Poster Presentations
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Session I:
New reactor and energy technology
ABSTRACT

The IRIS integral reactor is on the path towards licensing. The paper deals with the study, design and set up of the experimental facility for the performance of the Integral Effect Tests (IET). IRIS owns specific layout, i.e. the integral configuration, and enhanced safety features, obtained also by exploiting a coupled dynamic behaviour during the accident sequences among the primary vessel and its components, the steel spherical containment and the passive safety systems. These design choices, very positive for the safety point of view, however require that the scaled simulator of the reactor must be able to simulate the dynamic behaviour of all the systems, containment included. The PIRT (Phenomena Identification and Ranking Table) analysis and the H2TS (Hierarchical, Two-Tired Scaling Analysis) process were adopted, leading to a 1:1 scaling in height, 1:100 in volume and power and with the fluid in prototypical conditions. A brief description of the facility is reported and a preliminary Text Matrix is presented as well.
1. Introduction

With the resurgence of nuclear power there is an increasing need for a range of new reactor designs, including smaller units of several hundred MWe. The International Reactor Innovative and Secure (IRIS) offers an advanced, modular 335 MWe design. IRIS features an integral primary system configuration with all main components located within the reactor vessel. This configuration enables a simplified design with enhanced reliability and economics and supports its safety-by-design™ approach, which results in exceptional safety characteristics. IRIS is being developed by an international team, led by Westinghouse, incorporating 19 organizations from 10 countries, about half of them European. IRIS development started in 1999 and has reached the level of maturity indicating potential for being commercially offered by the mid of next decade. Economic analyses indicate that IRIS will be competitive with other nuclear and non-nuclear energy sources, whether deployed gradually in single units in smaller grids, or in multiple twin units for larger grids. Additionally, IRIS fits well the recently announced US DOE initiative, GNEP (Global Nuclear Energy Partnership) aiming to support worldwide expansion of the use of nuclear energy in a responsible and proliferation resistant manner. Within the GNEP framework, IRIS can in the near term offer an advanced reactor design to satisfy needs for smaller, grid-appropriate reactors.

On the R&D path towards licensing, currently the preliminary design has been completed and the testing needed for design certification has started. The centerpiece of the experimental program is the integral system performance testing to be performed at the SIET facility in Italy. The pre-application review process with the US NRC was initiated in 2002 to address long-lead items, and enable obtaining the Final Design Approval (FDA) by 2013.

The SPES-3 integral test facility of the IRIS reactor project is a key point in the development of the integral reactors concept and mainly for the IRIS licensing phase. Due to the reactor configuration, the scaling choices and the simulation constraints, SPES-3 is going to represent a unique facility in its kind. The large scaling factor and the need to simulate both the primary reactor vessel, the containment and their coupled dynamic behaviour, represent a significant challenge for the designers.

Fig.1 reports the general scheme of the IRIS integral layout, the safety systems and the functional links among the reactor pressure vessel, its internals, the safety systems and the containment. Since a positive coupling effect among the components is exploited in the safety strategy [1], all the relevant coupled phenomena have to be simulated in the facility. The scaling process and the resulting features of the SPES-3 facility are depicted in the following paragraphs.

2. Scaling approach and requirements for the SPES-3 facility

Scaling of the SPES-3 facility is a part of the Evaluation Model Development and Assessment Process (EMDAP) [2]. Based on the specified figures of merit, and identified and ranked phenomena and processes [3], which is EMDAP Element 1 (Establish Requirements for Evaluation Model Capability), the scaling analysis and identification of similarity criteria needs to be performed as a part of EMDAP Element 2 (Develop Assessment Base). The initial scaling analysis was based on Hierarchical, Two-Tired Scaling Analysis (H2TS) [4, 5]. The first two Stages of H2TS analysis (IRIS System Decomposition and Scale Identification) were performed [6] and followed with Sage 3 – Top-Down System Scaling Analysis and Stage 4 – Bottom-Up Process Scaling Analysis. These last two steps need to be performed iteratively and simultaneously with the design of the test facility. In the meantime the Fractional Scaling Analysis (FSA) [7, 8, 9, 10] was applied, as well. Both, H2TS and FSA use concepts from the hierarchical theory presented by [11], and the concept of time-scale modeling [12, 13]. However, introduction of the effective Fractional Rate of Change (FRC) in FSA provides the proper time constant for scaling a time-dependent evolution process in an aggregate (assembled of several interacting modules) and makes it more appropriate for scaling Integral Test Effects [8].
In the case of SPES-3 test facility the overall accepted volumetric scaling factor is 1:100 and the height scaling factor is 1:1. The fluid is water at prototypical pressure and temperature conditions.

The advantages of the accepted scaling approach are summarized in the following points.

Full Height of the test facility provides:
- Prototypical distance between heat sources and heat sinks centers to properly simulate natural convection effects.
- Both, single phase and two phase natural convection loops can be simulated simultaneously.
- Prototype and model fluid velocities and residence times in the loops can be adjusted to be the same.
- Horizontal inter-phase areas (transfer area concentrations) are properly scaled.

Prototypical Pressure and Temperature eliminates:
- Distortions due to the different fluid properties are not present (scaling analysis does not generate additional terms related to property distortions and interpretation of the results is easier).

However, the overall volumetric scaling factor 1:100 and full height bring some disadvantages:
- Resulting in 10 times larger Transfer Area Concentrations for heat transfer (energy exchange) and wall friction (momentum exchange) at vertical side walls.
- Some components (like heat exchangers) might be represented with limited number of tubes (not adequate to address side/bundle effects).

Transfer Area Concentration for Heat Transfer through the Side Walls might be adjusted passively (by applying an insulation material), or actively (by applying electrical heaters). The design of the test facility heat exchangers needs to address side/bundle effects (see next paragraph).

3. Main features of the SPES-3 facility

The SIET labs

The SIET company operates since 1983 as a Centre for Studies and Experiences with the primary purpose of carrying out safety tests on components and systems for nuclear power plants. Due to the “experimental
structure” of very high technology content, SIET is able to simulate the thermal-hydraulic loops of both existing nuclear power stations and new generation plants, at prototypical fluid thermodynamic conditions. In the past, SIET has qualified components and systems for customers like ENEA, ENEL, Ansaldo, General Electric, Mitsubishi, Doosan, Toshiba and Westinghouse. As for the Westinghouse AP-600, for which SIET provided the experimental results for the qualification and licensing process on the SPES-2 facility, in the frame of the IRIS program, it is going to build SPES-3 simulating IRIS in all its primary, secondary and containment components suitable to verify the effectiveness of interaction among them and qualify the system.

**The SPES-3 IRIS facility**

The SPES-3 is an integral test facility modelling the IRIS reactor, in particular:
- the primary circuit including the RPV with power channel and fuel box, lower riser and RCCA, upper riser, pressurizer, upper downcomer, steam generators, riser-to-downcomer connection check valves, lower downcomer, lower plenum, circulation pump;
- the secondary circuit up to the Main Isolation Valves, including Steam Generators (SG), Feedwater lines and Steam Lines;
- the safety system including the Emergency Boration Tanks (EBT), the Emergency Heat Removal System Heat Exchangers (EHRS_HE) located in the Refuelling Water Storage Tank (RWST) and the Automatic Depressurization System (ADS);
- the containment system including the Dry Well, the Quench Tank (QT), the Pressure Suppression System (PSS), the Reactor cavity and DVI room, the Long Term Gravity Make-up System (LGMS), and the DVI line.

Fig. 2 shows the resulting general layout of the SPES-3 facility.

**The primary circuit**

The RPV is a cylindrical tank of about 22 m height and 0.65 m diameter with the all main internal hydraulic paths reproduced. The fuel bundle consists of 235 rods powered by indirect heating plus 1 dummy rod, assembled in a standard Westinghouse 17x17 fuel assembly configuration. The maximum available power is 6.5 MW while the full scaled power should be 10 MW. This affects only the steady state and early phases of the transients and is considered acceptable from the system general behaviour investigation with a correct flowrate scaling in those phases. The pressurizer has an inverted hat shape and maintains the required pressure by means of a vertical electrical heater.

The SG zone consists of three annular sections suitable to locate the helical tube rows of the SGs: a single row in the inner and intermediate annulus, a double row in the outer annulus. An outer pump, injecting directly on each SG annular section, provides the required flowrates.

**The secondary circuit**

Eight IRIS SGs are simulated with 4 helical tube rows, each consisting of 14 tubes wrapped concentrically around the cylindrical riser. The global height of the SGs is 8.2 m with an average coil length of 32 m. In order to keep the same thermalhydraulic behavior for each steam generator and to ensure a prototypical fluid dynamic behavior on the primary side, different design solutions have been identified and compared by means of CFD analysis.

As an example, Fig. 3 reports the primary flow velocity field between two helical coil tube rows, simulated as a periodic bundle in the vertical direction, for two different solutions: the two bundles are bended in a parallel way or in a crossing way. The results show a better homogeneity for the crossing way, while the parallel way offers better solution for the manufacturing and the test section instrumentation.
Fig 2: Layout of the SPES-3 integral facility.

Fig 3: Velocity field of the primary flow between two helical coil tube rows (steam generator simulators): crossing (left side) and parallel (right side) rows layout.
(Red area: space between helical coil tubes; blue area: empty space in coil bundle helix; circles: holes in the tube bundle supports)
The safety systems

The two EBTs are cylindrical tanks connected to the RPV and DVI lines. The three EHRS.HEs consist of 3 and 5 vertical tubes, connected to cylindrical headers, suitable to simulate the heat exchangers belonging to a double or quadruple loop. The total height is about 3 m. They are located in the same RWST simulator of 12 m³ volume and 9 m height. The three IRIS ADS trains are simulated with a single train in SPES-3 by installing proper orifices to simulate the single or multiple intervention.

The containment

The IRIS containment compartments are simulated in SPES-3 by different tanks connected by pipe lines. Each tank shape is suitable to reproduce the same volume versus height trend of IRIS. As the pipe lines do not exist in IRIS, they are designed to limit their influence on the flow.

4. Preliminary Test Matrix

The preliminary Test Matrix of the design base cases is presented in Table 1. The test types, the test initiating events, purpose and additional comments are specified.

For the Low and High Elevations SBLOCA-s several additional group of tests are under consideration, like: demonstration of the long term cooling with all, or only two ADS trains available, tests beyond design cases with no, or only one EHRS.HE active, and tests with split breaks (instead double-ended guillotine breaks). ADS break is to demonstrate SBLOCA response with the maximum steam space (volume in the pressurizer) involved.

<table>
<thead>
<tr>
<th>Test Type</th>
<th>Test Initiating Event</th>
<th>Purpose</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>Low Elevation SBLOCA</td>
<td>Double-ended guillotine DVI line break</td>
<td>Establish $P_{CV}(t)$, $\Delta P_{CV,RV}(t)$, $P_{Cross}(t)$, LGMS injection initiation. Verify design basis case system response and mixture level</td>
<td>All safety systems OK except for a single failure on one ADS train</td>
</tr>
<tr>
<td>High Elevation SBLOCA</td>
<td>Double-ended guillotine RHR/CVCS line break</td>
<td>Establish $P_{CV}(t)$, $\Delta P_{CV,RV}(t)$, LGMS injection initiation. Verify design basis case system response and mixture level</td>
<td>All safety systems OK except for a single failure on one ADS train</td>
</tr>
<tr>
<td>ADS break</td>
<td>Double-ended guillotine ADS line break</td>
<td>Establish $P_{CV}(t)$, $\Delta P_{CV,RV}(t)$, LGMS injection initiation. Verify design basis case system response and mixture level</td>
<td>Maximum PZR steam space break</td>
</tr>
<tr>
<td>Feedwater Line Break</td>
<td>Double-ended guillotine feedwater line break inside containment</td>
<td>Show non-LOCA plant response with partial EHRS actuation (by design for all non-LOCA events).</td>
<td>To determine if the SG Makeup tank is needed.</td>
</tr>
<tr>
<td>Steam Line Break</td>
<td>Double-ended guillotine steam line break inside containment</td>
<td>Show non-LOCA plant response with partial EHRS actuation.</td>
<td></td>
</tr>
<tr>
<td>Safe shutdown sequence</td>
<td>Loss of all power</td>
<td>Demonstrate safe shutdown sequence</td>
<td>Observe primary coolant shrinkage, switch to primary coolant natural circulation, EHRS HX cool-down capability</td>
</tr>
</tbody>
</table>

Table 1: Preliminary Test Matrix.
The Feed Line and Steam Line breaks are to demonstrate non-LOCA plant response with partial EHRS actuation. Finally, the safe shutdown sequence is to demonstrate the plant response under the loss of all power.

5. Conclusions

The Integral Effects Testing of an integral layout PWR represents a significant challenge on the experimental side of the R&D effort. This is particularly true for the IRIS reactor, since its innovative layout integrates the dynamic behavior not only of the components into the integral reactor pressure vessel but also of the containment. The complexity of the item has been addressed by the IRIS R&D team and the strategy and final layout of the facility have been identified. A significant effort will be devoted also to set up the facility instrumentation, to obtain a valuable set of data for the development and assessment of the evaluation models. A coupled code able to simulate both the containment and the primary and secondary reactor systems will be developed and adopted.

Acknowledgements

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References

ECONOMIC FEATURES OF SMALLER SIZE, INTEGRAL REACTORS

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ABSTRACT

A misguided application of the economy of scale would label the small-medium size reactors as not economically competitive with larger plants because of their allegedly higher capital cost (€/kWe). The historical trend of capital costs vs. plant size is estimated from literature, and a reference exponent factor for the economy of scale law is obtained. Specific models are adopted for the estimation the various factors which, beside size, contribute in determining differentiating the capital cost of smaller reactors with respect to large reactors. The results show that when all the factors are accounted the capital costs of small and large plants installations are practically equivalent. Moreover, the historical increment in the O&M costs shows that the small size reactors are not dramatically affected. The IRIS reactor is used as the example of smaller reactors, but the analysis and conclusions are applicable to the whole spectrum of small nuclear plants.

1. Introduction

To fulfill the growing energy needs of developing countries and emerging markets, smaller size reactors are needed. This has been identified within the US DOE Global Nuclear Energy Partnership (GNEP) initiative as one of the key elements, “Grid-Appropriate Reactors”, needed to enable worldwide expansion of the peaceful use of nuclear power. Smaller size reactors (IAEA defines “small” those reactors with power <300 MWe and “medium” with <700 MWe) are the logical choice for smaller countries or those with a limited electrical grid. Small reactors have attractive characteristics of simplicity, enhanced safety and require limited financial resources. However, the other side of the coin is that they are not seen as economic because of the accepted axiom of the economy of scale: the specific capital cost ($/KWe) of a nuclear reactor decreases with the size increasing. Thus, in large, developed countries, the reactor size has steadily increased from a few hundred MWe 40 years ago to 1500 MWe and more today. But, the economy of scale applies only if the reactors are of a very similar design, as it has been the case in the past. This is no longer true today, where smaller modular reactors have very different designs and characteristics from the large ones. Thus, assuming by definition that, because of the economy of scale
principle, the capital cost of a smaller size reactor is higher than for a large size reactor is simplistic and wrong.
The awareness and realization of the economic potential of smaller reactors has grown significantly in the last few years (even though some work is as old as 15 years like the seminal paper\cite{1} by UKAEA, which has been the guide of the present work).
In addition to individual studies, the IAEA has launched in 2006 a collaborative project to address the competitiveness of Small-Medium Reactors (SMRs). As part of the IRIS (International Reactors Innovative and Secure)\cite{2} development, Westinghouse had already initiated investigation of the economic characteristics of IRIS. A more comprehensive outlook at the various components which make up the economics of SMRs was then undertaken by Westinghouse and some of its IRIS team partners, as a contribution to the IAEA study.
The general approach to smaller reactors economics and some preliminary results obtained by Westinghouse and the Polytechnic of Milan, Italy (POLIMI) are reported in this paper.

2. Cost factors affecting SMRs vis-à-vis Larger nuclear plants

When evaluating the competitiveness of SMRs versus large reactors, the various individual factors can be grouped into two classes:

- Factors which are either applicable to SMRs only or are critically affected by the difference in design and approach brought in by the SMRs (SMR specific factors)
- Factors which affect SMRs and large plants in a comparable way (common factors). Even for the common factors, a comparative quantitative evaluation might not be straightforward.

The SMR specific and the common factors are listed in Tables 1. The list is not exhaustive and others might be considered. Presented here are the ones judged to have higher priority for a quantitative evaluation; Six factors (identified by (*) in the Tables) have actually been addressed, as discussed in Section 4.

<table>
<thead>
<tr>
<th>SMR Specific Factors</th>
<th>Common Factors</th>
</tr>
</thead>
<tbody>
<tr>
<td>Design Related Characteristics (*)</td>
<td>Size (*)</td>
</tr>
<tr>
<td>Compactness</td>
<td>Modularization</td>
</tr>
<tr>
<td>Cogeneration</td>
<td>Factory Fabrication</td>
</tr>
<tr>
<td>Match of Supply to Demand (*)</td>
<td>Multiple Units at a Single Site (*)</td>
</tr>
<tr>
<td>Reduction in Planning Margin</td>
<td>Learning (*)</td>
</tr>
<tr>
<td>Grid Stability</td>
<td>Construction Time (*)</td>
</tr>
<tr>
<td>Economy of Replication</td>
<td>Required Front End Investment</td>
</tr>
<tr>
<td>Bulk Ordering</td>
<td>Progressive Construction/Operation of Multiple Modules</td>
</tr>
<tr>
<td>Serial Fabrication of Components</td>
<td></td>
</tr>
</tbody>
</table>

Tab 1 – List of Specific and Common factors for a differential evaluation

3. The life cycle cost breakdown for a nuclear power plant

The Life cycle cost for a nuclear power plant is conventionally subdivided in the following macro categories: Capital Cost, Fuel cost, Operation and maintenance cost and Final or decommissioning cost. Since different studies use different drivers to allocate the cost, in the literature there are different percentage for breakdown cost (Table 2). There are studies (SPRU, University of Sussex) and NERA\cite{3} that gives an indicative range representing the proportion for the accounts in the cost breakdown (Table 3). Therefore this paper is focused on two principal accounts: the Capital cost and Operation and Maintenance Cost.
4. Capital Cost

The ad hoc and common factors showed in table 1 do not by any means represent a complete list but they are the one judged as most representative. An initial quantification of some of these factors has been attempted. The SMR representative was the IRIS reactor, which is offered in single (335 MWe) or in twin (670 MWe) units. The large reactor used as reference was an hypothetical 1340 MWe PWR. The IRIS reactor was used because of the obvious familiarity and interest of the authors, but the evaluation conducted here is fully applicable to SMRs in general.

Six factors were evaluated: size; multiple units at a single site; learning; construction time; match of supply to demand; and, design related characteristics. The results are reported in Figure 1 and Table 4.

The first factor represents the economy of scale, assuming that the two plants are comparable in design and characteristics. The usual correlation

$$OCC_{SMR} = OCC_{LARGE} \times \left( \frac{size_{SMR}}{size_{LARGE}} \right)^{n-1}$$

was adopted with $n = 0.62$. All other things being equal, the overnight capital cost (OCC; $/KW) of the SMR would be 70% higher than the large pant. But all other things are not equal and other common factors will tend to reduce the SMR disadvantage.

The multiple units factor was evaluated considering that there are fixed, un-repeatable costs only incurred for the first unit and there are costs which are shared by the multiple units. The experience reported in the literature for Korean and French units on the same site was factored in our evaluation. For the four versus one plant comparison, it was evaluated that a 14% savings exists for the multiple SMRs.

The learning factor was evaluated from the various models reported in the literature. It was found that for the four units’ case the cost reduction is between 8% and 10%. The 8% value was conservatively chosen.

The next two effects, construction schedule and matching of supply to demand (or “timing”), were evaluated together, assuming a construction schedule for the large plant and SMRs of five and three years respectively, a discount rate of 5%/year and calculating the cumulative expenditures for the two cases. A 6% savings was estimated for the shorter construction time SMRs capability of better following the demand curve.

The principal design related characteristics for IRIS are: elimination of the pressurizer, steam generators vessels, canned pump housings, all large piping, vessel head and bottom penetrations and seals; elimination of safety systems such as the high pressure emergency core cooling system due to the safety-by-design approach which eliminates several postulated accidents; compact containment; lower amount of commodities. A conservative evaluation of these effects indicated a 17% cost savings.
When the various factors are combined, a single SMR of 335 MWe if deployed as part of a pack of four has a capital cost only 5% higher than the monolithic 1340 MWe reactor. Some sensitivity studies were also conducted, for example, to allow also the large plant to take advantage of multiple units on site and to investigate the effect of “worldwide” type learning. The reference case reported here and yielding a cumulative 1.05 factor considered four IRIS and one large plant on site, with no prior experience for either (i.e., no worldwide learning). A case of eight IRIS and two large plants on site, still with no prior experience yielded a total factor of 1.16, reflecting the proportionally higher effect of two large units on site. On the other hand, a case of four IRIS and one large plant on site, but with a prior worldwide experience of 2680 MWe for both (which means two large plants and eight IRIS) yielded a total factor of 1.0, reflecting the much larger learning deriving from the higher number of units. All the other sensitivity cases fell within the 1.0-1.16 range.

![Chart](chart.png)

**Table 4** Quantification of Factors Evaluated in SMRs/Large Plant Comparison of Capital Costs (Figure 1)

<table>
<thead>
<tr>
<th>Factor</th>
<th>Individual SMR/Large</th>
<th>Cumulative SMR/Large</th>
</tr>
</thead>
<tbody>
<tr>
<td>(1) Economy of scale</td>
<td>1.7</td>
<td>1.7</td>
</tr>
<tr>
<td>(2) Multiple-unit saving</td>
<td>0.86</td>
<td>1.46</td>
</tr>
<tr>
<td>(3) Learning</td>
<td>0.92</td>
<td>1.34</td>
</tr>
<tr>
<td>(4) (5) Construction schedule and timing</td>
<td>0.94</td>
<td>1.26</td>
</tr>
<tr>
<td>(6) Design specific</td>
<td>0.83</td>
<td>1.05</td>
</tr>
</tbody>
</table>

5. **Operation and maintenance cost**

After the Capital cost the most important account in the life cycle cost for a nuclear power plant is the “operation and maintenance cost”. Considering the table 3 seems that the operation and maintenance cost are a small part of the total cost for a nuclear power plant. However it is important to notice that it

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1 SMR: One 335 MWe plant, as part of four units; Large: One single 1340 MWe plant
becomes very important for an economic life cycle for a plant. In fact there are cases which nuclear power plants have been closed due to a dramatic increase in the operation and maintenance cost [9].

The model used to quantify the O&M cost is based on an ORNL Technical Report[10] and identifies three main cost categories: labour cost (on site and off site), material cost (shop supplies), and a third category including other minor cost items. The model assumes a reference cost for each cost category (estimated from DOE information), that is adjusted by means of two coefficients. The first one considers the economy of scale effect whereas the second takes into account the number of units built in the same site. The estimation functions for both coefficients use an exponent less than one, due to the nonlinear correlation between the number of units, the size of reactors and O&M costs.

According to the model is possible to conclude the a site with four SMR (335 MWe) has an O&M cost 24% greater than a site with one 1340 MWe Large reactor, or likewise a site with three SMR has an O&M cost 22% greater the a site with one 1005 MWe Large reactor.

It is also important to notice that the model doesn’t consider the specific advantages coming from the SMR technology. A correct quantification of them will be able to reduce the gap.

6. Conclusions

Smaller and larger reactors address different markets and there are many market related factors favouring one versus the other, independently from their capital cost.

When however they are competing on the same market the capital cost is not a discriminator and the two types of nuclear plants are practically equivalent under this respect.

The O&M costs increase more than the Capital Cost, but less than how is foreseen by a rough computation with the economy of scale canonical equation. They also represent a small part of the total cost which is composed mainly by the capital cost.

The so-called economy of scale is actually no longer an absolute advantage of larger reactors since it could be compensated by a variety of other factors.

This paper presents only the beginning of the evaluation of the competitiveness of SMRs and expanded, more detailed investigations will follow.

7. References

The reconstruction of the research reactor IRT-2000 in Sofia into a low-power reactor is being carried out from 2001. The future utilization of IRT aims to satisfy the society needs for: development and preservation of nuclear science, skills, and knowledge; implementation of applied methods and research; education of students and training of graduated physicists and engineers in the field of nuclear science and nuclear energy; development of boron neutron capture therapy. It will support the keeping up specialists with researcher’s approach and skills who are able to give adequate responses to the challenges of complex modern technologies and the associated environmental problems. It will be used for production of isotopes needed for medical therapy and diagnostics; in element activation analysis having a number of applications in industrial production, medicine, chemistry, criminology, etc. The reconstructed research reactor IRT will use low-enriched uranium fuel IRT-4M, with uranium-235 enrichment below 20%, is in accordance with the current norms on the security of transport and storage of nuclear and other radioactive materials which are vulnerable to theft by terrorists.

The research reactor IRT-2000 (IRT) in Sofia to the Institute for Nuclear Research and Nuclear Energy (INRNE) was built and put into operation in 1961. It was temporarily shut down in 1989 for improvement. The reconstruction of the IRT is being carried out under the decision of the Council of Ministers of Republic of Bulgaria from 2001. The strategy for sustainable utilization considers the IRT as a national base and aims to satisfy the society needs for:

- education of students and training of graduated physicists and engineers in the field of nuclear science and nuclear energy,
- implementation of applied methods and research,
- development and preservation of nuclear science, skills, and knowledge.

The IRT Technical Design is being in process of elaboration. The IRT will be reconstructed into a reactor:

- of thermal power 200 kW;
- with low enriched fuel, with uranium-235 enrichment below 20% in accordance with the current requirements of the security of transportation and storage of nuclear and other radioactive materials which are vulnerable to theft by terrorists;
- with ten vertical and seven horizontal experimental channels which will supply maximal fast neutron flux about $3 \times 10^{13} \text{n/cm}^2\text{s}$, and maximal thermal flux about $8 \times 10^{14} \text{n/cm}^2\text{s}$;
- with channel which will supply epithermal neutron flux about $0.9 \times 10^9 \text{n/cm}^2\text{s}$ suitable for medical Boron Neutron Capture Therapy (BNCT) application.

The MCNP 4C three-dimensional neutron transport code using the DLC-200 neutron cross-sections library has been applied for calculations of different variants of reactor core and BNCT filter. The core model used in MCNP 4C calculations is presented in Fig. 1.
The INRNE together with the Technical University in Sofia have proposed to the Ministry of Education a new programme for education of students in nuclear energy. The Nuclear Energy course will be obligatory for obtaining the Master of Science Degree of the Technical University in Sofia. The educational classes refer: types of research reactors, main characteristics and design of the reconstructed IRT, safety assuring and licensing, reactor physics and thermo-hydraulic characteristics determination, accident analyses, fresh and spent fuel management, radioactive waste management and governmental categorization norms and rules. Acquaintance with calculational codes as the MCNP code for neutron transport and criticality calculations, WIMS-ANL code – for preparing of neutron cross sections for diffusion calculation, REBUS code for the fuel burn depth calculation, SCALE code system for spent fuel transport and storage devices safety assessment, PLTEMP/ANL code for calculation of thermo-hydraulic steady-state, and RELAP5 code – for transient operation, etc. is planned too. Preliminary acquaintance with the neutron activation analysis and BNCT is included in the educational programme.

The INRNE and the reconstructed IRT will be used for carrying out specific training exercises on the reactor: reactor start, manual and automatically control, control rod calibration, delayed neutron group measurements, sub-critical multiplication/shutdown margin measurements, excess reactivity and shutdown margin measurements; reactor-physics measurements of static and kinetic reactor parameters, reactor dosimetry, measurements of the spent fuel characteristics in the hot laboratory, radiological characterization survey - alfa, beta and gamma measurement techniques, contamination measurement, etc.

The reconstruction of the IRT includes an arrangement for a BNCT facility. Preliminary neutron transport calculations for BNCT channel regarding the geometry and material composition design have been carried out (Fig. 2). Feasibility studies within the national network of the Medical University in Sofia, the National Centre of Radiobiology and Radiation Protection, the Institute of Experimental Pathology and Parasitology and Institute of Electronics of the Bulgarian Academy of Sciences, and the Faculty of Physics of Sofia University are carried out. Contacts with institutes, experienced in BNCT as EC JRC, Petten, the Netherlands, VTT, Finland and NRI-Rez, the Czech
Republic, were established. Human, social and economical results due to the BNCT for patients from Balkan region are expected.

Besides the financial support of the Bulgarian government the IRT has the IAEA support through the project BUL/4/014 “Refurbishment of the Research Reactor” and the support of the US Department of Energy in the frame of the RERTR program.

The reconstructed IRT is a basis for keeping up specialists with researcher’s approach and skills who are able to give adequate responses to the challenges of complex modern technologies and the associated environmental problems. The reactor will be used for production of isotopes needed for medical therapy and diagnostics; it will be the neutron source in element activation analysis having a number of applications in industrial production, medicine, chemistry, criminology, etc.

Nuclear energy has a strategic place within the structure of the country’s energy system. A new nuclear power plant Belene with two reactors of 1000 MeV will be built. The extremely high requirements regarding nuclear safety call for the availability of scientific and technical potential, and for an adequate culture of safe use of nuclear energy. The acquired scientific experience and qualification in reactor operation is a basis for participation of the country in the international cooperation within the European structures. In that aspect, the operation and use of the IRT brings economic benefits for the country.

Figure 2. The BNCT beam tube model:
1. Vessel of Channel;
2. Filter 3. Lead Shielding;
DEVELOPMENT OF AN ITER RELEVANT INSPECTION ROBOT

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ABSTRACT

Robotic operations are one of the major maintenance challenges for ITER and future fusion reactors. In particular, in vessel inspection operations without loss of conditioning will be very useful. In this context, an Articulated Inspection Arm (AIA) is currently being developed by CEA within the European work programme framework, which aims at demonstrating the feasibility of a multi-purpose in-vessel Remote Handling inspection system using a long reach, limited payload carrier (up to 10 kg). After qualification, the arm will constitute a promising tool for generic application. This paper deals with the integration of the robot into Tore Supra and presents the associated processes for inspection tasks.

1 Introduction

The aim of this R&D program is to demonstrate the feasibility of in-vessel Tokamak inspection tasks for the future fusion reactor ITER [1]. An Articulated Inspection Arm (AIA) is being developed to satisfy requirements in terms of maintenance and in-vessel component inspections. The mean specifications of the project are: large operational range, ultra high vacuum and temperature ambiance at 120°C. Operations under magnetic field and nuclear ambiance are not yet considered in the implemented technology for the AIA demonstrator. Preliminary tests of the AIA under relevant vacuum and temperature conditions are scheduled on the TORE SUPRA Tokamak at CEA/Cadarache by the end of 2007. The TORE SUPRA Tokamak is equipped with actively cooled components and operates with similar vacuum and temperature conditions as ITER (120°C to 200°C). Integration of the AIA demonstrator, associated with development of inspection processes will demonstrate in-vessel operating capabilities (viewing, leak detection on water loops, erosion or deposition characterization, abnormal events diagnostic…).

2 The Articulated Inspection Arm

The AIA is an 8 meter long multi link carrier with 5 modules of 160 mm diameter. The length of the AIA robot is consistent with that required for ITER. The modules include pitch (±40° in the vertical plane) and yaw (±90° in the horizontal plane) joints linked with a parallelogram structure that keep yaw joints axis always vertical [2, 3, 4]. Combination of elevation and rotation motions gives to the robot 8 degrees of freedom. The total weight of the AIA is about 150 kg. The robot is moved along its support with a linear trolley named Deployer. The payload carrier is limited to 10 kg.
All electronic systems are embedded in each AIA module. These components are enclosed at the atmosphere pressure in tight boxes while the mechanical structure is under vacuum. Moreover, components shall sustain a temperature of 200°C during the conditioning phase and 120°C for operations and in-vessel Tokamak deployments.

A successful vacuum and temperature test campaign on a prototype module was performed in 2005 in CEA/DRFC test facilities. In particular, a baking phase at 200°C was performed during a couple of days and the final spectrum has shown a good component conditioning. To overcome pollution issues with using grease, the design of free lubricant joints is based on thermal treatment with Teflon coating. An endurance testing was also performed at room conditions to qualify the 5 modules performances under representative loading (see Fig. 1).

These encouraging first results have pre-qualified the selected technologies for the AIA project.

![Fig. 1: The AIA robot 5 modules assembled with a 10 kg load at its end (on the right); Assembling and tests performed in CEA/LIST laboratories.](image)

### 3 Integration on TORE SUPRA Tokamak

A scale one demonstration of the AIA under ITER requirements is planned on TORE SUPRA which should lead to significant improvement in R&D results for in-vessel fusion remote handling equipments.

A long storage cask was designed for conditioning of the robot (vacuum and temperature) and for accurately guiding it during in-vessel Tokamak deployment. This stainless steel large structure (11m long, 3m height and 5 tonnes) is carried by 2 rolling wagons. One integration objectives is to connect or fold up the entire device in about 1 hour on a dedicated port of TORE SUPRA. For this purpose, the cask is equipped with a double valve that allows disconnection of the vessel without loss of the vacuum conditions. Moreover, all electro-technical equipments are embedded to realize a compact and an autonomous system (see Fig. 2).

A first deployment of the AIA robot into Tore Supra vacuum-vessel in planned by the end of 2007 (see Fig. 3). Robustness, reliability and flexibility will be tested and improved between successive plasma operating campaigns [5].
Development of interchangeable processes

The AIA is designed to allow accurate displacements of the head in front of the Plasma Facing Components. Several processes are in development to be implemented at the front end of the AIA robot. All these processes shall be interchangeable. This specificity will offer flexibility in the operation tasks.

The first developed process is a viewing system to make close visual inspection of Plasma Facing Components. The video process is designed with a CCD sensor embedded in a tight box made of stainless steel and glass (see Fig. 4). This box is linked to the head of the robot through a vertical joint actuated from inside with the same system as the yaw joint of the robot. All the electronic components inside this box are nitrogen-gas actively cooled by the means of a flexible umbilical. This system is
Currently tested and will be integrated for the first AIA deployment inside the TORE SUPRA Tokamak by the end of 2007.

Fig. 4: The visual inspection process.

A process based on helium sniffer is being considered to improve and facilitate maintenance operations on water loop leak tests.

Some analysis and operations on the Plasma Facing Components could be performed by laser systems [6]. Several characterizations and treatments are proposed for:

1. **The deposited layer depth** on the Plasma Facing Components can be measured using a repetitive laser pulse in a heating regime (0.1 to 0.4 J/cm², 100ns, 10 kHz, Nd-YAG laser).

2. **The removing of the deposited layer** is possible with the same laser device in the ablation regime (0.4 J/cm²). This technique can be considered for recovering the tritium trapped into ITER Plasma Facing Components.

3. **The composition of deposited layer** can be estimated via Laser Induced Breakdown Spectroscopy (LIBS). Integration of 2 optical fibres could be added into the AIA, both connected respectively to laser source and spectrometer located outside of the Tokamak.

5 Conclusion

Future deployment on TORE SUPRA of this multipurpose robotic device will give new perspectives on in-vessel maintenances and operating activities for fusion reactor like ITER. Several processes are foreseen to be associated on the AIA robot carrier to inspect, diagnose or treat Plasma Facing Components. Preliminary tests with dedicated vision device under relevant vacuum and temperature conditions will be performed in TORE SUPRA by the end of 2007. Other developments are under development for a further integration beyond 2007.

6 Acknowledgements

This work, supported by European Communities under the contract of Association between EURATOM/CEA, was carried out within the framework of the European Fusion Development Agreement. This views and opinions expressed herein do not necessarily reflect those of the European Commission.
The authors would like to acknowledge the technical staff of CEA/DRFC/STEP/GARV for its assistance.

7 References

CONCEPT OF A FUTURE
HIGH PRESSURE - BOILING WATER REACTOR, HP-BWR

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ABSTRACT

Some four hundred Boiling Water Reactors (BWR) and Pressurized Water Reactors (PWR) have been in operation for several decades. The presented concept, the High Pressure Boiling Water Reactor (HP-BWR) make use of the operating experiences. The best parts of the two reactor types are used and the troublesome components are left out. This means improved safety. The increased thermal efficiency is beneficial to the environment as less warm cooling water is released per produced kWh. With some modifications the presently used components can be used making this design cost effective and possible to realize in a not too distant future.

1. Introduction
Since the 1950s several hundreds Boiling Water and Pressurized Water Reactors (BWRs and PWRs) are in use. There is a wealth of operating experiences. During the years many difficulties occurred with a number of important components. This concept, the High Pressure – Boiling Water Reactor (HP-BWR) offers a solution to use the best parts from each type (BWR and PWR) and leave out the troublesome components. This means an important increase of safety. As an extra benefit, also a higher efficiency is attained beneficial for the environment as less warm cooling water is released per produced kWh. The HP-BWR is using –with some modifications- presently manufactured parts making this a cost effective, realistic concept.

2. The High Pressure – Boiling Water Reactor HP-BWR
The High Pressure Boiling Water Reactor (HP-BWR) is a promise for improved nuclear safety and increased leniency to the environment. The HP-BWR is an environment friendly cost effective alternative.
The HP-BWR is using a modified PWR reactor vessel and BWR type fuel and control rods. However here the cross formed control rods are gravity operated with ample space between the crosses and the fuel boxes. The control roads are manoeuvred electromagnetically which means that they will drop into the core at a loss of electrical power as in the PWRs. The traditional PWR control rods are finger shaped and are surrounded by a tube with a minimum of clearance. The traditional BWR control rods are operated from below with hydraulic pressure. Therefore in the bottom of the traditional BWR reactor vessel there are a great number of penetrations for the control rods. Directly below the reactor vessel there is an elaborate system of numerous high pressure hydraulic pipes to actuate the control rods. Taking the best and leaving out the difficulties of both the traditional BWR and PWR systems is a substantial safety improvement.

All the pipe connections to the reactor vessel are well above the reactor core. This allows the omission of the core spray. The moisture separators and steam dryers are outside the reactor vessel, leaving free space for the control rods.

Internal circulation pumps. These allow using orifices at the inlet of the fuel boxes so that the one phase pressure drop will dominate over the two phase pressure drop. This reduces the risk for hydrodynamic oscillations. However if suitable methods are found to facilitate natural circulation even the circulation pumps can be left out.

The use of the HP-BWR means improved Carnot cycle thermal efficiency up to about ~40% instead of about ~30%. The reason is that the HP-BWR steam temperature corresponds to 15MPa while the traditional BWR’s steam temperature corresponds to 7MPa and the traditional PWR’s steam temperature corresponds to 6MPa. The HP-BWR is lenient to the environment as less warm cooling water is released per produced kWh to the recipient, sea or river or to the air via a cooling tower.

Using direct cycle the system is simplified. Still, the usual PWR steam lines can be used through the containment wall to the turbine. A great advantage is that the complicated and costly steam generators are left out.

The moisture separators and the steam dryers are outside the reactor vessel in the containment instead of the huge troublesome steam generators.

Simple dry containment is used instead of the complicated, inert, pressure suppression wet containment which requires a great deal of surveillance.

3. The Traditional Boiling Water Reactor, BWR
The basic principles of the traditional Boiling Water reactor are well known

As there are pipe connections to the reactor vessel below the reactor core, a pipe break can empty the vessel leaving the core uncovered, without the cooling water. Therefore a core spray is required. This is a common feature for the BWRs with external circulation pumps or jet pumps. However this drawback is eliminated at the later design at the Advanced Boiling Water Reactor, ABWR. All BWR
control rods are inserted to the core with hydraulic power, some with electric motors too. This makes the lower part of the reactor both inside and outside the bottom of the reactor vessel extremely elaborate. To make things worse, in the past, cracks, corrosion and leakage have occurred at the penetrations at the lower part of the reactor vessel.

The huge reactor vessel would require an enormous dry containment building; therefore, instead a pressure suppression containment system is used. The containment is separated into two parts, the upper drywell and the lower wet well with the suppression pool. If the separation is not near perfectly leak tight the wet well cannot fulfill its function to suppress the pressure in the drywell in case of a pipe break. Further complication is that the traditional BWR containment operates inert, making the entrance into it more difficult.

The nice thing about the BWR is that it operates in direct cycle mode without the troublesome steam generators.

4. The Traditional Pressurized Water Reactor, PWR
Most of the World’s operating reactors are traditional PWRs.

The control rods are operated from above. Undoubtedly some leakages were observed at the penetrations which in a few cases led to the need to replace the reactor pressure vessel head.
The simple electromagnetic devices which manoeuvre the rods have worked reliably. This assures a high degree of safety. A basically continues, uninterrupted bottom of the reactor vessel avoids causing suspicions about its integrity.
The curse of the traditional PWRs is their steam generators. These complicated and costly huge pieces of equipment are disappointingly short-lived because of the corrosion of the internal tubes causing leakages. Many steam generators have been changed after some 15 years. This is an extremely expensive and troublesome and also time consuming operation.

In the upper part of the steam generators there is the moisture separator and the steam dryer. The HP-BWR is “borrowing” this equipment which can be used without the troublesome steam generators.

5. References

All university text books written for Nuclear Engineering students contain detailed descriptions of both Boiling Water Reactors and Pressurized Water Reactors. Also manufacturers in Europe, Asia and America publish data about their particular designs. There is a wealth of information about BWRs and PWRs on the internet.
Poster Presentations

Session II:
The nuclear Fuel cycle / Nuclear Operations
A Study on the Determination of Disposal Priority for Low and Intermediate Level Radioactive Wastes (LILW) in Korea

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1. Introduction

In Korea, the major LLW generator, called KHNP (Korea Hydro and Nuclear Power Co.) has generated about 67,000 drums (200L) of low and intermediate level radioactive waste (LILW) stored in the temporary storage facilities at each reactor sites since 1977. Especially, the amount of DAW (dry active waste) has been accumulated around 36,600 drums and this number represents about 56% of total LILW generated by all NPP in Korea. The amount of evaporated bottoms, concentrated wastes, spent ion exchange resins, and spent filters have been accumulated around 19,000 drums, 9,700 drums, and 1,600 drums and these numbers represent about 28%, 14%, and 2% in respectively.

![Figure 1. The amount of all wastes generated in Korea (As of Dec. 31, 2005)]
These LILW drums have been stored in temporary storage facility of each NPP site and have been prepared to be disposed in the Final repository which will be operated in 2009.

2. General Characteristics of LLW in Korea

2.1. DAW

DAW has included a variety of materials such as plastic, rubber, paper, metal and so on. The radioactivity level of DAW is turned out to be relatively low compared to other waste streams. However, some of DAW may have possibility to contain free liquids and harmful materials, including some wastes with high radioactive level. The detailed characterization of DAW is important and required to first step prior to the disposal of the waste. The free liquids in DAW can be easily detected by NDA (non destructive analysis) method using the X-ray, neutron, microwave, etc. However, the harmful materials in DAW can not be readily checked whether those are in drum or not [1].

2.2. Evaporated Bottom (Concentrated Waste)

In general, the evaporated bottoms and concentrated wastes are considered to be homogeneous waste that the activity of the waste can be easily estimated. Activity level of evaporated bottoms and concentrated wastes is also relatively low compared to that of spent ion exchange resins and filters. Activity estimation of all evaporated bottoms and concentrated wastes by DTC (dose to curie) method shows that no drums exceed the value of the disposal limits, which are regulated by Korea regulation law. Some of evaporated bottoms and concentrated wastes was solidified by paraffin wax and be relatively homogeneous. However, solidified wastes with paraffin can not be readily disposed because characterization methodology for those wastes has not been known yet in Korea.

2.3. Spent Ion Exchange Resin and Filter

For spent ion exchange resins and filters, although the radioactivity level is relatively high compared to other waste streams, the wastes from spent ion exchange resins and filters are showing good homogeneity same as that of evaporated bottoms and concentrated wastes. Therefore spent ion resins and filters could be considered to have early disposal priority because of relatively easy and accurate determination of the contents of the waste.
Table 1. The number of all waste drums generated per year (converted as the drum of 200L)

<table>
<thead>
<tr>
<th>Year</th>
<th>DAW</th>
<th>Evaporated Bottom</th>
<th>Spent Ion Exchange Resin</th>
<th>Spent Filter</th>
<th>The number of total drums generated per year</th>
</tr>
</thead>
<tbody>
<tr>
<td>1977</td>
<td>23</td>
<td>0</td>
<td>0</td>
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<td>23</td>
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<td>1978</td>
<td>95</td>
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<tr>
<td>1979</td>
<td>154</td>
<td>308</td>
<td>49</td>
<td>30</td>
<td>541</td>
</tr>
<tr>
<td>1980</td>
<td>137</td>
<td>471</td>
<td>22</td>
<td>33</td>
<td>663</td>
</tr>
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<td>239</td>
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<tr>
<td>1983</td>
<td>342</td>
<td>417</td>
<td>77</td>
<td>29</td>
<td>865</td>
</tr>
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<td>486</td>
<td>788</td>
<td>105</td>
<td>44</td>
<td>1423</td>
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<td>1985</td>
<td>385</td>
<td>628</td>
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<td>9</td>
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<td>691</td>
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<td>190</td>
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<td>714</td>
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<td>1293</td>
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<td>538</td>
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<td>349</td>
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<td>58</td>
<td>2200</td>
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<tr>
<td>1997</td>
<td>1801</td>
<td>258</td>
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<td>53</td>
<td>2534</td>
</tr>
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<td>2001</td>
<td>1952</td>
<td>251</td>
<td>449</td>
<td>57</td>
<td>2708</td>
</tr>
<tr>
<td>2002</td>
<td>2240</td>
<td>235</td>
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<td>263</td>
<td>418</td>
<td>60</td>
<td>3097</td>
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<td>2004</td>
<td>2808</td>
<td>306</td>
<td>337</td>
<td>32</td>
<td>3483</td>
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<tr>
<td>2005</td>
<td>3676</td>
<td>191</td>
<td>405</td>
<td>24</td>
<td>4296</td>
</tr>
</tbody>
</table>

| The number of total drums generated per stream | 36610 | 19040 | 9650 | 1588 | 66888 |

3. Considerations for Determination of Disposal Priority

To determine the disposal priority of LILW in Korea, the two main parameters called the activity and homogeneity were only considered in this study. In order words, the concept of disposal safety was the major consideration for the determination of disposal priority.

3.1. Activity

In this study, the DTC method was used to estimate the activity of all LILWs in Korea. The activity of γ-radionuclide derived by DTC method has some difference compared to that of
γ-radionuclide derived by real detection because the DTC method has conservative characteristics in itself. Although DTC method has some limitation for activity evaluation of LILWs, it is very helpful that the result of activity estimation is used for understanding the activity tendency of LILW in Korea. Especially, this result represents that no evaporated bottoms and concentrated wastes exceed the disposal limits. The number of drums exceeding the disposal limits is shown orderly in DAW, Spent Filter, and Spent ion exchange resin. Although the number of DAW exceeding the disposal limits is more numerous than other waste streams, the ratio of Spent Filter exceeding the disposal limits is more lager than that of DAW.

<table>
<thead>
<tr>
<th></th>
<th>The number of total drums</th>
<th>The number of drums exceeding the disposal limits</th>
<th>The ratio of drums exceeding the disposal limits</th>
</tr>
</thead>
<tbody>
<tr>
<td>DAW</td>
<td>18,394</td>
<td>486</td>
<td>2.64 %</td>
</tr>
<tr>
<td>Evaporated Bottom</td>
<td>16,599</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Spent Ion Exchange Resin</td>
<td>6,601</td>
<td>181</td>
<td>2.74 %</td>
</tr>
<tr>
<td>Spent Filter</td>
<td>1,394</td>
<td>236</td>
<td>16.93 %</td>
</tr>
</tbody>
</table>

**Figure 2. The ratio of drums exceeding the disposal limits**

### 3.2. Homogeneity

The homogeneity of the radioactive waste may become one of main considerations for the determination of the priority that what kind of waste should be disposed first in order to fill in
the repository site orderly [2]. In order to allow the radioactive wastes to be safely disposed in the repository, first of all, inner characteristics of each drum must be identified. The PCP (Process control program) is one of the methodologies to verify the homogeneity of some wastes, which are being produced in present. The NDA (non-destructive analysis) method is another methodology for checking the homogeneity of old wastes, which were already produced.

4. Conclusion

Since 1977, the KHNP has generated around 67,000 drums of low and intermediate level radioactive waste (LILW) converted as 200L drum. To dispose these drums, each drum was only estimated in the activity and homogeneity point of view in this study. After considering the two parameters to establish the disposal priority of LILW in Korea, the following order can be proposed:

(1) concentrate wastes solidified by the cement and DAW whose radioactivity level is very low and detailed characterization of the waste is identified easily because of the homogeneity, (2) spent ion exchange resin solidified by the cement, (3) spent filter, (4) DAW whose radioactivity level is relatively high but the characterization is well documented, (5) the wastes that require the additional researches including spent ion exchange resin in HIC, evaporated bottom and concentrated waste solidified by the paraffin, and DAW contained with some possible harmful materials.

<table>
<thead>
<tr>
<th>DISPOSAL PRIORITY</th>
<th>WASTE STREAMS</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>- Concentrate wastes solidified by the cement</td>
</tr>
<tr>
<td></td>
<td>- DAW whose radioactivity level is very low and detailed characterization of the waste</td>
</tr>
<tr>
<td>2</td>
<td>- Spent Ion Exchange Resin solidified by the cement</td>
</tr>
<tr>
<td>3</td>
<td>- Spent Filter</td>
</tr>
<tr>
<td>4</td>
<td>- DAW whose radioactivity level is relatively high but the characterization is well documented</td>
</tr>
<tr>
<td>5</td>
<td>- the wastes that require the additional researches spent ion exchange resin in HIC evaporated bottom and concentrated waste solidified by the paraffin DAW contained with some possible harmful materials</td>
</tr>
</tbody>
</table>

Acknowledgement
This study was financially supported by the KHNP (Korea Hydro and Nuclear Power Co.)

References

Abstract

The review of the water regime used for the Units of Paks Nuclear Power Plant was carried out in 2005, after 18 – 23 years of operation. It was clearly concluded after processing the huge database of the water regime, that there is nothing to prevent the power upgrading and service life extension of the Units.

In 2006, a new water regime was developed that will be applied during the preparation for the service life extension and the extended service life. In connection with this work, recommendations were made for some modifications in the previously used water regime.

Currently there is no uniform startup and shutdown water regime for WWER-440 Units. Therefore, special attention was paid to the development of a Unit startup water regime, which will be applied for the outages as early as the year 2007.

The summarised recommendations for water regime modification will be subject to international expert review in 2007, and the modifications judged to be implemented will be finalised after the review.

1. The purpose and the function of the primary and secondary water regime

   The primary and secondary water regime has several purposes and functions that can be summarised in the following list:
   - To provide for compensation for the reactivity margin by continuous reduction of the boric acid concentration and for the control/controllability of the reactor power (with follower assemblies).
   - To ensure that the overall corrosion of the structural materials of the equipment is minimum.
   - To minimise the risk of local corrosion of the structural materials.
   - To minimise the deposit of corrosion products on the structural components and the fuel clad.
   - To keep the rate of transport of corrosion products in the coolant and their deposit on the surfaces at low a level.
• To confine the rate of radiolytic decomposition in the primary coolant.

2. Analysis of the water regime currently applied for the Paks Units

2.1. The work to be done

Three fundamental issues were addressed by the complex and topical analysis:
• Is there anything to prevent the service life extension of the WWER-440 Units of Paks Nuclear Power Plant from water regime point of view?
• To what extent the primary and secondary water regime of Paks Nuclear Power Plant complied with the design requirements and the knowledge level deriving from the recent technological developments?
• How was the corrosion degradation of the major primary and secondary equipment and to what extent was the major primary and secondary equipment degraded by the applied water regime?

2.2. Conclusions for the primary circuit

It was possible to keep the impurity concentration of the primary coolant as low as achievable from process system point of view.

The periodic increase in the quantity of the disperse corrosion products can be attributed to the Steam Generator decontamination work performed at Unit 1, 2, and 3.

Altogether two cases of fuel leakage were detected for the 79 campaigns involved. The total iodine concentration was greater than 3.7 MBq/dm³, but never exceeded the Unit shutdown criteria of 37 MBq/dm³.

The average corrosion loss for the items of equipment of a material grade of 08H10N10T was 0.3-0.35 μm/year, i.e. 15-17.5 μm during 50 years, which does not prevent the service life extension.

2.3. Conclusions for the secondary circuit

The erosion rate of carbon steel surfaces being in contact with wet steam was high with the original water regime and therefore, significant amount of deposits settled down in the Steam Generators. This deposit had to be removed by chemical treatment method from the secondary side surfaces, 2 times for each Steam Generator.

Due to the insufficient tightness of the initially used condensers and because of the copper alloy structural material, significant amount of corrosion activator (chloride, sulphate, copper) was carried into the Steam Generators. The contaminants accumulated in the structural spaces of the Steam Generators are potential source of hazard. The shape of the gaps between the SG tube support spacers and the tubes are presented in Figure 1. The chemical cleaning work performed before were not even completely effective in these gaps.

![Figure 1: Structural gaps in the Steam Generators](image)

Various number of heat exchange tubes had to be plugged in each Steam Generator. This number depends on several factors, e.g. on the number and extent of leakages that occurred before the replacement of the turbine condensers of the given Steam Generator, the deviation of the material composition of the Steam Generator tubes (nickel content), etc. The number of tubes plugged as a result of the Eddy Current testing is presented in Figure 2. In this figure, the first digit of the figures on
the horizontal axis represents the number of the Unit, while the second one is the number of the Steam Generator. The largest leakage of primary coolant into the secondary side, during the use of the initial condensers, was found for the Unit 2 and 3.

The main condensate system, that became absolutely tight as a result of the condenser replacement, significantly reduced the amount of corrosion activators carried into the Steam Generators. Though, the efficiency of the blow-down of ionic contaminants from the Steam Generators had not changed, the decrease of the ion concentration of the feedwater resulted in the reduction of concentrations (chloride less than 10 µg/dm³) by an order of magnitude in the Steam Generators.

Due to the partial replacement of the copper containing structural elements, some copper content can still be detected in the feedwater and the Steam Generator.

The adoption of high pH water regime resulted in a reduction, nearly by an order of magnitude, of the amount of corrosion products transported into the Steam Generators that will be further reduced by the replacement of the High Pressure Pre-heaters. The replacement has taken place for Units 3 and 4 to date.

The installation of the Steam Generator feedwater headers and the use of the high pH water regime had a favourable effect on the removal of the corrosion products. The corrosion products transported into the Steam Generators in a significantly smaller amount do no longer deposit on the surface of the heat exchange tubes but as sludge on the bottom of the Steam Generator, thus allowing the effective removal by blowing-down.

In the following, we present a summary of the corrosion loss of the items of the secondary equipment during the period until the end of the 50th year of operation. $s_0$ represents the initial tube wall thickness, while $s_{50}$ is the wall thickness calculated for the end of the 50th year of operation:

- Condenser tubes: $s_0=0.6$ mm $s_{50}= 0.38$ mm
- Low pressure pre-heater tubes: $s_0=1.0$ mm $s_{50}= 0.95$ mm
- High pressure pre-heater tubes: (new pre-heaters of Unit 3 and 4): $s_0=1.4$ mm $s_{50}= 1.26$ mm
- Superheater, stage 1 and 2: $s_0=2.0$ mm $s_{50}= 0.86$ mm
- Steam Generator tubes: $s_0=1.4$ mm $s_{50}= 1.36$ mm

### 3. Recommendation for the implementation of the service life extension

#### 3.1. Recommendations for the primary water regime

The sampling tap (TV20) before the Water Purifier 1 does not provide representative sample for the disperse corrosion products under startup and shutdown transitional conditions. Therefore, the installation of a new sampling tap is necessary. On the basis of the inspection, acceptable samples can be taken from the sampling system of the operational boron acid measurement, which receives sample
directly from the reactor vessel.

For the on-line measurement of the most important chemical parameters it is necessary to improve the reliability and to ensure the full scope use of the primary circuit analytical monitoring system (FAM). The validation of FAM commenced and the results will be used to identify the measuring systems that can be used longer and those that will need to be replaced.

The development of a water regime for transitional (startup and shutdown) conditions is a task with highlighted importance for the service life extension. The startup and shutdown processes significantly affect and decisively identify the water regime parameters of the Unit for the entire campaign therefore, it is necessary to make the filtration of the primary coolant general at low (40 – 50 °C) temperature in these operation modes, in addition to the more frequent checking of some major chemical parameters. The improvement of the practice of feeding the chemicals (hydrazine, ammonia-hydroxide, potassium hydroxide) to the coolant will also be initiated.

Physical and/or chemical procedures will need to be developed for the removal of the corrosion products that accumulated in the primary circuit, which will be applied as required.

The reduction of the hydrogen concentration of the primary coolant from the range of 30-60 Nml/dm³ to 25-50 Nml/dm³ to reduce the pH fluctuation range.

It is recommended that the simultaneous addition of ammonia and hydrazine, rather than the use of hydrazine water regime is considered for one or two Units.

The reduction of the equivalent boron acid – potassium control range has a positive effect on the migration of corrosion products, being unavoidably present in the primary loop, and on their removal from the primary coolant. The difference between the presently used so called co-ordinated water regime and the proposed overall boron acid alkalinization control is presented in the Figures 3, 4, 5 and 6. The reduction of the control range of the potassium equivalent (overall alkalinization = potassium+lithium+sodium) and its break-down between the boron acid concentrations of 3 and 3.5 g/kg and again below 1 g/kg is well marked in Figure 5. The result of these changes can be studied by a comparison of the Figures 4 and 6. It is visible in Figure 4, that the difference between the corrosion product concentrations (C_R) calculated for the reactor vessel and those for the Steam Generators (C_SG) has a negative value for a significant portion of the campaign. Consequently, the precipitation of the corrosion products will take place mostly in the active core, and will turn to the Steam Generators at the end of the campaign only. This imposes unnecessary load to the active core and has an increasing effect on the radiation doses during maintenance outage.

It can be achieved by the implementation of the proposed modification as presented in Figure 6 that the precipitation of the corrosion products takes place in the Steam Generators from the beginning of the campaign and it turns to the active core at the end of the campaign only. With this solution it is expected that the active core can be maintained in rather clean condition and the portion of the corrosion products that precipitated in the active core can be removed all years together with the fuel assemblies unloaded during refuelling outage. The reorientation of the transport before maintenance outage will have a favourable effect on the radiation doses received during maintenance outage.

To achieve the above outlined aim, the overall alkalization concentration should be kept, from the beginning up to the time of achieving a concentration of 3.5 g/kg, in the upper part of the control range presented in Figure 5. After this point, it should be changed to the lower part of the range and below 0.5 g/kg boron acid concentration further reduction of alkalization is required. The change over of precipitation from the Steam Generators to the active core takes place during this period of some 500 hours.
Further tasks for the secondary circuit

The small amount of deposit in the structural gaps of the Steam Generators increases the risk of corrosion of the austenitic steel tubes therefore, the elaboration of an effective cleaning procedure, optimised for this environment is necessary.

The full replacement of the copper containing structural materials within the secondary system should be performed.

The modification of the anion exchanger of the Water Purifier 5 to mixed bed ion exchanger will allow a more efficient purification of the water blown down from the Steam Generators, and thanks to the cleaner water returned to the Steam Generators, the water quality of the Steam Generators can be further improved. On line conductivity meters should be provided for testing the quality of the purified water to ensure that the exhaustion of the ion exchangers is detected in due time.

The so called additional condensate accumulating at various places in the secondary system is collected in the condensate tank. To allow the location and elimination of the impurity sources as soon as possible, it is necessary to install an on-line conductivity measurement system for testing the water of this tank downstream of the cation-exchange column.

For the on-line measurement of the most important chemical parameters it is necessary to improve the reliability and to ensure the full scope use of the secondary circuit analytical monitoring system (FAM). The validation of FAM commenced, and the results will be used to identify the measuring systems that can be used further and those that will need to be replaced.

The blow-down efficiency of the ionic contaminants and disperse corrosion products from the Steam Generators will be reviewed by modelling and by performing dedicated measuring programs, and recommendations will be made for the improvements of the efficiency, as required.
The water regime risk factors of the Steam Generators are summarised in Table 1 below:

<table>
<thead>
<tr>
<th>Risk factors</th>
<th>Expectation</th>
<th>Actual values for Units 1 to 4</th>
<th>Required actions</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>The condition of the heat transfer surfaces</strong></td>
<td>Free of deposits (&lt; 50 μm)</td>
<td>minimum deposited</td>
<td>More efficient blow-down of the Steam Generators for disperse iron corrosion products</td>
</tr>
<tr>
<td>Concentration of the disperse iron corrosion products transferred into the Steam Generators [μg/dm³]</td>
<td>&lt; 3 – 5</td>
<td>&lt; 10 (3 – 8)</td>
<td></td>
</tr>
<tr>
<td><strong>Concentration of the stress corrosion activators in the Steam Generator water</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Maximum chloride-ion concentration during hide-out re-dissolving [μg/dm³]</td>
<td>&lt; 0.1</td>
<td>&lt; 0.1</td>
<td></td>
</tr>
<tr>
<td>Sulphate-ion concentration [μg/dm³]</td>
<td>&lt; 20 (5 – 10)</td>
<td>10 – 15</td>
<td></td>
</tr>
<tr>
<td>Maximum sulphate-ion concentration during hide-out re-dissolving [μg/dm³]</td>
<td>&lt; 0.15</td>
<td>&lt; 0.15</td>
<td></td>
</tr>
<tr>
<td><strong>Exclusion of oxidising materials from the water of the Steam Generators</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Concentration of the disperse copper corrosion products in the feedwater [μg/dm³]</td>
<td>~ 0</td>
<td>&lt; 0.5</td>
<td>Complete removal of the copper from the secondary circuit. Removal of the copper from the structural gaps.</td>
</tr>
<tr>
<td>Oxygen concentration of the main condensate [μg/dm³]</td>
<td>&lt; 5 – 10</td>
<td>&lt; 5</td>
<td></td>
</tr>
</tbody>
</table>

Table 1: Summary of the water regime risk factors in the Steam Generators

**Literature**

ABSTRACT

The European Community, in the EURATOM 6th Framework Programme, supports numerous R&D projects aiming to a possible development of Minor Actinides transmuters. This study aims to provide a critical approach in the context of P&T strategy, necessary to treat radiotoxicity data from different sources. The work, performed in SCK-CEN (Belgium) and Pisa University (DIMNP), concerns the state of the art revision of internal dosimetry in ICRP Publications, 10 CFR Part.20, 96/29/EURATOM and 2001 Belgian rules on Radiological Protection. In this regard, preliminary evaluation of an Industrial ADS (400MWth, 2.5mA) burning capability, using inert matrix fuel was performed. The main result obtained is an accurate assessment of ORIGEN2.2 last version, showing a difference of more than one order of magnitude in radiotoxicity values, if calculated using the most up to date coefficients from the latest ICRP Publications in comparison to the ORIGEN original ones.

1. Introduction

The panorama of studies about Minor Actinides (MA) transmutation, using an Accelerator Driven System, as a possible complementary approach to Spent Nuclear Fuel (SNF) geological disposal, has uncertainties in radiotoxicity definitions and values that, nowadays, are not more acceptable. This work, joined to nuclear fuel cycle closure and in particular to Partitioning and Transmutation (P&T) strategy, aims to give a critical approach and to manage radiotoxicity definitions. In fact, data from different sources imply often difficulties in comparing values. Radiotoxicity coefficients evolution review across ICRP Recommendations, BEIR and UNSCEAR Publications was analysed in depth. Attention was focused on their influence on spent nuclear fuel potential radiotoxicity and on the US and European Countries rules in radiological protection[1]. The background is the present Belgian situation (7 PWR, 5800 MWe installed), where, despite decision constrained by guarantee of energy independence and by engagement to respect the Kyoto agreement, Belgian Government decided (2003) to phase-out nuclear energy production in next the 25 years. Nevertheless the phase-out decision can be re-opened if certain conditions are not met (guarantee of energy independence, engagement to respect the Kyoto agreement). In this latter case, we proposed an alternative scenario: LWRs substitution, consequent installation of advanced reactors (i.e. EPR-European Pressurized Reactor) in synergy with industrial scale ADS to support energy demand and to burn consistent amount of HLW, in connection to Generation IV reactors (fast spectrum)[2].

The starting point of evaluating SNF radiotoxicity evolution is the waste inventory calculation of a reference reactor: a 1000 MWe PWR was chosen, with 4.3% enriched and 50GWd/tonIHM discharge burn-up fuel, as Belgian panorama typical one (Doel 3).
To perform these calculations ALEPH-1.1.2[3], a burn-up code under development at SCK•CEN (Belgium) inside the MYRRHA Project[4], that couples MCNPX, ORIGEN2.2 and NJOY99.112 code, was used.
ORIGEN2.2 was used separately from ALEPH to trace radiotoxicity curves. Apposite radiotoxicity coefficients data-bases were created, suitable for ORIGEN2.2 and for ALEPH-1.1.2 that gives as output isotopic materials quantities (g/cm³) only.

2. Health hazard measure from Radioactive High Level Waste

Correct assessment of exposure risk for members of public shall be evaluated on the basis of geological disposal that will receive total or part of nuclear material from reactors, if a solution for nuclear waste management would be found. In this regard, Radiotoxicity concept is representative of the measure of health hazard from nuclear radioactive waste that is considered a biological hazard potential.

In literature different radiotoxicity definitions exist, such as:
- the "quantity" of water required to dilute the material to the "maximum permissible concentration for human consumption" (MPC), that is referred then to its own country annual law limit;
- the relative measure to reference level, i.e. natural uranium radiotoxicity or an other Reference Level (RL); unfortunately the definition of RL is ambiguous as it will be well shown further on;
- the related to dose value, i.e. ‘dose per unit of intake’ (DPUI) in Sv/Bq, or to the number of cancer deaths expected if a given radioisotope is swallowed by a person (CD/Bq). It is not an absolute measure of the biological hazard of a given amount of radioactive material, but it is very useful to construct a relative measure of the biological hazard potential, that do not depends on country law annual limit of intake for members of public.

We chose the last one as the best SNF radiotoxicity definition in this document; in particular, we measured our radiotoxicity results in Sv/GWe-y, that is a derivate unit of Sv/Bq referred to the energy produced by a nuclear reactor for each ton discharged.

Results obtained are shown in Figure 1, these curves are built with update radiotoxicity coefficient data-bases from ICRP 72[5] applied to ORIGEN2.2. The continue curve (red) represents SNF and reaches the Reference Level, constant value (pink), in 2·10⁵ years.

ALEPH calculation is referred to the reference reactor described above with an irradiation period of 4.5 years inside reactor and a decay period of 10⁸ years after discharge (logarithmic scale).

![Fig 1. Spent Nuclear Fuel Radiotoxicity in Sv/GWe-y obtained by ORIGEN2.2 and updated coefficients from ICRP 72.](image)

2.1 Radiotoxicity concepts evolution: ICRP Recommendations.

Since the first years of 20th century international organisms are involved in understanding health hazard from buried radioactive high level waste considering at first internal dosimetry and related quantities. The first comprehensive publication of internal standards was ICRP Publication 2 (1959)[6] where new concepts as "critical organ", "standard man" and "biological half-life" were introduced to describe the physiological removal of radionuclides. One of the most important
innovation introduced by ICRP2 was the respiratory and gastrointestinal (GI) tract model. For each radionuclide the Publication presented “Maximum Permissible Concentration values” (MPC), expressed in \( \muCi/ml \), and calculated the annual dose limits, expressed in “maximum permissible annual doses”, that included concentration in air and water (corresponding to inhalation and ingestion pathways respectively) for both a 40-h week (occupational) and a 168-h week (used also for non-occupational exposure).

In following publications other secondary limits were introduced: "annual limit of intake", ALI and "derived air concentration", DAC. The annual limit of intake is the intake during a year of practice that would result in a committed effective dose equivalent of 0.05 Sv or a committed effective dose to a single organ of 0.5 Sv to "Reference man" (that updates “standard man”). These new limits and new respiratory and GI tract models were collected inside ICRP Publication 30 (1979)[7].

A fundamental change to the previous philosophy was presented in ICRP Publication 60 (1990)[8], and subsequent. In fact, during ’80s knowledge about radionuclide metabolic behaviour (minor actinides mainly) increased and more significant approach was adopted. In particular, limits of dose intake changed: 1 mSv for annual effective dose for population and 20 mSv for occupational annual effective dose limit values (they are annual limits taken in European Countries referred to 96/29/EURATOM). ICRP 30 values were, then, replaced and the new ones are expressed in terms of “Inhalation and Ingestion Dose Coefficients” or “Dose Factors” (Sv/Bq). We referred to ICRP72 last version coefficients. We analyzed also complementary studies about biological hazard potential carried out by different international organisms (i.e. BEIR and UNSCEAR) to comprehend the “cancer dose” as alternative radiotoxicity definition.

2.2 Evolution in US and European Radiological Protection Rules

From year to year, dose limit changes and drives to revise Countries rules in Radiological Protection. This work analyzed its evolution inside European and US rules.

Our analysis can be resumed as follows:
- European Community adopted ICRP60, and subsequent, with European Directive 96/29/EURATOM, received by single states. In particular, this work performed in SCK•CEN is referred to Belgium, where 96/29/EURATOM was taken in by 2001 Belgian law.
- 10 CFR Part.20 includes US “Standards for Protection Against Radiations”. The code last version is dated 1994 and it is referred to ICRP 30: it incorporates internal dose concepts and primary and secondary dose limit coefficients adapted from the recommendations of the International Commission on Radiation Protection Publication 26 (1977) and 30(1978). Although 10 CFR Part.20, 1991 revision, included the ICRP 60 recommendation to reduce the annual dose limit for members of the public from 5 mSv to 1mSv, it didn’t adopt the new occupational dose limit recommendation of 20 mSv/year.

As just indicated above, differences in radiotoxicity coefficients affect evaluation of SNF radiotoxicity and of the time to reach Reference Level.

3. Reference Level best estimate

Reference Level best estimation is necessary to correctly express the health hazard from SNF in term of radiotoxicity. The decay chains that lead to the achievement of secular equilibrium were simulated, taking values from present natural uranium isotopic atomic fraction, (ORIGEN2.2 calculation).

In order to obtain a right value, Reference Level was calculated considering that 1 ton of U enriched at 4.3% requires the use of 8 tons of natural uranium (Eq. 1). We obtained, in perfect agreement with literature (NEA-OECD Report[9]), a reference level equals to 3.07·10⁶ Sv/GWe-y that corresponds to 1.61·10⁵ Sv/tonIHM.

\[
\frac{\text{tonNatU}}{\text{tonFuel}} = \frac{(A - \epsilon)}{(C - \epsilon)} = \frac{(4.3\% - 0.2\%)}{(0.710971 - 0.2\%)} \cong 8
\]

4. Results obtained

In order to show that data from different sources imply often difficulties in understanding values, we performed interesting comparisons.

In fact, one order of magnitude difference exists between radiotoxicity from 2001 Belgian law, referred to the latest ICRP 72 Publication, and from ORIGEN own coefficients (ORNL, 1982) taken in
by 10 CFR Part.20 (‘Standards for Protection Against Radiations’), 1982 edition, based on ICRP Publication 2 MPC values that are old data (Figure 2). In fact, studies carried on between 70s and 90s on radiobiology corrected radionuclides effects on internal organs.

Moreover, a useful comparison, Figure 3, was obtained among ORIGEN2.2 original coefficients (1982, ICRP 2), coefficients from Belgian law (2001), related to 96/29/EURATOM (“Basic Safety Standards Directive on radiation protection”, 1996) and ICRP 72, and coefficients from 10 CFR Part.20 (1994) related to ICRP 30.

10 CFR Part.20 last edition (green curves) has the most restrictive radiotoxicity values. We tried to explain this result: 10 CFR Part.20 adopted the 1 mSv annual dose limit for members of the public but applies dose coefficients from ICRP 30 Publication that overvalued actinides effects on tissues if compared with ICRP 72.

Part of this work was finalized to analyze single isotopes contribution upon the whole of radionuclides extracted from the reactor. We found an imprecision inside ORIGEN2.2 (2002) decay library referred to Pr-143 radioactivity concentration guide for continuous ingestion of nuclide (WRCG value). In fact the value in ORIGEN is $5 \times 10^{-9}$ $\mu Ci/ml$ instead of $5 \times 10^{-5}$ $\mu Ci/ml$; this last one is the correct value present in 10CFR Part.20 (1982). This mistake was underlined during a calculation at $t=0$ (exit from reactor) because $t_{1/2}$ of Pr-143 is 13.57 days. Figure 4 and Figure 5 show, respectively, the wrong and
the correct distribution of few isotopes contribution to buried nuclear fuel radiotoxicity at time of discharge from reactor.

5. Conclusion

This work enables the use of updated radiotoxicity coefficients and, also, ALEPH and ORIGEN updated versions in order to develop a more accurate approach to ADS burning capability.

At first a theoretical burning capability efficiency calculation was, in fact, performed as a term of comparison with the industrial ADS burning capability. The theoretical burning capability acceptable value, that is a MA’s residual concentration left in wastes, obtained is 27%wt. able to reach RL in 2-3·10³ years. So, by the industrial scale ADS simulations performed, a transmutation efficiency equal to 72% after 3 transmutation cycles was reached that is a good value because it is nearly the same one than the theoretical transmutation analysis performed before[1],[10].

References

PROFILING THE GAMMA-RAY DISTRIBUTION OF THE
CEMENT LINING OF AN ESTABLISHED NUCLEAR POND AS
A FUNCTION OF DEPTH

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ABSTRACT

An investigation to determine the relationship between the depth of contamination within the
concrete lining of a nuclear pond and the ratio of x-ray to gamma-ray photpeak intensity
obtained from sodium iodide detector is described. The environment was simulated using
established radiation transport codes. Caesium-137 is assumed to be the sole source of
contamination. A detector was simulated across the energy spectrum of a source, and rastered
across its location from ±10cm at intervals of 0.5cm. A combination of point and distributed
sources were simulated within a concrete structure, as well as in free air to create a
comprehensive set of data. The results were then validated against experimental data and
demonstrate a high level of agreement. This investigation indicates that the x-ray / gamma-ray
ratio has the potential to localise contamination on the surface of the concrete lining; further
investigation is required to locate the contamination in three dimensions.

1. Introduction

The current focus of the civil nuclear industry in the UK has shifted toward the decommissioning of
obsolete facilities. This policy has been instigated by the UK government [1] via the foundation of the
Nuclear Decommissioning Authority (NDA) through the energy act [2]. This shift requires that many
legacy nuclear facilities are surveyed to optimise the design of processes and techniques to dismantle the
associated buildings that have often been contaminated through years of use. The early stages of this
process have already generated many diverse challenges that require novel measurement solutions.

An interesting challenge found at many nuclear facilities is that associated with the measurement of
entrained radioactivity within the walls of nuclear fuel storage ponds. These facilities are found at many
power plants and reprocessing facilities where fuel is stored following its use in a reactor. During the
lifetime of many of these facilities the pond linings absorb various nuclide species from the water in the
pond, as a result of contamination from the fuel stored in it. The water, used to shield and cool the spent
nuclear fuel, penetrates the concrete lining through hydroscopic action and areas of high concentration can
build up in defect areas, i.e. cracks and holes. This is especially relevant in very old facilities, that were
built at a time when little was known about the long-term effects of residual activity and hydroscopic
ingress and, often, ponds were not lined to prevent degradation of the concrete itself.

It is widely understood and expected that the penetration of radioactivity into porous media, such as
concrete, will be limited in depth. It is also expected that this depth will be dependent on the quality of the
concrete and the nature of the fuel stored in the pond, including such factors as the extent of clad corrosion
etc. In many cases the distribution of the entrained radioactivity is not uniform but, often, this distribution
is not known. If the concrete lining is to be safely removed during the decommissioning phase, it must
first be profiled in terms of nuclide penetration depth as well as resultant activity in order that dose
estimates can be established. Furthermore, and very importantly, the destiny of waste arisings from the
demolition of the plant can be determined before the decommissioning process starts if the depth profile of
the entrained activity is known, which improves planning and will potentially reduce the cost of waste
disposal significantly. A detector capable of profiling in this manner is the principal goal of the
Radioactivity Depth Profile Analysis Tool (RADPAT) project that is the subject of this paper. RADPAT is
a PhD bursary research project being conducted at Lancaster University, UK and is sponsored by the
NDA. The research presented in this paper describes an initial method trial and concerns the localisation
of caesium-137 at depth within the concrete lining of a contaminated nuclear facility.
2. **Background**

Caesium-137 is often one of the significant radioactive isotopes present in the water surrounding spent fuel stored in ponds [3] and, due to the tendency of caesium to be absorbed easily, it often becomes distributed throughout the concrete comprising the pond structure. On this basis, caesium-137 has been assumed to be the major nuclear species contaminating the concrete lining in this research. Caesium-137 has a half-life of ~30 years, eventually decaying to a meta-stable state of barium-137m via beta decay. The excited barium-137m has a half-life of ~2.5 minutes and decays with via \( \gamma \)-ray emission to its ground state; however this decay is usually assigned to the caesium-137 nuclide. The major contribution to the resultant energy spectrum comes from a mono-energetic \( \gamma \)-ray peak at 662 keV, with a smaller contribution from x-rays at lower energies; with the most significant peak situated at around 32 keV [4]. Due to the lack of kinetic energy the emitted x-rays from a caesium source have a limited penetrative depth in comparison with higher-energy \( \gamma \)-ray emissions. This is compounded in relatively dense materials such as concrete where, for example, the mass attenuation coefficient is 0.9601 cm\(^2\)g\(^{-1}\) at 30 keV [6]. If this is compared to the associated \( \gamma \)-ray, which has a mass attenuation coefficient of 0.0823 cm\(^2\)g\(^{-1}\) [6], it is clear that for increasing depth the ratio between the x-ray and \( \gamma \)-ray peak will vary predictably in a nonlinear fashion that has potential uses in depth analyses.

The results of an investigation into the relationship between the x-ray peak and the \( \gamma \)-ray peak of the various energy spectra obtained from rasters over a caesium-137 source are presented in this paper. Whilst scanning areas close to caesium-137 contamination, the x-ray peak at 32 keV will be relatively large compared to the rest of the energy spectrum. As the detector moves away from the contamination, this peak will decrease due to the attenuation of the x-rays within the medium between the detector and the source of the radiation. The mass attenuation coefficient of the emitted 662 keV \( \gamma \)-ray suggests that the relative intensity of this emission will attenuate at a much slower rate than that of the x-ray peak with respect to distance. Therefore, if the ratio is observed for each energy spectrum from a raster scan; the maxima of the x-ray/\( \gamma \)-ray ratio may indicate the position of contamination on a two-dimensional surface. The aim of this research was to determine whether this hypothesis is justified. Within the investigation the influence of two variables was examined;

- The difference between an open-air caesium-137 source and a source embedded in concrete. The fall in count rate due to the attenuation of photons from a localised source embedded in concrete will be affected as the detector moves away from it, as the photons have to travel through an increasing distance of concrete to get to the detector. This increases the probability of attenuation in a non-linear fashion and could significantly alter the relationship between the x-ray / \( \gamma \)-ray ratio and the distance from the source. To examine this variable scans was conducted with and without a layer of concrete.

- The effect of differently-shaped sources on the obtained projections. The intensity of radiation emissions from a radioactive source follow the inverse-square law with distance from the source, assuming it is a point source; otherwise the intensity distribution will exhibit an additional source of anisotropy that is dependent on the physical shape of the source. It is convenient in many cases, especially in simulation studies, to assume that the radiation propagates from a single point in space. However, in this research the shape of the source will influence the distance the radiation has to travel before it hits the detector and, very importantly, anisotropic source distributions in the concrete are highly-likely. Thus, the point source assumption can not be made. The discrepancy will be most apparent in the contrast of a point source and an evenly-distributed source across a plane normal to the axis of symmetry of the scanning detector. In this research both a point source and a distributed source have been examined as part of the investigation.

Two separate approaches were used during the investigation to obtain results:-

- **Simulation** – Radiation transport codes have been used to simulate the

![Figure 1: The attenuation of 32keV x-rays in aluminium](image-url)
environment on a standard PC prior to any experimentation taking place.
• Experimentation – A set of experiments were conducted. The results from these were used to validate the data obtained from the simulations.

2.2 Simulation Setup

Two transportation codes were used in the investigation, Geant4 [7] and MCNPX [8]. The initial parameters in both codes were matched as closely as the different input methods would allow. This technique was used to produce two separate sets of results that were sufficiently similar to be ascribed to the same set, which enabled any spurious results to be identified. This approach increased the confidence in the data as a different mechanism was used to obtain data from each of the codes. For photonic simulations Geant4 uses classical equations preceding a run to produce a set of tables that contain all the relevant simulation cross sections [9]. MCNPX uses an evaluated set of cross section libraries, such as ENDF-B VI, to interpolate the relevant information during the simulation run.

The material and geometric definitions of both codes were equivalent and were based upon the collimated ORTEC detector arrangement mentioned previously. All the materials and densities were defined from the NIST database of materials [10] and assumed to be at 270ºK. For convenience the signal processing chain of the detector was simulated as a single block of Mu metal which, due to the relatively low energy of the x rays under evaluation, would not represent a significant source of error. The disintegration of the source was defined using a probability distribution derived from [4]. This definition included all possible particles and energies through the full decay chain and was assumed to be fully isotropic. The point source was assumed to be infinitesimally small and located at the origin. The plane source, whilst similar in every other respect, was distributed with respect to the scanning plane from -2cm to 2cm.

2.3 Experimental Setup

The approach used throughout the investigation was based around a collimated ORTEC 905 7.62cm sodium iodide scintillation detector [5]. The collimator used in the experiments was manufactured from aluminium and designed such that the entire detector surface area was covered, except for the face used to create the scanning aperture. At the scanning face the collimator was designed to overhang by 0.3cm to limit the size of the aperture. The wall thickness of the collimator was specified by theoretical calculation of resultant intensity. The mass attenuation coefficient for x-rays at 30 keV, taken from [6], was used to derive the relationship between intensity and the thickness of aluminium. This was then validated using MCNPX [8]. This relationship can be seen in Fig. 1, where the trend of the simulated data agrees well with that of the theory. However the intensity of the simulated data is enhanced which is due to MCNPX using 32 keV x-rays as a source; as well as geometric effects being taken into account by the simulation. On the basis of the trend exhibited in Fig. 1 it was decided that a wall thickness of ~ 0.3cm would be sufficient for the collimator, as this thickness would attenuate ~90% of the incident x-rays.

The source location was defined as the origin in the experiments with the collimated detector rastered across from -10 cm to 10 cm at a height of 0.5 cm. Although every effort was made to keep the parameters consistent throughout the investigation, there were inevitable discrepancies between the various methods used that are detailed in subsequent sections.

The apparatus used in the experimentation is shown in Figure 2. The detector was coupled to an ORTEC Digibase module [11] which incorporates the necessary signal processing electronics. This device was connected to a standard portable PC through a USB cable which used ORTEC Maestro MCA emulation software to collate all the data fed back from the detector. Clamp-stands were used to hold both the collimated detector and the source so that any contribution due to scatter from surrounding materials was minimised. The source used in the experiment was a 39.7 kBq caesium-137 + barium-137m disk, aged at 1 year giving a relative intensity of 38.79 kBq.
3. Results

![Figure 3: MCNPX x-ray count rate for a) Free air source b) Concrete embedded source](image)

![Figure 4: Geant4 x-ray count rate for a) free air source b) source embedded in concrete](image)

During the simulated raster scans each energy spectrum was generated using 500,000 events as this gave sufficiently small uncertainties. The results obtained from Geant4 were normalised during analysis. MCNPX produces an output which is already normalised. For the experimentation a spectrum at each position was given 5 minutes to accumulate. This corresponded to an average of 1.16 million events per scanning iteration. The FWHH values were taken for both the x-ray and γ-ray peaks to account for the statistical response of the scintillation detector. The normalised results for the scans carried out in
MCNPX are shown in Figure 3 whilst those obtained from Geant4 are shown in Figure 4. Experimental data for the source are included on Figure 3a and 4a for comparison purposes.

4. Discussion

The data in both Fig. 3 and Fig. 4 demonstrate a general trend that as the detector scans across the source (in both plane and point variants) the ratio of the 32 keV x-ray peak intensity to the 662 keV γ-ray peak increases. As expected, the distance from the detector to the source decreases the attenuation of the emitted x-rays decreases more quickly than that of the γ rays. For both the point and planar sources, the simulated ratio data increase as the detector aperture approaches the origin, albeit at different rates with the planar source increasing more slowly than the point source and peaking closer to the origin. As the detector scans further toward the origin the source moves into the aperture of the detector. This removes the effect of the collimator and increases the intensity of both the x and γ rays, though at significantly different rates. The differential rate causes the local maxima at approximately ±4 cm where the collimator influence is gradually removed, increasing x-ray intensity rapidly whilst the intensity of the γ-rays remains fairly constant. However, as the position of the detector moves across the region of origin (i.e. between -5 cm and +5 cm) the ratio falls in a nonlinear but symmetric manner, both in the planar and point source cases and for MCNP and Geant4, with the minimum corresponding to the position at the origin. In this region it is reasonable to assume that the x-ray detection efficiency will not increase much more than is observed at, say -5 cm or +5 cm. However, as more of the detector crystal moves into the path of the source as it approaches the origin, the absolute detection efficiency for γ rays emitted by the source will increase, as more sodium iodide material is presented within the aperture -5 cm and +5 cm. Thus the x-ray/γ-ray ratio falls in this region.

The experimental results in the free-air case demonstrate a somewhat different trend. Whilst the ratio increases in a consistent way with the simulated data, the fall in the ratio in the -5 cm/+5 cm region is not replicated experimentally and this inconsistency is currently poorly understood. However, the experimental technique employed in this research could be developed significantly to remove several sources of error which may improve the confidence with which the data is held in future. For example, the detector position is possibly too close to the source such that its non-point disc shape influences the data more consistent with the planar source limit.

5. Conclusion

This investigation has shown that it is possible to localise contamination on a 2D surface by using the x to γ-ray ratio to find the local minimum exhibited by the simulated data at the origin. Further research will take into account the depth of the source; with the aim of finding a relationship between it and the x-ray to γ-ray ratio. Whilst the penetration of the x rays in this context is unlikely to exceed concrete thicknesses of ~ 4 cm, this is largely consistent with the approximate scabbling capability of many decommissioning techniques currently in use to dismantle contaminated concrete surfaces. Irrespective of the distribution of caesium-137 contamination in the concrete and its specific activity i.e. whether planar or localised, the relative proportion of x ray to γ ray intensity has the potential to distinguish its depth due to the relative differences in attenuation of these photon emissions [12]. This technique will aid further research to produce a 3D radioactivity profiles in legacy facilities.

6. References

MAESTRO: A HYDRAULIC MANIPULATOR FOR MAINTENANCE AND DECOMMISSIONING APPLICATION

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ABSTRACT

Compared to electric manipulators, hydraulic manipulators can handle very high payloads with respect to their size and mass. However, due to their limitations in force-torque control, they are usually disqualified for precise manipulation.

CEA, in collaboration with CYBERNETIX developed a complete remote handling system featuring the advanced hydraulic robotic arm MAESTRO. The complete slave system, 10 kGy radiation-hardened, is composed of the MAESTRO arm, a 2 m long, 100 kg payload 6 degrees of freedom slave hydraulic manipulator, mounted on an embedded unit made of a 210 bars hydraulic power pack and a slave controller. The master station is made of the latest generation VIRTUOSE master arm supplied by Haption, including a dedicated master controller. The system specifications were defined according to the requirements of decommissioning activities in existing nuclear facilities and maintenance scenarios of the next step fusion reactor ITER. Specific attention has been paid to the decontamination and maintainability aspect of the robot, in nuclear conditions. Validation of the system has been achieved through test campaigns during 1000 hours of an endurance test completed with operational evaluation on a set of tasks.

Using the generic TAO (Computer Aided Teleoperation controller) control software designed by CEA, specific force-torque control loops were developed to improve the manipulator performance, thus allowing the MAESTRO arm to be used in a traditional teleoperation master/slave application with force feedback. In addition to this control software, the graphical supervision software Magritte provides the operator with an additional interface to manage and monitor the system. Repetitive tasks like tool picking can therefore be managed by the system while the operator focuses on the main task. Thanks to a collision detection algorithm, Magritte warns the operator when the tool or any part of the slave arm comes too close to a sensitive unit.

This paper presents the complete MAESTRO system and gives up an overview of the research and development activities currently carried out:

- Real-time failure detection: when dynamic simulation helps managing hardware failures.
- Making hydraulic cleaner and safer: from oil to demineralized water hydraulic.
- Collision free teleoperation: from collision detection to dynamic collision avoidance.
- Decreasing working load on the operators: providing assistance functions for enhanced teleoperation tasks.
1. Introduction

The MAESTRO hydraulic manipulator belongs to the class of servomanipulators. This class of telerobotic systems appeared in the early 80’s with the progress on computer assisted teleoperation. Compared to traditional through the wall workstations equipped with mechanical master-slave systems, or to power manipulators with limited control and speed performances, these new systems provide innovative features and improved capabilities, including:

- operation from a remote control room located in an unrestricted access (cold) area,
- use of different arm morphologies and technologies for the master and the slave,
- work in the Cartesian coordinates,
- compensation of weight of handled tool,
- adjustable force and speed ratios in the force feedback loop,
- handy automatic robot modes (tool picking, return to rest position...),
- virtual mechanisms to assist operator in tricky tasks (sliding, axial or even more complex),
- virtual reality to assist the operator in complex tasks,
- real-time collision-avoidance to protect both environment and manipulators from shocks.

If power of electric motors is enough to supply good force feedback capabilities to the operator in master arm stations, requirements in the hot zone sometimes require the capability to supply high forces that standard electric motors are unable to provide in a limited space. Starting from a hydraulic manipulator developed for offshore applications, CEA-List, in collaboration with the French robotics company Cybernetix developed a complete telerobotic system for nuclear operations. The MAESTRO Telerobotic System (see figure 1) is composed of:

- A master station including:
  - a graphical 3D supervision interface,
  - video display monitors,
  - a Haption Virtuose 6D master-arm,
  - a master-arm controller.
- A slave station with:
  - a 6 degrees of freedom hydraulic manipulator,
  - a rad-hardened embedded slave-controller,
  - an embedded hydraulic power pack,
  - a remotely controlled PTZ video camera with tool tracking capabilities.

After a brief overview of the current capabilities of the MAESTRO system, this paper presents the latest developments carried out at CEA-List in the field of terobotics.

2. Design description and performances of the manipulator

Made of Titanium, the Maestro slave arm is a 6 degrees of freedom manipulator with two length configuration possible. It is typically 2 meters long with a payload capacity up to 100kg. The latest design work took advantage of good performances of original design from Cybernetix and Ifremer dedicated to offshore applications, improving it to satisfy easy decontamination requirements, eg: with smooth surfaces, avoiding any contamination traps in the design, leak tight, and pressurized housing capabilities.
Qualification of the complete system for Remote Handling (RH) in nuclear facilities application ran through a validation process including long term reliability testing during 1000 hours of the complete arm. Tests profile was based on rehearsal of a typical working trajectory including various payload handling. The trajectory was defined according to position records of the real manipulator during a representative teleoperation task:

- Tool picking.
- Task completion with tool.
- Tool removal.

Rad resistance of the system was proved in an irradiation facility. A cumulated dose of 10.65 kGray at a dose rate of 74 Gray/hour was reached before the first stop of the system. The mock up consisted of:

- a single elbow joint,
- a resolver,
- a servovalve,
- two pressure sensors,
- a low level controller.

It has to be noticed that after a 2 hours power off phase it was possible to restart the mock-up due to a recovery effect of the electronics. Developments in progress for the slave arm focused on the following directions:

- minimize the impact of a leak in the hot zone,
- improve safety of the control loop,
- improve performance in force control mode,
- reduce the tuning procedures time.

A change of fluid from oil to water was proposed to reduce impact of leaks (see [1]). The use of pressure controlled servovalves improved safety, force control performance, and tuning time. Driving requirements to adapt a joint to use water were:
- use corrosion resistant materials,
- reduce clearances (direct impact on internal leaks due to water’s low viscosity),
- prevent contact between water and components with poor corrosion resistance,
- adapt seal materials and properties to water.

In addition, attention was paid to control properties and quality of the water used during the trials. Tests were carried out with demineralized water on a single vane joint mock-up (see figure 2). Performance achieved with water is equivalent or even better than with oil.

Figure 2: Water hydraulics test bench

At the time of this paper, the mock-up successfully ran during 530h before a failure of the power pack occurred, thus stopping prematurely the tests of the joint and the servovalve. Up to that level no significant reduction of the joint performance were noticed.

Improvement of the force control loop is achieved by using pressure controlled servovalves to drive each joint. In that scheme, the controlled parameter is directly linked to the force, and this has a direct impact on the stability of the control loop. Using these components also allows removal of all pressure sensors and therefore reduces the probability of failure of the system. Prototypes of oil pressure controlled servovalves with space and performances requirements needed by a Maestro manipulator were developed in collaboration with In-LHC. Integration of all servovalves in the slave arm proved the feasibility of the concept. Performance was better than observed with flow servovalve and a reduction of the time period of the control loop by a factor of two was possible.

3. Real time failure detection

To detect possible failure or collision with the environment, developments of model-based monitoring strategies were tested (see [2]). The global dynamic model of the arm developed for such applications takes into account all inertia parameters of the arm, the centrifugal and Coriolis effects, gravitational torques, a contribution of the friction to each joint and offset values of the pressure sensors. Identification of all parameters is achieved with help of numerical regression methods. Thirty five trajectories were defined and used to identify all parameters of the model. Descriptions of all
trajectories were made according to the kinematics capabilities of the arm and real remote handling tasks.

Comparison between the estimated torque (model) and the real torque (sensors) is used to detect collisions of the arm with the environment. Torque values need to remain within boundaries of the residual error identified for each axis. If not, a collision (or failure) is detected.

![Figure 3: Collision detection](image)

Results showed good agreement between predictions and measurements during the trials (see figure 3) although some improvements were necessary for low speed movements due to noise on speed evaluation, under-estimation of friction forces and hysteretic phenomena. Implementation of low pass filtering of the signal and definition of threshold values were necessary to overcome this trouble.

4. **Graphical supervision**

CEA-LIST developed a 3D graphical supervisor to ease control in real environment of teleoperated systems such as the Maestro [3][4]. It has been assessed by operators on the prototype of the system, with indirect viewing of the environment. A complete set of maintenance tasks (cutting, welding, grinding...) were carried out with this tool.

Teleoperation and especially decommissioning intervention tasks are very stressful for an operator since he must interact in real time with the environment dealing with complex systems. Remote tasks are not repetitive and usually undefined since work depends on observations during the interventions. Most of the time, the operator does not have direct viewing of the operating area.

This supervisor (named MAGRITTE) was developed to ease operator's task during intervention. The main requirements consist in letting the operator interact with the environment through a 3D model of the workplace. Through this interface, operators elaborate robot trajectories, play them and control their execution in an intuitive way. MAGRITTE offers graphic assistances, making easy robots programming and control. It interfaces to robots and tools through an execution controller, allowing updating model state according to the real situation. In MAGRITTE, specific processes functions (welding, ultrasounds inspection, grinding) are gathered in dedicated trade modules.
Moreover, collision test algorithm warns the operator, preventing collisions with the modeled environment. This feature is called passive anti-collision but can only be used to prevent collision (warns the operator or stops execution of the task) and so may not be of any help in case of use in very constrained environment.

5. Assisted teleoperation for enhanced teleoperation task

More recently, progress has been made in computing power and virtual reality makes it now possible to interact with an accurate mechanical model using a haptic device. Main applications are currently for training [5][6] and digital prototyping (accessibility, maintenance).

Concerning the teleoperation field, the main benefit of using a physics engine with a teleoperated system consists in providing the operator with an active collision avoidance feature [7][8].

A prototype of a supervisor has been studied on the basis of MAGRITTE concepts and haptic software library developed by CEA-LIST named XDE (eXtended Dynamic Engine). This function differs from the passive anti-collision feature as it ensures collision avoidance by generating repulsive efforts, thus maintaining execution of the teleoperation task. At that time, the real robot is coupled with a simulated one and the operator uses a 6 DoF mouse to generate movement on the simulation and the robot. Because the simulation not only detects the collisions but also simulates the robot behavior, the operator can then handle the robot safely, without taking care of obstacles of the environment.

Figure 4: passive collision detection
Future work will be done to link the system with a force reflective master arm, and then provides the operator with force feedback on simulated collision. This functionality will allow using the master/slave system in a real force feedback mode, preventing collision of known, modeled objects. The issue of localization of objects will also be addressed.
6. References

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CANBERRA solutions for source location and activity determination for investigations of ORCADE dismantling project at AREVA NC La Hague site

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Abstract

The operations of dismantling of nuclear installations are often difficult owing to the lack of knowledge about the position, identification and radiological characteristics of residual radioactivity.

As part of the investigation team for the ORCADE dismantling project at AREVA NC La Hague UP2-400 reprocessing plant, CANBERRA had the opportunity to use new nuclear measurement systems and modeling tools to develop a methodology to locate and characterise radioactive hold up.

The methodology uses different types of dose rate meters, gamma spectrometers and modelling tools, depending on the type of problem.

In this paper, the methodology and tools are described, followed by typical case studies, including hot cell activity evaluations and drum categorization methodologies. For each case, the detector and modelling tool choices are explained and justified.

This methodology helps to prepare and execute post-operational clean out and dismantling activities. These examples give concrete insights into their significance and the productivity gains they offer.

Keywords: D&D, Gamma spectrometry, radioactivity modeling, source location, source identification, MERCURAD, PASCALYS, ISOCS, CARTOGAM
1. CONTEXT OF D&D MEASUREMENT ACTIVITIES

Dismantling a nuclear installation is often difficult due to the lack of knowledge about the position, the identification and the radiological characteristics of the contamination. In that way, the contamination is particularly difficult to define in a significant global background when the activities are relatively high. For example, identification and estimation of the activity become more complex in hot cells, where space is limited and human intervention is costly in terms of accumulated dose.

CANBERRA, the Nuclear Measurement Business Unit of AREVA, not only designs, manufactures and sells a complete range of instruments, but provides solutions and services to take care of the global problem depending on customer needs. In that way, the following items have to be specified:

- definition of the needed investigations,
- detector choice,
- dose rate modeling,
- coupling dose rate measurement and model,
- coupling gamma spectrometry measurement and model,
- coupling neutron measurement and model.

To be able to propose the best solution, CANBERRA has developed knowledge and intervention strategies based on its feedback experiences in many countries.

Each part of the scene characterization is described in the following chapters. Thanks to this methodology, CANBERRA is able to give the customer not only measurement results, but also activities, localization and eventually to guaranty safety or process threshold corresponding to the customer’s needs.

2. ORCADE INVESTIGATION PROJECT STAKES

2.1. Generalities

The ORCADE UP2-400 dismantling project at AREVA-NC La Hague began in late 2002 when all new facilities in UP2-800 had started. The project concerns all the facilities of the first reprocessing plant: from dissolution to U, Pu and fission products (FP) extraction and storage in tanks. To support all dismantling sub-projects, a transversal investigation project was created in 2005.

2.2. Stakes of the Investigation ORCADE’s project

The Investigation project support the different missions of the global ORCADE project which are :

1. Definition of dismantling scenario,
2. Design of waste packaging installation,
3. Good waste categorization,
4. Radioactive discharges optimization,
5. Safety analysis (dose rate, criticality…)

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To be successful, the investigation project needs many skills and means as:
- video investigation capabilities,
- documentary research,
- laboratory analysis,
- nuclear measurements and modeling.

Inside the investigation project CANBERRA is in charge of this last item.

3. **NUCLEAR INVESTIGATIONS IN A DISMANTLING PROJECT**

3.1. **Necessary data retrieval**

The first phase of nuclear measurement investigation is crucial. A facility prepared to dismantling has usually a long history of operations with numerous events which have occurred its lifetime. The knowledge of these events is very important in the definition of measurement strategy. These first assumptions usually come from interviews with the facility previous operators, allowing definition of a first model. A good expectation in an activity evaluation is already to take in account right information (detector choice for example).

3.2. **Scene modelling**

For a high activity cell, a first model evaluation can save a lot of money for the investigation phase. From large activities panel assumptions and geometry descriptions of the scene, sources would be able to be placed in the model and dose rate evaluation would be determined as a range of magnitude, which will define the type of investigation:

- Is the staff able to enter the cell?
- Possibility to introduce nuclear measurement?
- Logistics required for nuclear measurements?

For gamma emitters, CANBERRA uses MERCURAD application based on MERCURE V6 (reference [1]) gamma attenuation code developed by the French Atomic Commission at Saclay (CEA/SERMA). This code is used but also sold by CANBERRA. It has a very convenient interface which can be used by a technician for modelling a complex scene within about 30 minutes. Figure 1 summarizes this first part of the methodology.

![MERCURAD dose rate evaluation code](image)

For neutron emitters, only MCNP is used because no simplified code for 3D geometries exists yet.
3.3. First measurements to confirm hypothesis
The first evaluation of dose rate will allow selecting detectors, electronics and associated shielding and/or collimators. For very complex problems (which concerns mainly high activity cells), several iterations are needed. Simple measurements have to be done first, before complex measurement on model and first hypothesis refining.

4. MEASUREMENT POSSIBILITIES

4.1. Main solutions
The measurement solutions are very numerous. Usually only gamma and neutron detectors are used. Alpha and beta emitters are detected by their gamma emission, which explains why the ratio between gamma and alpha has to be known in the case of total gamma counting (without spectrometry). This ratio can have a deep impact in waste categorisation. Gamma spectrometry measurement will be preferred when this ratio cannot be easily identified or when the gamma emitters are numerous.

4.2. Total counting measurement

4.2.1. Gamma measurement
Ionization chambers are usually chosen because of their sensitivity and their ability to be used in very hot cells. In that case, the electronics will be placed away from the detector and current amplification electronics will be preferred. Geiger-Müller dose meters can also be used. Such detectors can be very small and are very useful when there is little space to introduce the detector in the hot cell.

4.2.2. Neutron measurement
Because of their sensitivity, ³He tubes are preferred. But they are also sensitive to gamma. In the case of high gamma emission, BF₃ detectors will be chosen. For very high neutron emission, fission chambers can also be used. All these detectors can only detect thermalised neutrons. If the neutron spectrum in the cell is not a thermal spectrum, the neutrons will have to be thermalised, usually with polyethylene blocks which increase the space needed around the detector.

4.3. Source location: CARTOGAM possible use
The CARTOGAM (gamma camera) will be used (see reference [2]) when the location is not known and when it is very important to know the radioactivity’s location in order to define the dismantling scenario. This tool can drastically decrease the number of needed measurements. The main advantage of the CARTOGAM system compared to others is that it superimposes visible and gamma images using the same optics (see figure 2).
The use of CARTOGAM will also simplify the need for sampling. The localization of the needed sample is made easier as the sample contains radionuclides. Laboratory analysis on these samples will determine all radionuclides and specific ratios. To avoid expensive laboratory analyses, gamma spectrometry measurements can also be done in the field.

4.4. Source identification: use of NaI, CZT, or Ge detectors

While gamma spectrometers are useful for source identification, NaI detectors are a cheaper solution, with a poorer resolution (<10% at $^{137}$Cs) but a good efficiency for very low activity determination. They will be preferred for simple gamma spectra with known radionuclides only the proportion of which has to be determined.

CZT (Cd-Zn-Te) detectors (figure 3) are small and will be preferred in high activity cells. Their resolution is better than NaI detectors (usually 1.5% to 3%). CANBERRA frequently uses this type of detector coupled with CARTOGAM or adapted to the InSpector 1000 Hand-Held MCA (figure 4), which easily distinguishes $^{60}$Co from $^{137}$Cs.

Figure 2: CARTOGAM measurement head with its PC and scheme of the image processing

Figure 3: CARTOGAM coupled with a CZT detector

Figure 4: InSpector 1000 with NaI and CZT probes to be introduced into very small holes
The last and best solution is to use germanium detectors with a resolution usually around 0.2%. This is the solution for very complex spectra. This type of detector can determine the isotopic composition of U and Pu. CANBERRA developed BEGe (Broad Energy Germanium) detectors for 15 years. Such a detector coupled with an ISOCS modelling code (see reference [4]) is a very versatile solution. CANBERRA also proposes a wide range of Ge detectors for specific problems.

5. SCENE MODELLING SOLUTIONS

5.1. Why do we model the scene?

Detectors give counts per second. The modelling of the scene with sources location assumptions, allows determining not only counts, but also proportions, radionuclides and activities. The use of CARTOGAM before modelling can help to make better assumptions in source location and drastically decrease the uncertainties. The following figure illustrates this methodology.

Figure 5: Cart for Ge detector and associated collimator

Figure 6: Methodology for activities determining
The determination of geometrical efficiency and detector efficiency allow determination of activities from a spectrum in counts per second via the following formula:

\[
A(Bq) = \frac{M(cps)}{\varepsilon_{geom}(\gamma / Bq) \times \varepsilon_{det}(cps / \gamma)}
\]

Where:
- \( A \) is the activity in Bq
- \( M \) the measurement of the peak net area in counts per second (cps)
- \( \varepsilon_{geom} \) the geometrical efficiency in gamma rays per Bq or \( \text{gcm}^{-2}.\text{s}^{-1} / \text{Bq} \)
- \( \varepsilon_{det} \) the detector efficiency in cps per gamma ray or cps / (\text{gcm}^{-2}.\text{s}^{-1})

Different codes can be used as gamma ray attenuation calculation. The use of the code is discussed below.

5.2. Which model and calculation to be used?
A very precise model is not often needed. The model has only to take into account the objects and layers between the assumed sources and the detector. Often the main uncertainties come from not knowing the layers thickness between the source and the detector. In that case a very precise code is really not needed. Nevertheless, a more precise code can be useful when the scene is known more precisely, when the geometry is complex, or when there is a need to reuse the geometry model for other applications. This case often occurs for hot cells modeling. Where high activities are concerned, the facility’s personnel issues are quite important and the time for modeling is significant.

5.3. ISOCS use for easy modelling
ISOCS gamma code attenuation is the best compromise for simple scene. The user interface uses templates which cover the most common geometries. With this approach, simple geometries can be modelled by technicians in less than 15 minutes. It allows the model to be done in the field during the measurement.

5.4. MERCURAD-PASCALYS advantage
We have seen that MERCURAD is a complete 3D code for dose rate evaluation from any kind of gamma spectrum and activities. It can also be used to calculate activities from a measurement spectrum. This way to use MERCURAD is called PASCALYS. The main advantage of PASCALYS compared to ISOCS is its modeling capabilities. An other advantage concerns the detector efficiency. As PASCALYS considers the detector as a point, any kind of detector can be used easily with this modeling approach. The third advantage concerns the possibility of reusing the modeling scene. In a hot cell complete study, the geometry model can be used for other applications as health physics studies or dismantling scenarios.
With the MERCURAD-PASCALYS tool, a complex geometry can be generated and stored in a database. A complex geometry can be used by PASCALYS to determine activities from one field measurement, and then reused with MERCURAD to determine the activities dose rate. Objects can be easily removed from the scene (simulating dismantling activities) and dose rates recalculated.

5.5. General synthesis
During repairing and dismantling activities, multiple types of detectors can be used. As the source activities determination is very complex, CANBERRA proposes different approaches depending on customer environment. CANBERRA is able to propose a large range of solutions.

Therefore the model used in the activity determination can be reused when dose rates have to be determined. It provides a consistent tool for engineering studies and dismantling scenarios.
6. ORCADE PROJECT EXAMPLES

6.1. Calibration and quality insurance

Inside the AREVA-NC investigation project, CANBERRA has deployed the previous described tools and methodology to characterized different hot cells in the following facilities:

- HAO : High Activity Oxide facility where fuels are previously sheared and dissolved,
- HADE : High Activity facility where Decanting, fission product and actinides Extraction are performed,
- HAPF: High Activity facility where Products issued from Fission are stored.

All the measurement instruments used for ORCADE project were previously calibrated at an irradiator facility (at AREVA-NC La Hague or at CANBERRA Loches irradiator). Both are COFRAC certified. Technical notes, procedure, and storage of scene modelling insure a complete tracability of the results.

6.2. Activities determination in high activity hot cells

The following methodology is performed by:

- Using laboratory sampling analysis to determine gamma & beta emitters and ratios,
- Using equipment geometrical data as model and associated assumptions concerning liquid or sludge volumes,
- Confirmation of sample analysis by dose rate measurements via very thin GM tubes and CZT gamma spectrometry measurement,
- Model via MERCURAD code and MCNP to confirm MERCURAD results when gamma scattering effects occur,
- Determination of transfer function at each measured point (Gyh⁻¹/Bq)
- Quadratic minimization of differences between dose rate measured values and model results.

For example, the modelling of a very complex cell in the HAO facility was performed (see figure 8). After fitting the 44 dose rate measuring points to MERCURAD dose rate results (from few mGy/h up to 8 Gy/h), the total beta activity was estimated at around 45 TBq.

![Image](image.png)

**Figure 8:** Modelling of one complex HAO cell
A second example concerns the activity evaluation of tanks containing fission products. The only probe to be introduced in this cell was the very thin CANBERRA Geiger Müller detector (mounted inside an $\varnothing < 8$ mm thermowell). These GM tubes where previously calibrated in an irradiator to establish their response up to 200 Gy/h.

**Figure 9:** GM dose rate measurements on 2 tanks of fission products

MERCURAD modelling was performed to determine the total beta activity, established around 2300 TBq and an uncertainty about 50%.

### 6.3. Measurement in Uranium Middle Activity facility (MAU)

In the Middle Uranium Activity facility (called MAU), the stake of ORCADE project was to determine when the different tanks and equipments are considered as sufficiently rinsed. The aim it to make the equipments compatible with the very low activity wastes specification (called TFA). For Uranium, the TFA limit is about 100Bq/g of final waste. Consequently the ORCADE project asked CANBERRA to achieve a methodology to classify the equipments according to 3 different categories:

- TFA waste
- Not surely TFA waste: equipment to be investigated
- Surely not TFA: equipment to be rinsed

The equipments were differentiated according their geometry and weight:

- Light ($< 500$ kg),
- Intermediate (from 500 kg to 1 Ton),
- Heavy ($> 1$Ton).

The equipments were modelled with MERCURAD and the measure was performed inside the facility.

To types of spectrum were achieved:

- almost Uranium spectrum,
- almost Cs137 spectrum.
So, it was possible to categorize the waste:

Figure 10: Differentiation between TFA, undetermined and non TFA waste

For light equipment, it’s very difficult to distinguish TFA and non TFA equipment as the limit around 80nGy/h which is nearly the background. For large tank the limit value of 200nGy/h is easier to measure.

These results which are preliminary, have to be correlated with more precise gamma spectrometry Ge detectors in situ and on final drums.

7. CONCLUSION AND PERSPECTIVES

CANBERRA proposes a complete set of solutions for radioactivity location and determination. According to the problem facing the nuclear facility, different measurements and modeling approaches can be used. Many tools have been used for ORCADE dismantling project in La Hague to help the investigation team and increase the knowledge of the radioactive contents of the different facilities.

Within the ORCADE project, CANBERRA contributes to the three main drivers in accordance with sustainable development goals:

- Environmental aspects: reduction of the toxicity of the waste
- Finance: optimization of the global project cost by contributing to the planning of dismantling scenarios
- Social aspect: optimization of individual radiation exposure by detailed preliminary measurements facilitating use of ALARA methodology

New waste categorization challenges await CANBERRA at AREVA-NC La Hague site. Such a categorization doesn’t consist only in measuring the final waste at the end of dismantling, but also to help project teams during the realization phase. The target is also to provide a good level of decontamination and to optimize the final cost of dismantling.
8. REFERENCES


1. **Introduction**

In accordance with the Russian practice the NPP radiation safety is provided by the Automated Radiation Monitoring System (hereinafter referred to as ARMS). The aim of ARMS development is to obtain on-line comprehensive information of the radiation situation and process equipment condition. The system monitoring data shall confirm the fact that the radiation impact on the personnel and population from the NPP side is within the specified limits.

This paper considers the peculiarities of ARMS developed by PROM Engineering in the framework of Kudankulam NPP construction in India. ARMS has been developed on the basis of valid in RF normative documents which regulate the requirements of the personnel and population protection from ionizing radiation impact as well as the requirements of radiation monitoring arrangement, namely:

- Standards of radiation safety NRB-99;
- Sanitary rules of nuclear power plants design and operation SP AS-03;
- General provisions of nuclear power plants safety assurance OPB-88/97.

Moreover, specific requirements of Indian national standards of radiation safety have been taken into consideration during the system development.

2. **Monitoring scope**

ARMS is a system of radiation safety at the NPP monitoring which allows damage to protective barriers to be identified at an early stage and prevents the penetration of radionuclides into the environment. The information exchange between ARMS and the Automatic Process Control System (hereinafter referred to as APCS) enables continuous analysis of the NPP state to be carried out and failures of the main process equipment to be predicted.

In the framework of the united ARMS system of new generation a complex solution of all tasks of radiation monitoring at the NPP is provided. These tasks comprise the following kinds of monitoring:

1. radiation monitoring of processes including:
o protective barriers (fuel elements claddings, primary circuit equipment, steam generators, secondary circuit equipment, containment);
o radioactive media leaks in process equipment;
o efficiency of gas treatment, water treatment and ventilation systems;
2. monitoring of the radiation situation at the power unit and at the Site;
3. monitoring of gaseous and particle releases into the atmosphere;
4. monitoring of radionuclide releases into the open aquatic environment;
5. monitoring of the spread of radioactive contamination at the NPP site and outside the NPP;
6. monitoring of collective and individual doses received by the personnel.

The crucial role of safety assurance is played by measuring channels which trace the main parameters changes in continuous automatic mode. These very channels are capable to provide prompt assessment of the radiation situation in case of emergency situation development.

ARMS combines both continuous (remote) and periodical monitoring functions which are mutually complementary. Periodical monitoring consists of measurements made using mobile and portable instruments and it includes also manual sampling and spectrometric or radiometric analysis of samples under laboratory conditions. The automation of measurements data obtaining, storage and processing by means of specific programmes package is provided in ARMS for the purpose of periodical monitoring.

One of the differences of ARMS for Kudankulam NPP from other similar systems is the integration of Automated Personal Dosimetry Monitoring System (APDMS) into ARMS with the uniform software. During the development the high emphasis was placed on the fact that radiation safety service personnel can receive detailed information at the same time about individual doses and the radiation situation in different NPP rooms and about the process equipment state. It will improve the radiation safety service work and will allow to reduce radiation doses caught in the course of radiation-hazardous activities. The automation of measurements as well as the implementation of a set of functions pertaining to calculation and planning of the individual doses for the personnel, accounting of the time of radiation-hazardous activities implementation, issue of radiation work orders and others has been made for APDMS.

ARMS allow to extend its functional possibilities and, for example, to include the following kinds of monitoring:
- monitoring of the radioactive contamination of the environment;
- radiation monitoring of processes related to radioactive wastes management;
- monitoring of the meteorological situation.

Thus at Kudankulam NPP the radiation situation monitoring in the observation zone is performed by a specific automated system (ARSMNS in accordance with the Russian practice) using stationary monitoring posts. Then monitoring data are transmitted to ARMS for the purpose of their analysis and storage as well as to obtain the prediction of the radiation situation and assessment of radiation doses of the population in the NPP area in case of emergency situation.

Hence the united automated system covers all three main objects of monitoring as follows:
  a) nuclear power plant (processes);
  b) the personnel and population living within the NPP location area;
  c) the environment within the NPP location area.

This scale and heterogeneous scope of monitoring requires special approach to the system creation: its design and the software development.

3. Structure
ARMS has a distributed hierarchical structure. ARMS structural diagram for the nuclear power plants with two power units is provided in Figure 1. Its lower level is a set of measuring channels consisting of individual monitors or detection units and data processing and transmitting units which are united by data communication lines. Lower level devices ensure the measurement of the current values of the parameters to be monitored as well as local alarming in case the monitored parameters exceed the preventive or emergency limits. Besides, the especially important parameters preventive or emergency excess alarms are replicated directly from lower level devices on safety panel and generalized mimic panel installed in the Unit control room.

Upper level consists of concentrators made on the basis of industrial controllers, automated workplaces of operators (hereinafter referred to as AWP) and database servers. The upper level hardware carries out data accumulation and processing, recording into database, archiving and presenting the results of monitoring to the radiation safety service. The concentrators inquire by cycle the lower level measuring channels via communication channels RS-485, obtain the current data from them and process this data up to the unified accepted format ready to be transmitted to AWP. In addition the concentrators collecting data of radionuclides release into atmosphere through the NPP ventilation stacks calculate total release of radionuclides into atmosphere per day and compare the day release with the control level.

AWP makes regular diagnostics of the software and hardware. The equipment right up to individual devices, units and modules of the software are subject to diagnostics. ARMS and APCS data exchange is performed via concentrators (interface Ethernet 100Base-FX).

Two-level structure taken as the system basis makes it possible to improve the system reliability. The crucial point of the structural reliability improvement is the lower and upper levels autonomy (self-contained operation). Due to this fact the monitoring is going on to be performed in case of the upper level equipment failure or communication lines loss, since the alarms of preventive and emergency limits excess are transmitted to safety panel of Unit upper level control system (ULCS), and the results of measurement are displayed at the lower level devices displays and are saved into memory.
All AWP are unified and if one of them fails monitoring functions are transferred to any other computer complex. It is provided at the expense of links unification and the software proper structure.

Only unified self-contained measuring channels with standard interfaces for all hardware are used in ARMS. Taking into account the fact that 65% of total number of the measuring channels are used for gamma radiation dose rate monitoring a wide-range channel (covering eight decimal exponents) common for all NPP operation conditions that is for normal operation and for emergency situation – is used in the system. It further contributes to the system manufacture, adjustment and cost.

In the course of design process a differential approach was used according to which the distribution of all ARMS tasks between its sub-systems is determined by their relation to safety (classification as per the Russian standard OPB-88/97). Sub-systems and hardware included into sub-systems are divided into safety-related normal operation elements (safety class 3N) and not-safety related elements (safety class 4N). A number of increased requirements of resistance to external impacts, electric power supply, electromagnetic compatibility, error-free running time and etc is set for the equipment of 3N class. And as well a redundancy of power lines and communication lines shall be fulfilled for 3N class equipment related to the upper level. All these make ARMS design more complicated and set the additional requirements to data exchange between sub-systems of different safety classes. Thereby the data are transferred through interfacing gateway the role of which is played by the concentrator connecting local area network of safety class 3N and 4N.

Radiation monitoring system shall stay operable under all NPP operation conditions including design-basis accidents, so ARMS includes independent sub-systems of normal operation and emergency control. Emergency monitoring systems must continue functioning in case of any design-basis accident, they are made with the double redundancy of the measuring channels, concentrators connected with them, as well as of communication and power supply lines.

This flexible structure of hardware and software makes it possible to modify the system configuration and functioning conditions, if required. The same flexible structure allows ARMS to provide radiation monitoring during the NPP decommissioning.

Another problem is connected with ARMS service life. During the nuclear power plant whole operation life this system must stay serviceable within at least 30 years. Taking into account the fact that specific hardware service life is about 10 years, their future modernization will be required to be made without damaging the whole system operation. The way out has been found in using the hardware manufactured only at the series enterprises. These enterprises carry out the hardware modernization activities without any violation of the unified interface accepted in ARMS. The equipment manufacture is under supervision of the Federal Service for environmental, process and atomic surveillance (Rostechnadzor). It guarantees the high quality of the manufactured products.

4. Software and dataware

ARMS software has a modular structure and consists of self-contained modules thus enabling the system tasks composition to be modified and expanded.

The upper level software has the “client-server” architecture with the ARMS common database location at the server. The software has been developed as a multi-purpose client application which functions at AWP under control of the operational system Linux. Since the AWP get data independently of each other and work on the basis of the multi-purpose client application, they are functionally interchangeable thus contributing to the system viability.
The basis of the user’s interface development makes the perceptibility of the information by operators. The graphic interface is realized in the form of multimage application using modern approaches to the data presentation. ARMS makes it possible for the operator to monitor the general condition of all the systems covered by the radiation monitoring, to obtain the detailed data for specific parameters and sub-systems. As an example, Figures 2 to 4 provide the patterns of video frames.

5. Conclusion

ARMS presented in this paper is a new generation system developed for Kudankulam NPP taking into account the long experience of the similar systems creation for all Russian power units with reactors of WWER type. Due to the use of the modern measuring and computer equipment, application of the flexible two-level architecture, optimization of the monitoring scope and complex approach to the solution of all tasks of radiation monitoring at the NPP nowadays this system is one of the advanced products in the world in the field of the NPP radiation safety assurance.
ABSTRACT

Radioactive waste as arisen in nuclear power plants or similar installations is classified in different categories according to its radiological content and radiation emission rate. The components of the waste, which are small in mass but have a high activity inventory, are called Hot Spots. If Hot Spots can be removed more waste can be shift in a lower category resulting in a more economical waste management. The classification range comprehends from free release to high level waste which must be specially packed and stored in safe facilities for long time. The detection of Hot Spots requires specific instrumentation and methods which must be adapted to the radiation intensity of the Hot Spots and the speed of measurement. NUKEM Technologies GmbH has developed a series of new, economical monitoring systems for this purpose that are presented in this paper.

1. Introduction

Radioactive waste as arisen in nuclear power plants or similar installations is classified in different categories according to its radiological content and radiation emission rate. The classification range comprehends from free release to high level waste which must be specially packed and stored in safe facilities for long time.

For the radiological classification different limits are defined:

- Surface dose rates (i.e. dose rates at 100mm distance from the waste surface)
- Beta activity content (i.e. specific beta activity in units [Bq/g])
- Alpha activity content (i.e. specific alpha activity in units of [Bq/g])
- Isotope concentration with life time greater than 30 years

In general the cost for waste handling and storage is increased with increasing activity level.

Therefore, an advanced waste sorting and handling strategy should be used that considers economical aspects.

The components of the waste, which are small in mass but have a high activity inventory, are called Hot Spots.

If Hot Spots can be removed more waste can be shift in a lower category resulting in a more economical waste management.

Typical Hot Spots are debris from nuclear fuel elements, high activated metal pieces etc.
During the last decade NUKEM Technologies GmbH was highly engaged to realise nuclear waste treatment centres and has therefore developed and tested different methods and devices for this application.

In particular NUKEM Technologies GmbH has developed such kind of measurement systems for:

- Checking of excavated soil
- Sorting of solid waste
- Mapping the area of nuclear sites
- Checking of packages before leaving the nuclear plant

The measurement systems are optimised in respect to pick out small particles without the lost of the required throughput.

2. **Hot Spot detection for waste designated for release and/or landfill**

NUKEM Technologies has developed a Measurement System for Free Release Measurement (FMA) for waste from demolition of buildings and from ground. The waste is firstly scrapped to a size below 60mm and then filled in batches of 1000kg onto a conveyor belt. Above the belt four HPGe detectors are mounted. The detector spectra are analysed for each batch to measure the total activity of the waste. The throughput is up to 50 tons per hour. Details of the installation can be found in reference [1].

The online analysis of the total gamma spectra – especially in the case of very low gamma emission – is not suitable for Hot Spot detection. Therefore, the window analysis method is implemented: for each detector the regions of interest inside the spectrum are specified, which are readout in a short time sequence (typically 15 second). This procedure can be performed without interruption of the normal data collection for a complete spectrum during the passing of the waste batch.

The regions of interest are placed inside the gamma spectrum in such a way, that they cover the area for gammas from Co-60 and Cs-137. Some regions are placed in the neighbourhood of these lines where no gammas are expected and therefore, they can be used for background subtraction.

This method allows a time resolution of 1 second for a hot spot of 1000 Bq in a free release measurement. But in practice, the read out time has to be adapted to the intensity of the expected Hot Spot. The minimum time must be large enough to reliably measure a Hot Spot.

If a Hot Spot is detected, the conveyor is stopped, the Hot Spot is removed, the batch is transported back to the starting position and the measurement is started again. This procedure guarantees the success of the hot spot removing.

An alternative is to separate the complete batch with Hot Spot to a different place for further investigation. In this case a higher throughput is possible when many hot spots are expected. Normally, this special procedure is not needed.

3. **Hot Spot detection for middle and high active waste**

For the detection of Hot Spots in middle or high active waste – i.e. such waste type is managed in the waste treatment centre ICSRM in Chernobyl- NUKEM Technologies has developed a scanner system which has 3 CdZnTe gamma spectrometers, each mounted with its own collimator system. The complete device is moved by a crane over the sorting table. Each scanner scans a stripe of 1/3 width of the table.
The spectra are read out every second using the window technique as described in previous chapter. The system is able to detect Hot Spots and to associate these to specific isotopes for differentiation between activated or irradiated fuel material.

For better local resolution the gamma camera RAYMOS was added. [2]. The gamma camera takes pictures in the visual wavelength range, as well as, from gamma ray distribution affected by isotopes like Co-60 or Cs-137. After the measurement, both pictures are superimposed and delivered easily interpretable information about the location of Hot Spots.

The Fig. 1 shows such a picture which was taken during the tests of the camera. The source was a Cs-137 with intensity of $2 \times 10^8$ Bq, which could be detected even covered with 8cm of steel. In the example shown in Fig. 1 the source was covered by wood and plastic material.

![Fig. 1 Hot Spot detection by RAYMOS](image)

This type of RAYMOS has a relative low sensitivity: The picture presented in Fig.1 was taken from a 4m distance with an exposure time of 10 minutes. In this case a low sensitivity camera was used since it was applied for sorting of high level waste. The camera is based on the pin-hole principle. For application in the low radiation field NUKEM Technologies has developed a multi-aperture type of RAYMOS which has 256 pinholes instead of one single pinhole. This enhances the sensitivity by a factor 10. In addition a special background subtraction system was developed and integrated that further enhances the sensitivity.

4. **Hot Spot detection for large areas: the Groundhog™**

For mapping large areas outside of buildings, the monitor Groundhog™ [3] was developed (Fig. 2 and Fig. 3). It is a combination of a gamma measurement system and a GPS (global positioning system). The measurement data and the local position are stored in the database in real time. When the measurement is completed, an activity map of the terrain is available. Different types of gamma detectors can be used. The most advanced system is capable of guiding itself over large terrains.
5. **Hot Spot detection at drums**

This Hot Spot detection system has a different purpose: The drums are normally transported by containers. To transport the containers the regulations about the maximum allowed surface dose rate have to be fulfilled. If the drums have Hot Spot, i.e. the dose rate is above the allowed limit in some positions, since the hot spot can be placed near the surface the drum does not fulfil the requirements when it is stored in the container. When the location of the hot spot is known the loading procedure can take credit of this information to place the drum in the right position. A misleading can be avoided.

This method is implemented in our Drum Monitoring System DMS [4]. The drums are placed onto the turntable. From the side, as well as, from top and bottom dose rate meters equipped with rollers are pressed against the drum. In this way the distance between drum and dose rate meters is fixed. The dose rate meters are equipped with two Geiger Mueller counter tubes of different sensitivity. The dose rate meters are read out every second and the measured data are checked for Hot Spots. Once a Hot Spot is detected, colour is jetted on the drum to indicate the position of the Hot Spot (Fig. 4).

6. **Conclusion**

The detection of Hot Spots requires specific instrumentation and methods which must be adapted to the radiation intensity of the Hot Spots and the speed of measurement. NUKEM Technologies GmbH has developed a series of new, economical monitoring systems for this purpose that are presented in this paper.

**References**

Fig. 4 Drum monitor

1. Drum
2. Dose rate meters
3. HPGe detector
4. Hot Spot
DECOMMISSIONING OF AN URANIUM HEXAFLUORIDE PILOT PLANT

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ABSTRACT

The Institute of Nuclear and Energetic Researches has completed fifty years of operation, belongs to the National Commission for Nuclear Energy, it is situated inside the city of São Paulo. The IPEN-CNEN/SP is a Brazilian reference in the nuclear fuel cycle, researches in this field began in 1970, having dominancy in the cycle steps pilot scale from Yellow Cake to Uranium Hexafluoride technology. The plant of Uranium Hexafluoride produced 35 metric tonnes of this gas by year, had been closed in 1992, due to domain and total transference of know-how for industrial scale, demand of new facilities for the improvement of recent researches projects. The Institute initiates decommissioning in 2002. Then, the Uranium Hexafluoride pilot plant, no doubt the most important unit of the fuel cycle installed at IPEN-CNEN/SP, beginning decommissioning and dismantlement (D&D) in 2005. Such D&D strategies, planning, assessment and execution are described, presented and evaluated in this paper.

1. Introduction

Several groups of chemist and engineer at IPEN-CNEN/SP consolidated the developed researches in the facilities of the Uranium Conversion Project (PROCON), during more than twenty-five years, being responsible for the production of 35 metric tonnes of UF₆ by year, in the end of the 1980 decade. All PROCON plants comprised section from yellow cake to UF₆ production, such as the yellow cake dissolution operations, solvent extraction purification and the production of UO₃, its chemical reduction to and UO₂ followed by its fluoridation to UF₄, generation of the elementary fluorine for the fluorination of UF₄ to UF₆.

The UF₆ Production Pilot Plant began operation at IPEN-CNEN/SP in 1986 and stopped operation in 1992. It was projected and assembled for a nominal capacity of production of 20 kg×h⁻¹ of uranium, content in natural uranium hexafluoride, expressed as 27 kg×h⁻¹ of UF₄ or 30 kg×h⁻¹ of UF₆. Figures 1 and 2 depict some of the PROCON facilities.

![Fig 1. Pilot Plant Building](image1)
![Fig 2. Primary Crystallizer Inside Plant](image2)

During the operational period, the PROCON staff developed and improved components jointly with the national industry, and soon afterwards established know-how was been transferred to the company.
associated of the project. The country gained capability in the fuel cycle know-how, and trained more than two hundreds of technicians (engineers, physicists, technologists and others). The great majority of the trained professionals today apply their knowledge in the national nuclear establishments and in the private industry in a general way. Brazil has being recognized internationally as one of the few countries in the world with the complete domain of the nuclear fuel cycle (1,7). Being recognized internationally as one of the few countries in the world with the complete domain of the nuclear fuel cycle (1, 7). The figure 3 shows a flowchart of the Uranium Conversion Project (PROCON), that was operational at IPEN-CNEN/SP.

A search in the specialized literature pointed out several papers (2, 6, 8, 9, 10, 11, 13) covering the nuclear field decommissioning and dismantlement. Nevertheless there are not available open data concerning on decommissioning and dismantlement including nuclear material and dangerous chemical reagents, respectively uranium, hydrogen fluoride as that handled in an Uranium Hexafluoride production facility. The present study comprises the development of innovative methodology for decommissioning and dismantlement (D&D) of the Uranium Hexafluoride production facility. The present study comprises the development of an innovative methodology for decommissioning and dismantlement (D&D) of the Uranium Hexafluoride Production Pilot Plant, an integrating unit of the Uranium Conversion Project (PROCON), set up at IPEN-CNEN/SP (14).

2. Strategy
The decommissioning and dismantlement (D&D) of the UF₆ pilot plant comprised the Freon gas unit and the unit of Fluorine generation. This UF₆ plant remained out of operation for a range of 12 years (1993 to 2005). During this period all plant installations not received any maintenance. Several components and accessory of the plant had not received any maintenance. Several components and accessory of the plant were denied and relocated, impeding its operating again. Consequently, was impossible to discharge the residual gaseous UF₆, and any material contained inside of the connections and loading lines to crystallizers tank, making the dismantlement operation difficult and laborous. For this reason and because part of the equipments was exposed to the plant environment conditions, a detailed study was accomplished to verify the status of the equipment and to do an inspection in the UF₆ crystallizers and their internal parts. The possibility of finding Hydrogenfluoride (HF) condensed inside components of the fluorine generation section was evaluated as well.

As a consequence of the long time out of operation, all the trained operation and maintenance staff was moved from the UF₆ Plant to another tasks execution. Then, was necessary to contract an external company specialized in equipments dismantlement. However the contracted company has not inexperince in nuclear area. The alternative was to capacitate and assisted the employees of this company via an intensive course involving theoretical and practical lessons and training them in Radioprotection, handling of potentially dangerous products, (HF, UF₆) and also in the wearing of special personal protection equipments, such as total supplied air respirator and suits apparatus, as showed in the figure 9.

3. Decommissioning and Dismantlement Plan

3.1 Planning

The planning for the execution of these dismantlement tasks and decommissioning of the UF₆ Plant was submitted to a rigorous evaluation in the feeding pipes and discharge of UF₆, due to the possibility to find UF₆ (crystallized and gaseous) and residual UF₄. After the preliminary study, several blank tests were performed in the blind flanges set up with PTFE (polytetrafluorideethylen) parts. A series of tests was run for the inspection of the wash column (absorption). In the case of gaseous UF₆ lines, it was decided begin the dismounting operation starting with one of the secondary crystallizer, since the possibility of to find any UF₆ was remote.

3.2 Dismantlement

The dismantlement of the sections that comprise the UF₆ Pilot Plant (elemental fluorine and freon unit) conforms to the planning above described, except small alterations concerning handling of the hermetic flanges, with PTFE connections, mainly when inside the pipes or equipments crystallized UF₆ was found. As the chemical reaction for formation of UO₂F₂ is very fast, it was necessary to develop new techniques for replacement of each part of the lines, case to case. Figures 4 to 9 presents the steps relatives to dismantlement, transportation, storage and safety clothe used.

3.3 Decommissioning

The decommissioning of this pilot plant included specific procedures for Radioprotection (3, 4, 5), Safeguards (12), Environmental Monitoring (6) and Individual Protection (3, 4, 5, 6, 14). The dismantlement of the sections of elemental Fluorine and Freon gases were carried out according to the planning above described, except small alterations concerning on the handling of the hermetic flanges, with PTFE connections, mainly in the case that inside the pipes or equipments was found crystallized UF₆.
3.4 Transport and Storage of the Equipment

The choice and selection of an appropriate place for placement of the solitary equipments of the plants decommissioning it was very discerning, due to the dimensions of some equipments and the contained material, the proximity of the place of the plant also weighed a lot in the choice, there was the need of material remove of some deposits, to find the most appropriate place.

4. Conclusions

The UF6 Pilot Plant was successfully decommissioned and dismantled, according to initial planning, without serious adjustments. The reasons that contributed to this success were mainly planning, training, capability and technical attendance.

For the subcontracted company, the group training course was successfully, including theoretical and practical classes in Radioprotection, handling of Hydrogen fluoride, wear of special individual protection equipments (total supplied air respirator and suits apparatus with gas filters).

The decommissioning and dismantling of the UF₆ Pilot Plant was successfully accomplished with short financial costs, due to previous planning, direct accompanying and supervising of capable in charged group and personal responsibilities (14).
The decommissioning and dismantling of the first UF₆ Pilot Plant in the Brazil and probably in the world was a very important experience. The problems encountered in the course of decommissioning process allowed IPEN-CNEN/SP to find solutions, enabling their technicians to face future challenges not only in the others nuclear plants but also in NORM (natural occurring radioactive material) and conventional industries.

The works related to UF₆ Pilot Plant decommissioning and dismantling ended in 2006. All materials were decontaminated and stored.

5 References

4- COMISSÃO NACIONAL DE ENERGIA NUCLEAR. Diretrizes Básicas de Proteção Radiológica. CNEN, CNEN-NN-3.01 Norm, Rio de Janeiro, 2005 (In Portuguese).
In this work it is shown that the concentration of Pm-147 in spent fuel samples can be accurately determined by using liquid scintillation counting on a separated Pm fraction. Analyses were carried out on UO₂ and MOX samples. After dissolution of the fuel sample, Pm-147 has to be separated from the actinides, from the interfering fission products and from the other lanthanides. In this regard several separation methods were evaluated. The final separation was carried out by means of a column packed with Ln resin. To determine the obtained separation yields, an aliquot of irradiated Nd-146 spike is added to the fuel solution prior to separation. The progress of the separation can be monitored by measurement of gamma radiation of Pm-148m in several fractions. In this way separation yields of 90% could be obtained for all tested fuel samples.

1. Introduction

The objective of the MALIBU International Program was to provide improved knowledge of actinides and fission products in fuels irradiated at high burnups. This type of information is a necessity in many nuclear areas, such as, fundamental research, safeguard issues, the calibration of source term codes and waste management. This project proposed and managed by Belgonucléaire has been launched in June 2003 with the participation of 10 organisations, representing more than 20 different companies. A series of UO₂ and MOX samples, originating from both BWR and PWR reactors, were chosen to be analysed at SCK-CEN. Our group has already build up a lot of experience in the field of spent fuel analysis and burnup determination due to participation in several spent fuel projects [1, 4, 5].

After dissolution in the hotcell, the spent fuel samples were analysed by radiochemical and chemical methods covering an inventory of 51 different isotopes, among which Pm-147. A method was developed to determine Pm-147 in a separated Pm fraction using liquid scintillation counting. The separation of Pm-147 is necessary to eliminate all interferences of other β/γ emitters present in the sample during the LSC measurements.

2. Discussion

The spent fuel samples were dissolved in a hotcell following a two step procedure in which the samples are boiled under reflux in, respectively, 8M HNO₃ and 10M HNO₃ + 0.1M HF. The obtained solution is diluted until its activity is low enough to be brought out of the hot cell for the subsequent separations [2,3] and analyses.

Before starting the separations, the necessary spikes and tracers were added to the fuel solution. In first instance U and Pu are separated from the other isotopes. For this Pu is converted to Pu(IV) by applying a redox cycle. This solution is evaporated to dryness and redissolved in 9.0M HCl. The HCl 9.0 M solution, containing the dissolved fuel, is fed onto a Dowex 1x4 column preconditioned with HCl 9.0M. The fission products, the rare earth elements, Am and Cm are eluted with 9.0M HCl and collected for further treatment.
Consequently, the eluate fraction is evaporated to dryness and redissolved in 8.0M HNO₃. This solution is fed onto a mixed PbO₂-Dowex 1x4 column on which the tetravalent elements, e.g., Ce(IV) remain fixed. The trivalent rare earth elements and actinides, and most of the fission products are eluted with 8.0M HNO₃. From this fraction a 0.1M HCl solution is made by multiple evaporation and redissolving. Note that in each evaporation step, some H₂O₂ is added to help the breakdown of any organic material coming from the resin columns.

The 0.1M HCl sample is fed onto a di(2-ethylhexyl)orthophosphoric acid-kieselguhr column (Ln resin) from which the remaining Am and Cm can be eluted with a freshly prepared mixture of 2M lactic acid and 0.2M Na₅DTPA (Di-ethylenetriaminepentaacetic acid).

In a next step a method was developed to consecutively elute the lanthanides, and thus also prometium, from the Ln resin. Before applying this method to irradiated fuel samples, some experiments were conducted on test samples whereby HNO₃ or HCl gradients were used to elute the lanthanides. Usually HNO₃ is applied in traditional spent fuel separations, but for Pm-147 it was not really suitable. The eluate was collected in small aliquots of 2-5 ml that were analysed with ICP-MS. The Pm fractions were analysed with LSC. The results for the experiments in which a HCl gradient is used, are summarized in fig 1.

These results clearly show that when HCl is used, almost 100% of the Pm-147 is eluted from the column. If, on the other hand, HNO₃ is applied, the elution yield is below 50%. Moreover, it was not possible to get a well shaped elution peak. Therefore, it was decided to use HCl for the Pm elutions of spent fuel samples.

To determine the obtained separation yields, an aliquot of irradiated Nd-146 spike is added to the fuel solution prior to separation. This spike is irradiated in the BR2 reactor of SCK•CEN before use and contains low concentrations of Pm-148m, which is used as a tracer to track Pm-147. The progress of the separation can be monitored by measurement of gamma radiation of Pm-148m in the obtained fractions. In this way separation yields of about 90% could be obtained for all tested samples. The resulting Pm-148m elution curves of three spent fuel samples whereby this method was used, are presented in fig 2. The obtained separation yields do not seem to be influenced by the type of fuel (UO₂ or MOX) or the type of reactor (PWR or BWR). In all cases similar yields were produced.
Fig 2: Elution curves for Pm-148m obtained on three different spent fuel samples measured with gamma spectrometry

For the actual analysis of Pm-147 with LSC, several fractions are selected. These are mixed and from this new solution a sample, suitable for LSC, is prepared. The LSC analysis is done with a Packard Tri-Carb 1900CA detector which makes use of two photomultipliers to minimise the signal to noise ratio. For quantification a certified internal standard is applied. The measured concentrations are then recalculated to the analysis date and to the EOL (end-of-life) reference date, hereby taking into account the dissolution factors and separation yields. When the concentration values at EOL are compared, it becomes clear that for samples irradiated in the same type of reactor, the Pm-147 concentration (mg/g fuel) increases with an increasing burnup.

3. Conclusion

To measure Pm-147 in spent fuel samples, a suitable separation method was developed. By the use of HCl it was possible to separate Pm from the other lanthanides with a separation yield of about 90%. A few selected fractions were measured with LSC to obtain Pm-147 concentrations. It was found that samples irradiated in the same type of reactor, the Pm-147 concentration (mg/g fuel) increases with an increasing burnup.

4. References

Perception of management options for contaminated milk and implications for the decision-making process

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ABSTRACT

This paper analyses public acceptance of management options for radioactively contaminated milk and associated consumers' behaviour, focusing on the implications for the decision-making process. The results derive from a public opinion survey carried out in Belgium in 2006. The instrument used was Computer Assisted Personal Interviewing, complemented with a simulated news bulletin for a fast and realistic briefing on the situation investigated. Clean feeding of dairy cattle and disposal of contaminated milk are the preferred options in case of contaminations above legal norms. For contaminations below legal norms, normal consumption of milk seems better accepted than disposal. Nonetheless, the consumer’s behaviour reveals a precautionary tendency: the presence of radioactivity at some step in the food chain could lead to avoiding purchasing products from affected areas. Finally, public trust building appears as a key element.

1. Introduction

The management of a radiological food chain contamination requires a complex decision-making process involving multiple criteria and stakeholders. The aftermath of the Chernobyl accident (IAEA, 2006, pp. 83) proves that a robust and practicable restoration strategy should take into account not only radiological and feasibility criteria, but also the “acceptability of the countermeasures, ethical and environmental considerations, requirements for effective public communication, spatial variation and the contrasting needs of people in urban, rural and industrial environments”. The need to accommodate social concerns in the decision-making process (Otway, 1987; Allen et al, 1996) implicitly calls for a broad stakeholder involvement including the general public.

Depending on the characteristics of a radiological food chain contamination (geographical scale, type and quantities of radionuclides involved, etc), countermeasures (Nisbet et al, 2006) can be taken to reduce the health risk for the population, to bring social reassurance and to facilitate the return to normal life in the affected areas. However, little is known about the public acceptance of such countermeasures and one way to deal with the lack of information is by means of a survey. This paper reports on the results of a first survey on this topic. A couple of issues considered relevant for the decision-making process are addressed: i) public acceptance of various countermeasures; ii) consumer’s behaviour and its relation with countermeasures’ acceptance. Milk was chosen as case study since its continuous production requires urgent decisions due to the limited storage facilities. Moreover, dairy products are an important element of the diet, especially for children, who constitute the most radiosensitive population group. Finally, for certain radionuclides with high radiotoxicity such as radioceasium and radioiodine, maximal levels of activity concentration in milk are reached within few days after the ground deposition of radioactive material (Nisbet, 2002).

In the next section we introduce the problem studied and its particularities. In section 3 the methodology used is described, while in section 4 some results are discussed (see Turcanu et al, 2007, for an extensive analysis). Conclusions are drawn in the last section.
2. A hypothetical radioactive contamination of the food chain: problem framing

During recent years, the public confidence in food safety has declined due to multiple factors, e.g. the numerous food crises or the shortcomings in communication about food safety which is too often solely based on scientific risk assessments (Jensen & Sandoe, 2002). Frewer et al (2004, pp.1183) observe that public risk perceptions have often been dismissed as “irrational”, but these public concerns (and associated risk behaviours) “have direct consequences for human health, food safety and security, economic expansion, and international regulation”.

During a radiological emergency or in the restoration phase, authorities or food industries may decide on countermeasures for limiting the consequences of the contamination and maintaining or restoring public trust. Several food and agricultural countermeasures have been studied in the literature (Howard et al, 2005). Some of these target specific radionuclides, such as administration of feed additives, for instance, which largely reduces the transfer of radiocaesium to milk and meat. Countermeasures such as feeding dairy cows with uncontaminated fodder are effective against all radionuclides. Since the presence of radioactivity in food products is not an issue of concern for the Belgian public, the question is how are these countermeasures perceived by the public and to what degree are they accepted? From a legal viewpoint, after the Chernobyl accident in 1986 the European Union (CEC, 1989) laid down maximal radioactivity levels allowed for marketed food products in case of a future nuclear accident or any other radiological emergency. A radioactive contamination involves however a risk that would be new, involuntary and unknown to those exposed, with delayed effects and potentially affecting a large number of people. Due to a presumably high risk perception (cf. Slovic, 1987), the responsible authorities may decide to implement actions even for contaminations below legal norms. There are also situations, e.g. a large scale contamination, when consumption of specific products with contamination levels above the current legal norms could be envisaged. Paine (1992) reports a case related to the Chernobyl accident when the legal norms for radioactivity in reindeer meat were raised in Norway due to social and cultural reasons.

Finally, if countermeasures’ acceptance is important for the political decision-maker, the knowledge about consumer’s behaviour is essential in reducing the economic loss. Therefore we examine consumer’s behaviour for selected milk countermeasures with respect to a number of purchasing options, looking in particular if a high acceptance of a countermeasure is reflected by the intention of buying products originating from the area affected by contamination.

3. Methodological co-ordinates

Acceptance of management options and consumer’s behaviour after an accidental radiological contamination in the food chain were focal points of a public survey (Van Aeken et al, 2006) carried out in March-April 2006 among 1063 Belgian adults. Primary data were gathered through Computer Assisted Personal Interviewing, i.e. face-to-face interviews at the home of the respondent, the answers being directly stored on a portable computer’s hard disk. The selection of the persons interviewed is representative for Belgium for the following variables: province, region, level of urbanisation, gender, age (three categories) and professionally active status (retired or not). To better reproduce the real-life context, a video clip was shown to the respondents; this simulated a news bulletin reporting on an accidental release of radioactivity to the environment.

Two hypothetical situations were analysed: i) one causing contaminations above legal norms and ii) a less severe one, with contamination remaining below legal norms. In the first situation, the authorities are legally bound to implement countermeasures, since food products with contamination above legal norms may not be brought on the market. In the second situation, the main reason for taking countermeasures is public acceptance.

The milk countermeasures discussed were: keeping the dairy cows in stables and feeding them with uncontaminated feedstuff; reducing the radioactivity concentration in milk by administration of feed additives; processing of milk to dairy products having a low radioactivity retention factor; diluting contaminated milk with clean milk; disposing of contaminated milk; and slaughtering dairy cows in case of a long duration contamination. To these we added normal consumption of milk and decreasing contamination as much as possible.
4. Results and discussion
4.1 Acceptance of management options

For the situation when contamination of milk would exceed the legal norms (see Table 1), the countermeasures with highest acceptance are clean feeding and disposal of contaminated milk. Slaughtering dairy cows, which is an option for minimising the volume of contaminated milk produced in case of a long-lasting contamination, is also well accepted. These results confirm the increased sensitivity and intolerance of Belgian consumers (Vandecasteele et al., 2005), possibly leading to a strong pressure to dispose of any suspicious food product.

Administration of feed additives and processing of contaminated milk to butter or cheese are less accepted. This could be due to a fear that these countermeasures are less controllable, or to the fact that they leave a residual activity in milk, although being effective in removing the largest part.

<table>
<thead>
<tr>
<th>Management option</th>
<th>Degree of acceptance</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>strongly disagree</td>
</tr>
<tr>
<td>Clean Feed</td>
<td>7%</td>
</tr>
<tr>
<td>Disposal</td>
<td>9%</td>
</tr>
<tr>
<td>Slaughter</td>
<td>10%</td>
</tr>
<tr>
<td>Feed additives</td>
<td>15%</td>
</tr>
<tr>
<td>Processing to butter or cheese</td>
<td>19%</td>
</tr>
<tr>
<td>Dilution</td>
<td>47%</td>
</tr>
<tr>
<td>Normal consumption if experts ok</td>
<td>18%</td>
</tr>
</tbody>
</table>

Tab 1: Public acceptance of milk management options (N=1063 respondents)

That people prefer to take legal norms as a reference point is apparent from the low acceptance of normal consumption of milk with contamination above norms, even if experts declared it safe for consumption. Dilution is strongly opposed by an overwhelming majority of respondents, possibly due to a feeling that dilution would increase the scale of the problem, rather than reduce it.

For the situation when contamination of milk is expected to remain below the legal norms, clean feeding as a measure of preventing milk contamination enjoys again a high acceptance among the respondents. More than half or the respondents also agree with the idea that products complying with the legal norms can be consumed as usually, but the vast majority is in favour of reducing contamination as much as possible, even when already below the legal norms. Although one third of the respondents agree with disposal of all contaminated milk, the general acceptability of this radical countermeasure decreases compared to the previous situation: almost half of the respondents are actually against it. Processing of milk and administration of feed additives are again less accepted, while dilution is mostly considered unacceptable.

For a practical approach to geographical zoning of the affected area in view of applying certain countermeasures, it is useful to know if the countermeasures’ acceptance depends on whether the expected contamination is above or below legal norms. Acceptance of normal consumption and of disposal differs significantly in the two hypothetical situations considered.

This is however not the case for clean feeding and milk processing (see Table 1). For these two countermeasures, acceptance seems more related to the countermeasure itself than to the circumstances of its application. This conclusion is supported by a marginal homogeneity test which indicates that there is no significant difference between the associated acceptance distributions (significance level 0.01). For administration of feed additives, the difference between the corresponding values in the two situations considered is, for any agreement degree, less than 3%; therefore also for this countermeasure the acceptance is similar. In case of dilution, given the very
small agreement rate with this option (<20%), the difference in acceptance between the two situations analysed can be considered negligible as well.

From a decision-making point of view this implies that the geographical extent of the latter countermeasures may be larger than the area in which the contamination of milk is expected to be above legal norms. The inherent exceptions are normal consumption of contaminated products (confirmed as “safe” by legal norms or experts in the field) and disposal.

4.2. Consumer’s behaviour

Consumer’s behaviour was studied on the background of five possible contexts by presenting to the respondents the following purchasing options: i) stop buying milk or dairy products; ii) buy imported products; iii) decide if to buy local or imported -“clean”- products depending on their price; iv) buy local products.

Confirming the high acceptance expressed for clean feeding, 67% of the 984 respondents consuming dairy products at least once a month state that they would continue buying the local products (see Fig. 1), if this countermeasure were implemented. A significantly smaller number of respondents (36%) would buy the local products if feed additives were administered to dairy cows, which is in agreement with the smaller acceptance observed for this countermeasure.

A contamination below legal norms results in more than half of the respondents deciding to buy imported or stop buying dairy products. The expressed consumer behaviour is here in contradiction with the relatively high acceptance expressed for normal consumption of such products. From the 587 respondents consuming dairy products at least once a month and agreeing or strongly agreeing that it is acceptable to consume products with contamination below legal norms, only 43% would buy local products, while 30% would buy imported and 16% would stop buying dairy products. Dairy products with contamination above legal norms are refused by two thirds of the population even when declared safe by experts. The same is valid for dairy products with contamination below the legal norms, but processed from raw milk with contamination above legal norms. It is worthwhile reminding here (Henson, 1996) that “safer” food products may be refused due to peoples concern on how these have been treated or tested. A good communication on such policy will therefore be an essential factor.

While price is generally considered an important food attribute to the Belgian consumer (AFSCA, 2004), it appears inconsequential when considering a radioactive food contamination. Less than 10% of the respondents would decide between local or imported products depending on their price, for any of the management options considered.

5. Conclusions

Our study has focused on public acceptance of countermeasures for radioactively contaminated milk and associated consumer’s behaviour. The aim of the research was to acquire information that can be used to support not only the real-life decision-making process in case such a contamination would occur, but also the activities related to emergency preparedness.
From a public acceptance viewpoint, the preferred countermeasures are clean feeding and disposal, in case of contaminations above legal norms. For contaminations below legal norms, clean feeding and normal consumption are both well accepted. However, the results show that if normal consumption of milk is the chosen policy for contaminations below legal norms, a serious market loss could be expected. Public acceptance proved to be influenced by the initial contamination level mostly for the normal consumption and the disposal countermeasures. For clean feeding and milk processing, and to a lesser extent for feed additives and dilution, public acceptance seems to be an intrinsic characteristic. From a decision-making perspective this suggests that application of such countermeasures may be on larger areas than those where contamination of milk is expected to exceed legal norms.

Looking from the perspective of both public acceptance and consumer's behaviour, it appears that a majority of people favour a precautionary policy, aiming at preventing any contamination in the food chain. This upholds options like clean feeding and disposal of contaminated foodstuff. Administration of feed additives is an alternative, but this would presumably lead to a significant decline in the consumption of products from the affected areas. Milk processing would have to count on a limited market segment, whereas dilution of milk is strongly opposed in any circumstance.

Despite the inherent uncertainties, we consider that our study is useful for emergency planning purposes, especially for situations when there is a time constraint. It can also improve communication on the countermeasures to be implemented and may contribute to an increased public acceptance and efficiency of the decision-making process.

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ABSTRACT

The developed countries only live for years an unprecedented period of well-being. This means easy access to energy sources to keep the standard of living. Nevertheless when we watch in the news the consequences of some uncontrollable meteorological events or terrorist acts we realize that our society is not invulnerable. Additionally in the coming years the facilities producing raw materials, components and final assemblies will likely be located farther away one from each other and from the final customer, posing a major influence in the complexity of the supply chains. All this is becoming an issue in the energy market where the security of supply is of bigger concern especially for the countries with higher energy demand. This background place a challenge in the national administrations and utilities to secure energy and fuel supply from all different sources including nuclear.

As part of the effort to secure nuclear fuel availability both utilities and nuclear fuel vendors are merging forces to build a sustainable supply chain that can provide a reliable nuclear fuel in case of a potential disruption.

Wolf Creek Nuclear Operating Corporation (WCNOC) and Almaraz NPP (Spain) together with Westinghouse and ENUSA Industrias Avanzadas (Spain) have entered into a security of supply agreement aimed at demonstrating the feasibility of fuel supply to a US nuclear power plant from the ENUSA Juzbado fuel factory as well as to a Spanish power plant from the Westinghouse Columbia fuel factory.

This paper describes the project goals, challenges, planning and expected results. At the end a set of conclusions are presented despite the fact that the project is currently being executed.
Introduction

Europe, North America and in general the developed countries only live for years an unprecedented period of well-being. This means easy access to energy sources that allow to work to our appliances and automobiles and to keep on moving our public transportations and industries. For the developed world it seems inconceivable to reduce our standards of living and to give up something that we give for fact.

It is this belief the one that questions when we attend the news of some uncontrollable meteorological events (hurricane Katrina) or we witness with incredulity to the collapse of the twin towers of New York for a terrorist act. No we are invulnerable neither to the terrorism nor the nature, neither although to a lesser scale to the politics and the regulators..

Additionally in the coming years the facilities producing raw materials, components and final assemblies will likely be located farther away one from each other and from the final customer, posing a major influence in the complexity of the supply chains.

All these facts have stimulated the energy producers to revise their supply chains and to attempt protecting the weaker links. One of the initiatives to get it is to guarantee an alternative nuclear fuel supply in case of a potential disruption.

That is the exercise that Wolf Creek Nuclear Operating Corporation (WCNOC) and Almaraz NPP (Spain) together with Westinghouse and ENUSA Industrias Avanzadas (Spain) ENUSA, have outlined.

These four parties have entered into a security of supply agreement aimed at demonstrating the feasibility of fuel supply to a US nuclear power plant from the ENUSA Juzbado fuel factory as well as to a Spanish power plant from the Westinghouse Columbia fuel factory (Fig. 1).

Fig. 1. The logistics of the program at a glance

From the formal point of view the project is splitted in two subprograms

On the one hand, ENUSA will manufacture for Westinghouse four Fuel Assemblies (F/As) for one of the reloads that W has to deliver to WCNOC. This is known as American subprogram.
On the other hand, Westinghouse will manufacture for ENUSA four F/As for one of the reloads that ENUSA has to deliver to Almaraz NPP. This is known as Spanish subprogram.

The primary goal of this project is to demonstrate the reliability of an overseas manufacturer to supply nuclear fuel for a PWR Nuclear Power Plant in the case of a potential supply disruption affecting the primary fuel supplier. Such supply must meet not only technical but also licensing, logistics and timing requirements which must therefore be considered in an integral manner in the project.

As an aside objective, Westinghouse and ENUSA shall undertake a cross-qualification of their respective manufacturing facilities, by means of an auditing process, as part of which not only the product but also manufacturing process and inspection techniques shall be compared.

Partnership is one of the key words in this project. The relationship of Westinghouse and ENUSA of more than three decades allows the parties to approach the exercise outlined above with trust and in an atmosphere of collaboration.

**Fuel Designs**

It is logical that the designs of the fuel assemblies to be manufactured for the utilities should be equivalent to the resident fuel. Also one of the assumptions in the project is for each manufacturer to minimize the number of modifications to their respective fuel product lines needed to allow insertion in the plants (Figs. 2 & 3).

In the American subprogram the reference design that Wolf Creek NPP loads is 17 RFA (z+2) featuring ZIRLO™ tubing, Intermediate Flow Mixers (IFM), RFA-2 mid grids, protective grid and Integrated Upper Nozzle (WIN). The assemblies that ENUSA will manufacture have the following particularities:
- Bottom Nozzle fabricated in Spain by a domestic supplier (ENSAs)
- UO2 rods with neither annular pellets nor integrated poisons and with end plugs manufactured by ENUSA

In the Spanish subprogram the reference fuel assembly design that Almaraz NPP loads it is very similar to the RFA design although with the specific designation of 17 MAEF. The assemblies that Westinghouse will manufacture have the following particularities:
- Westinghouse Integrated Top Nozzle (WIN)
- Bottom Nozzle supplied by W (now it is manufactured by ENSA)
- Gad rods manufactured in Västeras with Sandvik ZIRLO™ tubing and end plugs manufactured by Westinghouse.
Fig. 2. Main design differences vs. the reference design in the Spanish domestic PWR product

**MAIN DESIGN DIFFERENCES**

**SPANISH SUBPROGRAM**

**DESIGN OF RECORD**

**SPANISH DOMESTIC PWR PRODUCT**

- **Integrated Top Nozzle (WIN)**
- **Gadolinia rods manufactured at Värstervästan**
- **Bottom Nozzle supplied by WH**
- **End plugs manufactured at Cetara**
- **ZIRLO tube from SAFDVIK (only Gad rods)**

Fig. 3. Main design differences vs. the reference design in the US domestic PWR product

**MAIN DESIGN DIFFERENCES**

**AMERICAN SUBPROGRAM**

**DESIGN OF RECORD**

**TFRA [242]**

- **UO2 rods without annular poison or integrated poison**
- **End plugs manufactured by ENSA**
- **Bottom nozzle supplied by ENSA**

**DESIGN OF RECORD**

**TFRA [242]**
The fuel designs from Westinghouse and ENUSA, which are to be supplied respectively to Almaraz and WCNOC, have been analyzed by both fuel fabricators to assure compatibility with the resident fuel in the core. The analysis includes not only the core physics and mechanical design but also the manufacturing and quality processes used during fabrication of the fuel.

As stated above, most of the components shall be fabricated by Westinghouse, the main component supplier to ENUSA. However, the Lower Nozzles for the Wolf Creek fuel assemblies shall be manufactured by ENSA at its factory in the North of Spain. Pressure drop tests have been performed to demonstrate the feasibility of using this component at Wolf Creek.

For the Almaraz fuel, a specific modification has been required by design to incorporate Gadolinium as burnable absorber. The Gadolinium fuel rods shall be fabricated at the Westinghouse fuel facility at Västerås (Sweden) and delivered to the Columbia factory for the final fuel assembling. A specific fuel rod design study shall be carried out to allow the insertion of those Gadolinia rods.

For the Wolf Creek fuel, the core was specifically designed to not require burnable absorbers in the fuel rods provided by ENUSA. Wolf Creek normally uses the Integral Fuel Burnable Absorber (IFBA), a boron coating on the fuel pellet, which is not available from ENUSA. This design change was possible for the delivery of four fuel assemblies; however, a full reload would require the use of gadolinia, or discrete burnable absorbers.

**Licensing**

Due to the similarity of both product lines a relatively smooth licensing effort is expected. Key points on this process shall be the different origins and characteristics of the UO2 powder. ENUSA utilizes an IDR process with powder delivered from the Springfields facility (UK) while Westinghouse uses an ADU route. Additionally, Västerås AUC process shall be used in the Gadolinia rods for the Almaraz fuel.

Features like the WIN nozzle are to be first delivered into the Almaraz plant.

**Logistics**

This Security of Supply project involves three nuclear fuel manufacturing facilities and two nuclear power plants located at both sides of the Atlantic. Therefore, an extensive effort must be put in place to guarantee the smooth delivery of fuel to the different facilities. The transatlantic transportation will be carried out through shipping lines with previous experience in transport of nuclear materials.

For both deliveries the new Westinghouse Traveller container shall be used, although for the Wolf Creek assemblies an additional operation at the Columbia factory must be performed to load the fuel into MCC containers for later delivery to Wolf Creek.

The Gadolinia rods for the Almaraz fuel shall be shipped following an existing route and procedure at Westinghouse for shipment of Gadolinia rods for the US market. The ZIRLO™ tubes shall be manufactured by Sandvik Europe.

The fissile materials will be accounted for as a balance of the Uranium through book transfer among the accounts that as much ENUSA as Westinghouse have in SFL (UK).
Conclusions

Although the project is currently ongoing some of the expected results are anticipated:

- The project will show to the clients of ENUSA and Westinghouse that in the event of a disruption the alternate fuel supply from foreign manufacturing facilities is a feasible back up from the logistics, design and licensing standpoints.
- The exercise will demonstrate that the designs that ENUSA and Westinghouse are able to manufacture and deliver with their current supply chains are equivalent and do not need either further qualification or additional licensing.
- The program will allow exploring new routes for supply of UO2 powder and components to eventually improve the current ones and to open alternatives before the foregone increase of the demand in coming years.
- The security of supply program intends to get the above-mentioned goals with the minimum impact in the licensing process before the regulatory authorities. The objective is to get the authorization of fuel loading without necessity of revising the licensed safety analyses. This is a major advantage versus qualification of an alternate vendor where new safety analyses are normally required, nearly prohibiting any benefit in an emergency situation.

Both Westinghouse and ENUSA consider that this project is a key step in their long history of mutual partnership which shall tighten even more their historical links to better serve their customers in Europe and the US.
TECK-IN :
Skills Center for Containment Techniques and Working Methods

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1. Introduction: competitiveness clusters

In 2005, France launched a new industrial policy mobilizing the key factors for competitiveness, one of the most important of which is the capacity for innovation. A competitiveness cluster is a group of companies, research centers and training organizations in a given area, all working together as partners and pooling their actions under a shared development strategy. The cluster is designed to harness synergies and encourage close cooperation on innovative projects. The French government has decided to officially approve and support 67 competitiveness clusters. The TRIMATEC competitiveness cluster (Tricastin Marcoule Technologies) covers six R&D zones and includes a central zone covering Tricastin and the east of the Gard department. These zones bring together all the major research and development organizations. TRIMATEC received the government’s seal of approval in 2005 and aims to produce projects that are essentially based on technologies developed by the nuclear industry but which can now be applied to other sectors. The cluster is located over five departments (Ardèche, Drôme, Gard, Hérault and Vaucluse).

2. The project initiated by MELOX and its partners

Numerous industrial processes require the use of containments in fields as varied as the pharmaceutical industry, medicine, microelectronics or the nuclear industry. The AREVA group, world leader in the nuclear industry, has over 30 years’ experience in this field. The MELOX plant is at the forefront since it is here that plutonium is used to fabricate fuel assemblies.

When the plant was designed, and indeed ever since it has been in operation, there have been numerous initiatives to improve the tools, methods and techniques used when working in glove boxes. Likewise, a variety of training sessions are held regularly to maintain a high level of expertise in this field.

It was therefore decided to set up a Skills Center for Containment Techniques and Working Methods, with the participation and involvement of several partner companies whose activities necessitate this kind of work. The project was approved by the TRIMATEC competitiveness cluster on November 23, 2006 and officially launched at a kick-off meeting held at MELOX with the various partners on January 19, 2007.

Current partners
Companies: MAPA Advantech, Piercan, La Calhene, Sovis (St Gobain), Plastunion, STMI, Robotic Concept, Axilys, Trihom, Sinaptec, and Novintec.
Training, teaching and research organizations: École des Mines d’Alès, INSTN Valrhô, and CEA Valrhô.
3. Creation of TEKH-IN and objectives of the association

- **Structure**

The Skills Center for Containment Techniques and Working Methods was formed as a non-profit organization on June 15, 2007. Its aim is to pool, develop, improve and spread the competencies, techniques, methods and tools used when working in containments. The project aims to create a consistent set of academic and industrial competencies to provide a center specializing in containment work and focusing on three main areas:

- research and development,
- professional training,
- consulting and expertise.

Pooling skills makes it possible to launch, finance and steer actions with the joint aim of perpetuating a high level of technicity in the target field. This is an innovative project in that it brings into contact organizations that would normally have very few dealings with each other, such as the nuclear industry, the pharmaceutical industry, the food processing industry, depollution, asbestos removal and other activities confronted with the problems associated with work in glove boxes. It will encourage performance improvement as regards worker training and system operation in containments, particularly concerning nuclear and occupational safety and the environment in an industrial setting.

- **Objective**

More precisely, the aim is to:

- develop special tools, in situ work processes and technical materials to offer workers improved ergonomics, and greater occupational safety and precision,
- devise and dispense training for those working in an industrial environment or enrolled in a specialized university course,
- provide consulting services and expertise to support industrialists.

A market opportunity study is currently being launched. It will provide information on the major containment work improvement trends, and the projected requirements of companies in a number of sectors.

4. Resources and partnerships

The center will have its own resources to:

- carry out the tests required before any new techniques and tools are implemented on an industrial scale,
- provide training in containment work, notably the use of special tools and techniques.

And resources provided by each partner to:

- design, study and produce new techniques and tools,
- carry out standardized tests prior to qualification,
- design appropriate training courses,
- carry out research work to acquire the basic knowledge that supports applied developments.

5. On-going actions

**Academic training**

The aim is to create "relevant" partnerships with schools (technical schools, engineering schools, vocational schools, etc.) with a view to transferring knowledge and competencies within the framework of a diploma course. The center’s contribution should be incorporated under certain conditions:
- participation in diplomas leading to true job opportunities.
- consistency between our areas of competence and the general content of the course work.

This initiative has many benefits. It will lead to Department of Education approval and the high-quality contribution of the center will be acknowledged.

**Research and Development**

The following topics have been identified so far:
- introducing equipment into containments
- removing equipment from containments
- very long-lived HEPA filter
- self-standing micro vacuums
- dispensing with the need for gloves

The above R&D projects will be the subject of special agreements binding the TEKH-IN members interested in developing the subject. New partners are currently being sought for these topics.

**Competency mapping**

The current and potential know-how of the project partners will be listed to produce a map of the competencies required to improve containment work.

The following macro-competencies have been identified to date:

- **Design/engineering**: integration of the operating and maintenance constraints specific to containments

- **Ventilation, filtering and purification systems**

- **Transparent materials**: providing visibility and an integrated barrier

- **Special gloves**: isolating the operator from the product without hindering the manual dexterity required for operations

- **Lighting**: external facility generating sufficient intensity without reflecting or dazzling
- Monitoring systems measuring the quality of the environment (special sensors to detect dust, radioactivity, etc.)

- Leak detection methods

- Operating and maintenance know-how: focusing on preserving containment integrity and creating the interface between the operator and the hostile environment
  - workstation organization and ergonomics
  - operating technique with appropriate gestures
  - risk awareness
  - self-control
  - questioning attitude

- In situ waste management: special tools and techniques for waste removal and packaging

- Dismantling operations in hostile environments: coordinating the various trades and making allowance for the constraints associated with containments

- Operating, maintenance and investigation techniques in containments: endoscopes, vacuums, airlocks, personal and collective protection devices

- Specially-adapted tools: conventional mechanical tools subjected to the constraints of containments and modified accordingly

- Design and production of protective clothing for work in hostile environments
ADVANCED SOLUTIONS FOR MITIGATING HYDROGEN RISK DURING TRANSPORTATION OF RADIOACTIVE MATERIALS

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ABSTRACT

During transportation of radioactive materials, radiolysis or thermal degradation of the transported product can lead to the generation of hydrogen, which continuously enriches the gaseous mixture. Among the functions to be satisfied, transportation systems shall thus allow the control of the hydrogen content below its flammability limit. This can be achieved by limiting the transportation duration so as to reopen the cask before the critical hydrogen concentration is reached. Development of new technologies that would mitigate the hydrogen risk is all the more motivated because it would allow an extension of the transportation duration.

AREVA-TN International has developed catalytic systems which aim at buffering the hydrogen concentration far below the flammability limit. The principle of these catalysts is to recombine the hydrogen with the oxygen contained in the gaseous mixture.

Hydrogen generation is either due to radiolysis of water or organic compounds. The latter are usually transported in the form of solid waste, whereas water is either in the form of residual water is the case of a dry transportation or of free water in the case of a wet transportation. AREVA-TN International has thus developed two types of catalytic systems.

The first is dedicated to the recombination of hydrogen coming from radiolysis of organic compounds and residual water. It allows a significant recombination even in the presence of other gaseous species generated by radiolysis of organic compounds. This catalytic system also absorbs the water generated by the recombination reaction, preventing from subsequent radiolysis of this water.

The second catalytic system is specifically dedicated to wet conditions of transport and is particularly adapted to these conditions including successive immersions into borated water.

The present paper gives an overview of both catalytic recombining systems. It describes the laboratory tests undertaken for the evaluation of the recombining efficiencies.

The catalyst dedicated to dry transportation systems was especially studied in the presence of several gases that may be generated by radiolysis of organic compounds, i.e. CO, CO$_2$, CH$_4$, C$_2$H$_6$, C$_2$H$_4$, HCl, I$_2$. The water absorption capacity of the catalyst has also been determined.

In case of wet transportation, particular attention is placed on the recombining efficiency after immersion of the catalysts in borated water, which would occur in a nuclear reactor pool during loading of used fuel. Laboratory investigations, carried out in an autoclave simulating a wet transportation cask, showed that, after immersion in borated water, the catalytic system allows the recombination of 3% hydrogen in less than 24 hours at temperatures as low as 35°C.

INTRODUCTION

Catalytic systems have already been developed for mitigating hydrogen risk in case of a severe accident in a nuclear power plant <1>. These catalysts were based on the use of a mixture of platinum and palladium deposited on a support (for example alumina). This catalytic system enhances the oxidation of hydrogen with respect to the following reaction:
It also allows the recombination of about 1 µmol of hydrogen per second and per gram of catalyst at 90°C.

As a matter of fact, the applicability of this technology to transportation of radioactive materials requires the system to be acceptable at lower temperatures, down to about 50 °C. In addition, the catalytic support for the platinum and palladium elements shall in some cases withstand the humid environment of wet transportation. In case of dry transportation, alumina support becomes an advantage since it allows the water produced by the recombination reaction to be absorbed. The latter prevents from further radiolysis and hydrogen generation.

Consequently, AREVA-TN International developed two types of catalytic systems based on the deposition of palladium and platinum on a stainless steel grid or on aluminium. At first, the present paper shows the qualification results with respect to laboratory tests carried out with the stainless steel recombiner, which we dedicate to wet transportation. Secondly, details are given on the use of an alumina recombiner for dry transportation.

1. QUALIFICATION OF A HYDROGEN RECOMBINER FOR WET TRANSPORTATION

1.1 Experimental protocol of the qualification campaign

The qualification protocol for the catalytic grid consisted in 15 immersion/recombination cycles simulating 15 loading/transport cycles. The experimental set-up consisted in a reactor combined with gas injections and a sampling device dedicated to the chemical analysis of the gas. For laboratory tests, a cycle corresponds to one week of testing defined as follows:

1. **2 days immersion of the catalytic stainless steel grid into borated water containing 2662 mg/l of boron.**
2. **setting up a gaseous mixture of N₂-O₂ containing 3 to 7 vol% oxygen.**
3. **homogenization of the reactor temperature at 40 °C.**
4. **injection of 3 vol % hydrogen.**
5. **measurement of the evolution of the hydrogen content as a function of time.**
6. **as soon as the hydrogen content falls below 1%, an additional injection is performed in order to readjust the hydrogen concentration to 3%.**
7. **the cycle is stopped after 5 days (1 week). The catalyst is then immersed into borated water for 2 days (week-end).**

1.2 Qualification results

Appendix 1 shows the evolution of the hydrogen and oxygen concentrations during the 15 immersion/recombination cycles.

The first 4 cycles were carried out with initial oxygen content of 6 to 7 vol% whereas for the last 11 cycles, this concentration was comprised between 3 and 4 vol%. It appears that this initial oxygen content has a direct influence on the amount of hydrogen injection during the corresponding cycle. With 7 vol% O₂ initially, 4 hydrogen injections are performed on a weekly basis. With 3 or 4 % O₂, only 3 injections are possible per week. Indeed, with an initial concentration of 3 to 4 % O₂, the catalytic recombination leads to a drop of the oxygen content to concentrations below 1% after about 2 days. The oxygen supply is then too small to allow the catalytic recombination with respect to the chemical reaction presented in paragraph 3.1. Consequently, for the last 11 cycles, the hydrogen content remains stable during the last few hours that follow the last hydrogen injection. Nevertheless, appendix 1 shows that the recombination capacity remains similar for the whole 15 cycles.
1.3 Kinetics of the recombination reaction

It can be assumed that the \( \text{H}_2 \) recombination rate in \( \text{cm}^3/\text{h}/\text{cm}^2 \) of catalytic grid can be written as follows:

\[
V_{\text{recomb}} = A_0 [\text{H}_2]^\alpha
\]

Eq. 1

where:
- \( V_{\text{recomb}} \) designates the \( \text{H}_2 \) recombination rate in \( \text{cm}^3/\text{h}/\text{cm}^2 \) of catalytic grid,
- \( A_0 \) and \( \alpha \) (the order of the oxidation reaction with respect to the hydrogen content) are the couple of kinetic constants for each test and
- \([\text{H}_2]\) is the hydrogen concentration in volume %.

Based on the measured evolution of hydrogen concentration in the course of time (Appendix 1), the recombination rate was calculated for each of the 15 qualification tests performed in \( \text{N}_2-4\%\text{O}_2 \) at \( 40^\circ\text{C} \). Figure 1 illustrates these recombination rates \( V_{\text{recomb}} \) for the two tests which show the slowest recombination reaction.

With respect to Figure 1, a conservative kinetic law for hydrogen recombination can be written as follows:

\[
V_{\text{recomb}} = 0.014 [\text{H}_2]^{1.5}
\]

Eq. 2

Figure 1: Evolution of the \( \text{H}_2 \) recombination rate per unit surface of the catalytic grid (in \( \text{cm}^3/\text{h}/\text{cm}^2 \)) as a function of the \( \text{H}_2 \) concentration in the reactor at \( 40^\circ\text{C} \).

For design calculations the kinetic law Eq. 2 of an order of 1.5 with respect to the hydrogen content will be considered for the oxidation of hydrogen.

1.4 Applicability to the transportation of used fuel

Hydrogen generation due to radiolysis is usually given by a linear expression of the volume \( V_{\text{prod}} \) of hydrogen produced by unit time <2>. \( V_{\text{prod}} \) is thus given in \( \text{cm}^3 \) of \( \text{H}_2/\text{h} \).
Consequently, in order to recombine the total quantity of hydrogen generated by radiolysis, the minimum surface $S_{\text{min}}$ of catalyst required is given by (in cm$^2$):

$$S_{\text{min}} = \frac{V_{\text{prod}}}{V_{\text{recomb}}} = \frac{V_{\text{prod}}}{0.014[H_2]^1.5}$$

Eq. 3

As for equation Eq. 2, the hydrogen concentration in Eq. 3 is expressed in volume percentage.

An hydrogen concentration of 1% is acceptable with respect to safety since it is far below the flammability limit of 4% hydrogen in air. Therefore, it is conservative to consider that the minimum surface of catalytic grid to be put in place is given by Eq. 3 with 1 vol. % hydrogen.

As an example, R62 is a wet transportation package for used fuel presented at Patram 1986 <3>. The results of measurement of hydrogen concentrations within the package showed a hydrogen generation rate below $V_{\text{prod}}=3.5$ cm$^3$/h. This means that for the particular design of R62, a total surface of 250 cm$^2$ of the catalytic grid would be enough for maintaining the hydrogen concentration below 1%. It appears that such an amount of recombiner is acceptable for an application in usual transportation casks, as it should be easy to insert this low amount of catalyst within the cavity. Even higher amounts of recombiner should be easy to put in place in order to allow a safety margin with regards to recombination capacity.

Furthermore, with a higher amount of catalytic recombiner within the transportation cask, the hydrogen risk can be mitigated for a higher level of water radiolysis, i.e. for larger casks or for used fuels with higher radiation energies.

2. QUALIFICATION OF A HYDROGEN RECOMBINER FOR DRY TRANSPORTATION

The recombiner dedicated to dry transportation is made of alumina beads impregnated with palladium. Some characteristics are given in the following table:

<table>
<thead>
<tr>
<th>Diameter of the beads</th>
<th>Specific surface</th>
<th>Density</th>
</tr>
</thead>
<tbody>
<tr>
<td>3 mm</td>
<td>300 m$^2$/gr</td>
<td>800 kg/m$^3$</td>
</tr>
</tbody>
</table>

The aim of the qualification tests was to:

a) Assess the hydrogen recombination capacity as well as the water absorption capacity of the hydrogen recombiner at different temperatures;

b) Determine the recombination kinetics at different temperatures;

c) Assess the influence of other radiolysis gases on the H$_2$ recombination capacity.

2.1 Hydrogen recombination capacity

The hydrogen recombination capacity has been determined by laboratory tests at 25°C, 45°C and 65°C.

The catalytic beads were previously dried at 80°C within the reactor. Tests were carried out in a 5 L reactor, which was linked to an argon-hydrogen supply. The latter allows the hydrogen content to be maintained at about 2%. Hygrometry is measured all along the testing duration. The composition of the gas is periodically analyzed using gas chromatography.

The recombination capacity (given in moles of H$_2$/kg of catalyst) is measured from the total quantity of oxygen consumed (2 moles of H$_2$ for 1 mol of O$_2$).

The results are shown in the following table:

<table>
<thead>
<tr>
<th>Recombination</th>
<th>25°C</th>
<th>45°C</th>
<th>65°C</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>18,5</td>
<td>23</td>
<td>&gt; 40</td>
</tr>
</tbody>
</table>
2.2 Water absorption capacity of the alumina beads

For the three tests mentioned previously, the hygrometry was measured above 90%. The water formed by the recombination reaction is thus either absorbed by the alumina from the recombiner or contained in the gas phase. The ratio between the water content in the alumina and in the gas phase depends on the temperature, i.e. the higher the temperature, the lower the amount of water absorbed by the catalytic support, thus the higher the water content in the gas phase. The absorption capacities are given below.

<table>
<thead>
<tr>
<th>Testing temperature</th>
<th>Ratio of the mass of water absorbed by the catalyst with respect to the mass of catalyst</th>
<th>Amount of water produced by the recombination and trapped in the catalyst</th>
<th>Amount of water produced by the recombination and released by the gas phase</th>
</tr>
</thead>
<tbody>
<tr>
<td>25°C</td>
<td>32%</td>
<td>90%</td>
<td>10%</td>
</tr>
<tr>
<td>45°C</td>
<td>31%</td>
<td>80%</td>
<td>20%</td>
</tr>
<tr>
<td>65°C</td>
<td>17%</td>
<td>45%</td>
<td>55%</td>
</tr>
</tbody>
</table>

2.3 Hydrogen recombination kinetics of the recombiner

The recombination kinetics has been studied at different temperatures, i.e. 28°C, 45°C, 55°C and 65°C. For each test, a mass of 6.25 g of catalyst was previously dried at 80°C. The testing reactor had a capacity of 5 L and the starting hydrogen content was about 4% hydrogen. The evolution of the hydrogen concentration was measured in the course of time using gas chromatography. Experimental results are given in Figure 2.
Figure 2: Evolution of the hydrogen content in the reactor at different temperatures
(caracterisation of the H₂ recombination kinetics)

With respect to figure 2:

a) the evolution of the hydrogen content follows a decreasing exponential law

\[ \%H = Ae^{-Bt} \]

where A and B are constant values (see figure 2).

b) the H₂ recombination rate increases with temperature; compared to 25°C, the H₂ recombination rate is multiplied by 3 at 45°C, by 4 at 55°C and by 6 at 65°C.

When the temperature increases in a transportation/storage cask, gas generation from the radiolysis is generally enhanced. The present results thus shows that the recombiner fully answers this requirement since its recombination rate increases by increasing temperature.

2.4 Efficiency of the catalytic recombiners in presence of flammable and potentially poisoning gases

The efficiency of the recombiner was tested in presence of 5 other gases: CH₄; C₂H₆; C₂H₄; CO₂; HCl; I₂. The purpose was to ensure that these gases have no poisoning effect on the efficiency of the catalyst. Indeed, these species were identified by AREVA as being potentially generated by radiolysis of organic compounds. The latter are contained in canisters dedicated to residues from the reprocessing of used fuel. Each type of gas was tested separately in combination with hydrogen. The proportions were the following:

<table>
<thead>
<tr>
<th>Ratio of gases with respect to H₂</th>
<th>H₂/CH₄</th>
<th>H₂/C₂H₆</th>
<th>H₂/C₂H₄</th>
<th>H₂/I₂</th>
<th>H₂/CO₂</th>
</tr>
</thead>
<tbody>
<tr>
<td>2/1</td>
<td>40/1</td>
<td>40/1</td>
<td>1200/1</td>
<td>2/1</td>
<td></td>
</tr>
</tbody>
</table>
Concerning HCl, a capsule containing a concentrated HCl (36 %) solution was placed inside the testing reactor just above the catalysts. Figure 3 shows the evolution of the hydrogen concentration for each of the six tests. These curves can be compared directly since they refer to the same quantity of recombiner.

It appears that the catalyst shows a significant recombination capacity since it allows the hydrogen content to fall below 1% in less than 24 hours. In addition there is no poisoning effect for the gases CH₄; C₂H₆; C₂H₄; CO₂; HCl; I₂.

![Figure 3: Evolution of the hydrogen content in presence of potential poisoning gases.](image)

**Effect of CO:**
Moreover, the effect of carbon monoxide on the recombinating efficiency of the catalyst was also tested. The first tests were carried out in the same experimental conditions as previously, with a ratio of H₂/CO of 2/1. The evolution of gaseous concentrations showed that on the contrary to other gases, the carbon monoxide is consumed during the test. CO is oxidized into CO₂. This reaction is also catalysed by the recombiner and thus comes in competition with the H₂ recombining reaction. Consequently, the H₂ recombination becomes much slower, i.e. the hydrogen oxidation into H₂O actually starts when the quasi-total quantity of CO is removed. (Figure 4)
An additional test was carried out using the catalytic recombiner associated with carulite, which is a mixture of manganese and copper oxides. Contrary to the above mentioned tests, this experiment was carried out in dynamic conditions. Therefore, $H_2$ and CO were continuously injected into the reactor with a ratio flow $H_2$/flow CO equal to 11.5. The dynamic conditions are thus closer to the real conditions of a transport/storage cask, were the hydrogen and carbon monoxide are continuously generated by the radiolysis of the transported content.

Figure 5 shows the evolution of the of the hydrogen content as a function of time. It has to be pointed out that the hydrogen concentration is rapidly stabilized at about 0.5 %. This means that the hydrogen recombination takes place since the very beginning of $H_2$ and CO injections. Consequently, the presence of carulite allows a rapid stabilization of the hydrogen concentration far below the flammability limit of hydrogen in air.
CONCLUSION

Two catalytic systems based on the deposition of palladium and/or platinum on either a stainless steel grid or alumina beads were developed by AREVA TN International. On the one hand, the first system has shown to catalyse the oxidation of hydrogen at temperatures as low as 40 °C, even after immersion into a boric acid water solution. The catalysing capacity has also shown to remain stable after 1400 hours of testing. Furthermore, the recombination capacity is sufficient for stabilising the hydrogen concentration below its flammability limit, even with a low amount of recombiner. The required amount of recombiner can thus easily be introduced in a free space of the casks cavity.

On the other hand, concerning the palladium deposited on alumina, the recombination is ensured by the noble metal whereas the alumina allows the water produced by the recombination to be absorbed and avoid further radiolysis. The efficiency of this catalyst in presence of other radiolysis gases, i.e. CH₄; C₂H₆; C₂H₄; CO₂; HCl; I₂ has been demonstrated, whereas the presence of CO requires the use of carulite in combination with the catalytic recombiner. This second system has already been presented in TN International's patent <4>.

Both systems developed by AREVA-TN International answer the whole range of requirements for recombining systems either for wet transportation of used fuel or dry transportation casks such as transportation of vitrified residues.

REFERENCES


APPENDIX 1: Qualification results.
Evolution of the oxygen and hydrogen contents during 15 immersion/recombination cycles at the laboratory

Evolution of H$_2$ content

- week 1
- week 2
- week 3
- week 4
- week 5
- week 6
- week 7
- week 8
- week 9
- week 10
- week 11
- week 12
- week 13
- week 14
- week 15

O$_2$ content
METHODS FOR FABRICATING GAMMA – URANIUM – MOLYBDENUM (γ-UMo) ALLOYS AND THEIR INFLUENCE ON POWDER OBTENTION BY THE HDH TECHNIQUE

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ABSTRACT
Gamma uranium-molybdenum (γ-UMo) alloys has been widely considered as the best low enriched – high density fuels candidates for the substitution of the previously utilized high enriched ones, according to the RERTR requirements. For its usage as dispersions in plate type research reactor fuels, some of the techniques to transform the ingots into powder are highly influenced by the alloys’ properties achieved in the previous steps of melting and solidification. In this work we will briefly introduce the study of two of the main techniques to melt (γ-UMo) alloys, the induction and arc melting, and show some of the differences in properties presented by the casts and its powders, obtained by the technique of hydration-dehydration (HDH) thermal treatments. Samples of the ingots and powders prepared in the range of compositions of 5 to 10 weight % Mo, were characterized by means of scanning electron microscopy, hydrostatic density, and X-ray diffraction. It was verified that highly homogenous alloys can be obtained by the induction technique in only one step, and for those produced by arc, even with smaller loadings, several microstructural problems arises, leading perhaps to its invalidation as a technique for the fabrication of nuclear (γ-UMo) alloys and powders.

1. Introduction
In the nuclear area, the main techniques for fabricating the powders of γ-UMo alloys for its usage as dispersion fuels in nuclear research reactors are cryogenic milling, machining, atomization and hydration-dehydration (HDH). Atomization is the commercially most accepted, but there are works indicating that hydration-dehydration (HDH), as studied by BALLART et al. [1], SOLONIN et al. [2], or its variation, the hydration-milling-dehydration (HMDH) technique, as studied by PASQUALINI et al [3] and PASQUALINI [4], are also suitable to produce powders which obeys more closely the specifications requirements in terms of dimensions and granulometric distributions. This is the main motivation for the adoption of the HDH technique at IPEN / CNEN – Brazil, to the production of γ-UMo powders.

The responsible for the success of the HDH techniques is the high hydrogen affinity presented by the alpha uranium phase. In the ranges of compositions here studied, an alloy that presents some alpha as intergranular precipitates reacts readily with hydrogen, leading to high yieldings in terms of powder production. The reaction:

\[ 2 \alpha U (s) + 3 H_2 (g) \rightarrow 2 (\alpha U)H_3 (s) \] (01)

are related to the hydride formation, mainly in the grain boundaries. The hydride phase has a volume bigger than that of the alpha phase, and this volumetric difference generate tensions in the alloy that leads to its fragmentation, even without the step of dehydration. Thus, the ease of fragmentation and subsequent powder obtention implies necessarily in the gamma decomposition during the HDH thermal treatments. The reaction of the decomposition is:
\[ \gamma_{\text{matrix}} \rightarrow \alpha + \gamma_{\text{poor}} \]  \hspace{1cm} (02)

which produces precipitates with properties that are function of the HDH temperatures and, also, of the methods of the alloys preparation. The above equilibrium was extensively studied by REPAS et al. [5], VAN THYNE & McPHERSON, D.J. [6], VAN THYNE & McPHERSON, D.J.[7], SALLER, H.A., et al. [8], and more recently by HOFMAN et al. [9]. Their most important conclusions refer to the fact that the higher the molybdenum contents, the higher the gamma stability. Thus, alloys with higher alpha contents are, according to the reactions above, are more convenient to produce higher powder yieldings.

But the methods of powder production are highly influenced by the methods of alloys production, since that, according to the previous works, structure stability is a function of the molybdenum content and thus to the initial composition of the alloys. In all the alloys prepared by the arc melting technique, a subsequent thermal treatment was needed, even after remelts, to enhance their homogenization. In the induction ones, a high degree of homogenization could be obtained in only one step.

In the present work a brief introduction about how the methods of fabricating the alloys influence the choice of HDH condition is given. As there are no references on this issue in literature, our main objective is to show its importance in the nuclear technology, mainly in the fabrication of high density \( \gamma \)-UMo powders.

2. Experimental procedure

To the arc melted alloys, natural metallic uranium discs were used as charge. Molybdenum was used as a charge in small cylinders with 3 mm height and 3 mm diameter. Both materials were assembled in the copper plate of the furnace, its chamber closed and mechanical vacuum was applied to the system. After a suitable level of pressure, the vacuum valve was closed and a flow of argon was inserted inside the chamber. The arc was opened by means of an arc-starter, and applied over the sample until a high level of mixing between uranium and the molybdenum charges was obtained. The typical time to reach this configuration over the charges was about 40 seconds to 1 minute, maximum values to avoid damages in the chamber.

This procedure was repeated several times until, after visual inspection, the observation that the molybdenum charge was well homogenized in the sample. The main disadvantage in applying several remelts over the samples is the formation of an external oxide layer on the samples, deleterious to the quality of the alloy. Maximum masses of the alloys were about 30g.

For the induction melts, natural metallic uranium cylinders with 7 cm height and 2 cm diameter, and the same small cylinders of molybdenum were used as charge. Uranium cylinders were surface cleaned with 65\%vol. nitric acid, and inserted together with molybdenum, in a zircônia crucible, inside the furnace chamber. A cycle of purge and mechanical vacuum was applied and, after 3 operations, argon was inserted and the power of the furnace was raised until the melting of the samples. Masses of 700 g were utilized in each melting operations. The internal surfaces of the as cast alloys were analyzed by scanning electron microscopy, X-ray diffraction and its hydrostatic densities were also measured. Some of the results are discussed bellow.

3. Comparison between Arc and Induction Melting Techniques

Theoretical densities were obtained from some existent data in literature, for the 5 to 10 % weight molybdenum, and were shown in figure 1, together with the experimental determinations. The literature data are from the paper of TRYBUS [10].
In terms of densities, alloys prepared by both methods behaved the same way up to 8% Mo, the fall in the densities were almost at the same “rate”. After 8% Mo, the fall in the densities of the samples prepared by arc was substantial, and must be due to some closed porosity, which doesn’t constitute a problem in terms of powder obtention. It was observed also that this parallelism between the arc and induction density curves was possible only after the application of at least 2 remelts in the arc samples. In the case of the induction ones, no remelts were necessary, even working with charges 30 times heavier.

However, the most important difference in terms of quality is microstructural. It was observed that the structure of the induction alloys was mainly constituted by an homogeneous $\gamma$-UMo matrix plus some intergranular porosities, as we can see in the figure 2. In the case of the low-Mo alloys (5 to 7%wt. Mo), some $\alpha$-U is also present in the grain boundaries. In the arc samples, a big number of dendritic structures and some intragranular regions containing $\alpha$-U, even after the operations of remelts, were observed, which indicates some incompatibility between the speeds of cooling and diffusion of molybdenum in uranium in the samples, the first one faster than the second.

Structures containing high amounts of alpha uranium are the main responsible for the ease on the hydration-dehydration operations, but they are also related to a low degree of homogenization of both constituents of the samples. Thus, it is expected that, in the hydration of the arc melted samples, the rates of hydrogen absorption must be higher than that the induction rates. But if we are looking for homogeneous powders, it is necessary to work with the induction ones, and to try to find methods to enhance the hydrogen incorporation by these alloys.

Factors affecting the solidification of the alloys are mainly those related to the furnace’s project, like its geometry of melting (crucibles) and charges (how to assemble the charge into the crucible), possible impurities introduced in the charges by the crucible and the arc base materials, and mainly the cooling system.

As an example, X-ray spectra and micrographies of the $\gamma$-U8Mo compositions are shown in the figures 2 and 3, where we observe a high degree of homogenization presented in the induction sample. Dendrites are regions of low molybdenum concentration, and thus, the most suitable to promote high rates of hydrogen absorption, due to the high affinity presented by hydrogen and $\alpha$-U. They form, by chemical reaction, uranium trihydride, which leads, after dehydration, to the formation of powders. However, the remaining alpha uranium constitutes a loss of material, because there is no possibility to reconvert it to gamma, as it is usually segregated out of the gamma matrix.
4. Hydrogen Absorption

The experiments with hydrogen absorption for both alloys were carried out exactly at the same conditions of gas flow and sample’s masses and form. As an example, we can see below that the rates of absorption for those produced by arc melting was higher than for those produced by induction.
higher than that for the induction one. But, as denoted in the micrographies, inhomogeneities in composition are the main responsible for this high absorption rate. If gamma uranium is considered the more favorable phase for a high density fuel, such anomalies are undesirable, and must be treated as a process or method of fabrication loss.

Finally, in the case of the induction-melted alloys, the structures are much more homogeneous, presenting grains of a continuous gamma matrix and, sometimes, alpha precipitates in the boundaries, mainly in those of 5 to 7% weight Mo compositions. Thus, no homogenization thermal treatment is needed. If we take as comparison the same compositions, as the initial Mo content presented by the arc alloys, easiest is the powder obtention, but of an alloy with no homogeneous composition.

As an example, in the figure 5 are presented a micrography of a powder produced after thermal treatment under hydrogen applied on a $\gamma$-UMo induction alloy.

![Figure 5 – Powder particles of $\gamma$-UMo alloy.](image)

5. Conclusion

The difficulty of the obtention, in one single step, a microstructural homogeneous alloy by the method of arc melting, its abrupt reduction in the values of densities in compositions greater than 8% wt. Mo, lead us to the conclusion that, to the obtention of the same quality presented by the induction melted alloys, some features of the arc melting process must be changed, like the furnace’s geometry, number of remelts, geometry of loading, and mainly the cooling systems and thermal treatments conditions. Our main solution to avoid the problems of homogeneity in the arc samples was the change in the number of remelts. However, the remelts promotes also the formation of oxides, introducing impurities in the alloys.

Thus, the use of induction as a method of fabricating $\gamma$-UMo alloys are the choice here in IPEN-CNEN / Brazil. However, for low molybdenum alloys, where the problem of homogeneity is not too serious, arc could be used, conditioned to the application on the samples a sufficient number of remelts.

The important fact relating to the techniques of fabricating $\gamma$-UMo alloys is that the arc-melted alloys present, in all compositions, several dendritic structures, which are regions of low molybdenum concentration, mainly constituted by alpha-U phase, which leads to the application of an homogenization thermal treatment. But, as our experiences shows, they are not enough to eliminate all the dendritic structures, which are responsible to the differences in composition inside the grains.

This can be an advantage in the production of the powders, but at a cost of the loss of homogeneity. To the best preservation of the structural integrity of the alloys and their compositional
homogenization, the induction melting techniques is considered here the best choice to produce the powders of high gamma content alloys.

6. References


STUDIES ON THE AGING AND CORROSION BEHAVIOUR OF SIMULATED SPENT NUCLEAR FUEL

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ABSTRACT

To extrapolate data obtained on a laboratory timescale to time intervals relevant for extended interim storage or final disposal of high level nuclear waste in a geologic repository is a tough challenge. Emphasis is put on key aspects affecting waste corrosion: in particular, the effect of accumulating alpha-decay damage and helium on spent fuel is studied by monitoring relevant properties as a function of time and through annealing tests. The mobility/precipitation behaviour of actinides and fission products exposed to container/repository materials immediately after dissolution from the waste matrix is also considered. Finally, highlights from the effort to optimize suitable and complementary solution analysis tools for leaching experiments under repository conditions are schematically described.

1. Introduction

The aim of these studies can be summarized as follows:

a) Assessing the long term mechanical stability of spent fuel, which will determine the level of safety for transport/handling of the waste after intermediate storage times, and/or will define the surface exposed to groundwater attack, hence the radionuclides source term for the so-called instant release fraction in the case of final disposal scenarios.[1-3]

b) Defining and understanding the mechanisms and processes governing the chemical interaction between aged waste and groundwater, as these will directly determine the amount and the rate of release of radionuclides from the waste form.

This paper reports some highlights of these studies, including items belonging to the above mentioned lines of investigation and also an example of the effort to assess and optimize solution analysis techniques applied to the corrosion experiments on nuclear waste forms. Possibly the most challenging aspect of the studies on safe disposal of high level nuclear waste (in this case spent fuel) is the necessity to extrapolate to long storage times the behaviour of the waste form observed on an experimental timescale. Spent nuclear fuel available today is not fully representative of aged fuel after hundreds or thousands years of storage because it has not experienced the correspondingly long accumulation of microstructural defects and of He due to α-decay. UO₂ sintered samples containing short-lived alpha-emitters, the so-called alpha-doped UO₂, with specific activities spanning over six orders of magnitude were used to simulate spent fuel with different ages [1]. High activity material was used to rapidly accumulate levels of damage corresponding to spent fuel after thousands or tens of thousand years of storage [2].

If the aged spent fuel will become exposed to groundwater, corrosion mechanisms will cause the mobilization of radionuclides. It is important to assess the fate of these radionuclides, especially with regard to the capability of near field materials to act as retarding agents slowing down the transport of the dissolved species. This paper presents data obtained from spent fuel leaching experiments under simulated near field repository conditions [4] (in the frame of international cooperation) indicating immobilization of dissolved radionuclides on the iron canister material.
An adequate analytic support to corrosion tests involving multiple phases under realistic repository conditions must be provided. In addition to high sensitivity techniques particularly effective for trace concentration analysis, such as Inductively Coupled Plasma - Mass Spectroscopy, suitable techniques must be available that can be applied on a variety of solution types and capable to analyze solutions characterized by strong matrix effects. To this aim, a comparison of Inductively Coupled Plasma - Optical Emission Spectroscopy (ICP-OES) and Laser Uranium Fluorescence Analysis (LUFA) is schematically described.

2. Microstructural evolution of spent fuel during storage

As a result of the alpha-decay process, microstructural damage associated with the defects induced in the lattice by the energy transfer from the alpha-particle and the recoil nucleus will be produced; additionally, helium will accumulate in the fuel. The effects of alpha-damage accumulation on the microstructure of spent fuel have been studied on alpha-doped UO₂ specimens containing different amounts of either ²³⁸Pu or ²³³U. In terms of radioactivity level, these samples represent typical UO₂ fuels from reactor discharge up to million years of age [1].

Figure 1 shows a set of TEM micrographs of alpha-doped UO₂ specimens having cumulated alpha-damage ranging over 6 orders of magnitude. The early stages are characterized by formation of dislocation loops, progressively increasing in concentration and size. Small helium bubbles (1-2 nm) can be seen on the sample after a dose of 2 displacements per atom (dpa). The precipitation of helium into bubbles associated to the formation of the loops will result in swelling of the fuel during storage. The internal stresses caused by the formation of such defects might result in the embrittlement of this material. XRD data show an increase of the lattice parameter which is in perfect agreement with the TEM observations: a rapid increase at low dose followed by a smooth evolution towards saturation (not yet achieved) [1]. The evolution of the hardness follows the same trend [2].

All together the knowledge accumulated on the behaviour of UO₂ as a function of alpha-damage accumulation may contribute to define a model to understand the formation of the high burnup structure in irradiated UO₂ fuels [2-3].

A difficult aspect to treat in this type of studies is related to the kinetics of damage recovery. The assessment of diffusion coefficients, for helium for instance, but also for defects would help to predict the behaviour over long period of time of these materials. Radiation induced diffusion could contribute to the mobility of gas. Experiments are on-going to determine the solubility of helium in UO₂ at high pressure and high temperature and to study the interaction of helium with the defects, hence diffusion in a damaged lattice.

Fig. 1. TEM micrographs of alpha-doped samples showing the microstructure evolution with damage accumulation indicated as dpa values in the interval 10⁻⁵ - 2 dpa. The main feature in this range of accumulated dose consists of dislocation loops whose size and concentration increase with the dose.
3. Specific heat of $\alpha$-doped samples

Another example illustrating the effects due to the accumulation of alpha-decay damage in UO$_2$ refers to the energy stored in the defects produced in the lattice. Figure 2 shows the results of differential scanning calorimetry (DSC) measurements on alpha-doped UO$_2$ containing 10wt% of an oxide constituted mainly by $^{238}$PuO$_2$ after accumulating approximately 1 dpa. The apparent specific heat, $C_p^*$, of damaged samples was measured by DSC by applying ascending and descending temperature programmes in the range 400 K – 1500 K. The deviation of the measured $C_p^*(T)$ from the real heat capacity, $C_p(T)$, is related to the recovery of the latent heat of the lattice defects during thermal healing.

Calorimetry of strong $\alpha$-emitters is perturbed by the heat generated by radioactive decay. In fact, the apparent temperature-ascending curve of $C_p^*$ is lower than the real one, whilst the descending curve is higher. However, the average of these two curves gives exactly the value of the unbiased $C_p$. The $\alpha$-decay heat generated by the sample is known to be 0.0702 Wg$^{-1}$ for 10 at% $^{238}$Pu with 5.499 MeV energy per $\alpha$-particle and the same energy for the recoil daughter. Knowing this energy source, whose effects are perfectly anti-symmetric in the ascending and descending curves, the calorimetric signal produced during damage annealing could be accurately measured and converted into energy. The real $C_p(T)$ obtained from literature data [5] for $(U_{0.9},Pu_{0.1})O_2$ corresponds very well to the average obtained between the ascending and descending curves of undamaged samples.

![Fig. 2. Apparent Cp* curves for $(U_{0.9},^{238}Pu_{0.1})O_2$ obtained by DSC during temperature ascending and descending runs (i.e. before and after annealing). Two experiments were performed (duplicate runs).](image)

The peaks of the latent heat effects appearing during the annealing (ascending) runs at temperatures corresponding to different healing stages can be deconvoluted and analysed in order to identify the parameters of the latent heat effects. For each stage, the quantities derived are: concentration and energy associated to the annealing of a certain kind of defect, as well as its characteristic mobility [2].

4. Reduction and incorporation of redox sensitive nuclides in corroded iron surfaces

To understand the fate of redox sensitive radionuclides released from high level radioactive waste, secondary mass ion spectroscopy (SIMS) analysis was conducted on iron coupons reacted for two years in spent fuel leaching experiment performed at Studsvik Nuclear AB, Sweden in the frame of a SKB project [4].
The concentration of radionuclides in solution from the leaching of a 47 MW/kgU spent fuel sample in synthetic groundwater dropped quickly after inserting iron metal coupons in the leaching solution. This is explained as reductive precipitation of these redox sensitive radionuclides on the iron surface. SIMS analysis (Fig. 3) confirmed that U(VI) and Pu(VI)/Pu(V) are reduced and immobilized in the corrosion layer on the iron surface. Most U and Pu are coordinated with the inner parts of the corrosion layer, where Fe(II)-rich corrosion products are dominating and therefore more reductive than the Fe(III) oxides occurring in the outer periphery of the corrosion layer. The result of SIMS analysis also showed that Si was coordinated with iron corrosion products in both inner and outer parts of the corrosion layer. From these data it can be concluded that under simulated near field conditions the redox sensitive radionuclides released from spent fuel can be reduced and precipitated on corrosion layer of the proposed canister material, i.e. cast iron.

Fig. 3. SIMS analysis of the cross section of reacted surface of cast-iron coupons. Nuclide mapping shows the incorporation of U, Pu and Np in the corrosion phase of Fe. The bright yellow parts in Fe mapping represent the cast-iron metal matrix. Si was from the glass vessel or contained in cast iron.

5-Uranium analysis by ICP-OES and laser fluorescence techniques

A comparison between ICP-OES (Horiba Jobin Yvon Ultima 2 spectrometer) and LUFA (Scintrex UA-3) has been performed on uranium solutions, to assess the complementarity and range of effectiveness of the two techniques. In both cases, external calibration was performed using certified uranium standard solutions (Alfa Aesar Specpure). The samples analysed by ICP-OES had dilution factors between 0 and 15. A high generator power was used to minimize the matrix effects. The wavelength was detected using the profile function, and by using a semi-quantitative analysis mode using multiple wavelengths. Two analytical lines of uranium, 367.007 nm and 409.014 nm were used with best limit of detection of 1.11 ppb and 4.31 ppb, respectively. Three replicas per line and element were performed for each sample. The samples analysed by LUFA had dilution factors between 0 and 7400. The intensity was zero for a uranium concentration <2 ppb. The relative standard deviation of the method was 5.4%. Fig. 4 shows the uranium concentration measured by laser fluorescence analysis and ICP-OES with 67 % level of confidence. The results are in good agreement with the calculated values and with the semi-quantitative method. Both methods are effective in a wide range of
concentration levels (ppb to hundreds of ppm), are fast and precise, and can be used complementarily for the measurement of uranium. LUFA accuracy was ~5%, while in the case of ICP-OES accuracy better than 1% is achievable.

![Graph showing measured uranium concentrations](image)

Fig. 4. Measured uranium concentrations with 67 % level of confidence.

A procedure to analyze uranium in solutions characterized by strong matrix effects (brines) was developed for ICP-OES. Such effects constitute a limiting factor for the use of the LUFA. ICP-OES results for U in high concentration NaCl solutions indicated that the background shift caused by sodium can be corrected and reliable analyses of uranium can be performed in the brine matrix.

6. Summary

Predicting the behaviour of spent fuel during storage and/or in a geologic repository is a complex task requiring simulations and extrapolations to a remote future. The related studies must cover a broad spectrum of mechanisms and processes, including the evolution of the waste under the effect of accumulating alpha-decay damage and He, and the corrosion behaviour in presence of groundwater and repository agents. The first aspect is investigated by measuring property changes and recovery behaviour of materials under the effect of accelerated dose accumulation at a microscopic and macroscopic level; the second is studied by singling out the governing mechanisms determining the release and mobility of radionuclides. Appropriate analytical tools must be available to characterize the systems of interest, especially when simulating realistic, multi-phase repository configurations.

References

THE CURRENT STATUS AND FURTHER DEVELOPMENT OF RADIATION ENGINEERING FOR NUCLEAR POWER, INDUSTRY AND MEDICINE

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ABSTRACT

In the paper there are described the basic foundations of radiation engineering and there are reflected the questions of a current status and further development of radiation engineering in Russia. There are given some examples of practical use of the research and development in the field of radiation engineering for the solution of the problems of nuclear power engineering and nuclear fuel cycle, metallurgy, geology, mining, gas and oil and other industries, medicine, ecology etc. There are described the main methods and means of radiation engineering. Radiation technology installations are used for food treatment, for sterilization of medical products as well as for purification of drinking and waste water. Radionuclide power units are applied for power supply of the automatic equipment on the remote or unattended objects in Polar areas or in Space. Radiation instruments are used for quality testing, for analysis of substance composition and for control of production processes, as well as for radiation diagnostics and radiotherapy of human diseases. The information on gas discharge detectors of radiations, which are developed and are producing in VNIITFA, is given also.

1. Introduction

The artificial radioactive nuclides and other sources of ionizing radiations are widely applied today in various branches of industry, medicine and agriculture. It became possible only due to development of researches in the field of nuclear physics, progress in the nuclear power and accelerators engineering, large-scale manufacture of nuclides, success in instrumentation and computerization. The listed factors have caused appearance of a specific direction in a nuclear science and engineering – the radiation technology and engineering. The radiation technology and engineering is a complex of nuclear and physical methods and radiation devices based on use of interaction of ionizing radiations with substances. It is intended for treatment on materials and substance to receive new or to change the existing properties of materials, to realize the radiation therapy of various man’s diseases, to obtain the information of qualitative and quantitative data of composition and properties of materials and products, to control and manage production processes, and also to transform energy, which emits as a result of radioactive decay, in other kinds of energy.

To the present time three basic directions of radiation engineering were defined: radiation technology and equipment, radiation instrumentation and radionuclide power engineering. The aim of radiation technology is creation the methods and devices, which are based on chemical, structural or biological changes, taking place in substance under influence of the certain absorbed doze of ionizing radiation.

The aim of radiation instrumentation covers methods and equipment for obtaining the information on parameters of a condition, quality either composition of substances or products functionally connected to a flow of ionizing radiation.

The aim of radionuclide power engineering is creation various power devices, in which as a source of primary energy is the energy, which emits as a result of processes, accompanying radioactive decay. A complex of scientific and technical problems exists in all these directions, many of such problems were developed in independent scientific disciplines, for example, radiation chemistry, gamma-radiation therapy, activation analysis, radiation methods of the non-destructive testing etc. Application radioactive nuclides and the radiation engineering in various areas of human activity rapidly increase all over the world.

Many institutions and enterprises in our country and abroad are engaged in development and production of radiation engineering equipment and equipment for use of radioactive nuclides and other sources of ionizing radiation. The international contacts in this area are intensively growing.
The current status of works in the field of radiation engineering and radionuclides application is characterized by the following:

- There is an expansion of the areas of biomedical application of radiation methods for treatment and diagnostics of various diseases, for sterilization of medical production, for irradiation of food and agricultural production, for treatment of drinking water and wastes;
- There is a definite delay in an occurrence of new methods and development of essentially new types of the radiation equipment against a background of quantitative growth of used products of radiation engineering;
- There are qualitative changes in developing products, which are bound up with wide application of computer engineering and automatics, use of more perfect converters, detectors of radiations, optimization of the characteristics and activity of irradiators etc., that allows essentially improve the data of equipment on accuracy, sensitivity, productivity, enables to use the received data for automation of manufacture;
- The data of radiation safety of used equipment are improved owing to automation of irradiation equipment control, optimum choice of sources specific activity (down to application in some cases of sources with minimally significant activity), to application of new shielding materials and perfection of a radiation protection design.

The successes and achievement of radiation engineering in Russia in many respects are bound up with investigations in Russian National Technical Physics and Automation Research Institute (VNIITFA), which till 1989 referred to as All-Union Research Institute of Radiation Engineering. In institute alongside with other works in the field of technical physics, the works on radiation technology and creation of the irradiation equipment, gamma beam therapy machines, radionuclide power units, radiation means for non-destructive testing, nuclear analytical technique for the elemental analysis of substance, etc. were received the large development.

It is possible to show some tendencies of development of radiation engineering in Russia on examples of the most interesting researches and development of the radiation equipment, which has been carried out recently.

2. Nuclear analytical methods and equipment

Much attention is devoted in VNIITFA to researches and development of nuclear and physical methods and equipment for the analysis of substance composition. The especially significant progress was achieved in the neutron activation and neutron absorption analysis, in photonuclear methods and in radionuclide X-ray analysis.

It is can be said, that VNIITFA specializes in specific questions of nuclear and physical methods of analysis for industry. The researches spent for this area, carry the brightly expressed applied character directed on creation of devices and complexes of the equipment, which finds the main application in an industry, in mining and geology.

The neutron activation analysis possesses very high sensitivity of various elements determination when nuclear reactor is used as a source of neutrons. A number of reactor activation analysis techniques are developed in institute. In particular, the neutron-resonance method developed in VNIITFA is successfully used for instrumental determination of some noble metals.

When small-sized generators of neutrons and powerful radionuclide sources of neutrons made of $^{252}$Cf became available, the researchers of institute carried out researches and development on creation of stationary and transportable neutron activation analytical device for an industry and geology. The special attention in methodical development of institute was paid to activation analysis with use of short-lived nuclides. That allows to receive high efficiency of the analysis of separate samples and even to conduct the continuous analysis of technological products.

The neutron absorption analyzer was developed in VNIITFA for determination of elements with high section of neutrons absorption. In particular, now majority of atomic power stations in Russia with reactors of VVER type and many foreign NPP (in Bulgaria, Czech Republic, Slovakia, Ukraine) are equipped with developed in VNIITFA neutron analyzers of $^{10}$B in the coolant of the first contour of reactor.
The gamma activation analysis is applied as the analytical tool for solving of various problems. This method possess with sharp selectivity, sensitivity, high accuracy and rapidity. In order to achieve these advantages the scientists of institute carry out the important work on more precise definition the nuclear data for photonuclear reactions. The results of these researches were applied for development of the industrial gamma-activation equipment.

The laboratories for gamma-activation analysis of gold ores were created in VNIITFA. High efficiency and rapidity allow using the results of the ore samples analysis for directional ore mining. The linear electron accelerators are the most convenient gamma-radiation sources for realization of gamma-activation analysis. The electron accelerators allow receiving not only intensive bremsstrahlung, but also very high neutron fluxes. So, for example, in installation "Arctica" the accelerator LUE-15A with beryllium converter allows to have a neutron flux up to $10^{13}$ neutrons per second. It significantly expands a circle of analytical tasks can be solved by application of neutron activation analysis in a complex with gamma-activation with use of electronic accelerators.

The radionuclide X ray analysis is the method based on excitation of atoms of analyzed elements with the help of primary radiation from radioactive nuclide. It is one of the most widely used analytical techniques. Simplicity, the small dimensions and cost of the equipment in a combination with sufficient sensitivity for solving many practical tasks made the radionuclide X ray method the most convenient analytical tool for mass and multielement express analysis of technological or geological samples. It applies also for analysis of rocks and ores in-situ, automatic control of technological products in a flow etc. All these directions have developed in Russian National Technical Physics and Automation Research Institute. The scientists of institute have brought in the significant contribution to development of scientific bases of the radionuclide X ray analytical method and creation of devices for various purpose intended for operation in field, laboratory and workshop conditions.

At processing results of the nuclear-physical analysis the great importance is attached to correct decoding of radiation spectra, account of interfering influence of other elements. It concerns both to activation and to radionuclide X ray analysis. Therefore in institute the special computer methods and al-
gorithms of processing of spectra are created, the great importance is attached to development of various physical principles for exception of interfering influence of a matrix.

Fig. 3 Portable radionuclide X ray analyzer ПРАМ-1 for geology

3. Non destructive testing

The wide range of problems for the production control in various branches of industry is successfully solved with use of the equipment of the non-destructive testing and radiation gauges for determination of various technological data.

The means for non-destructive testing, applying mainly radiation methods, for an estimation of quality of products made of various materials and possessed a complex configuration and technology of manufacturing are developed in VNIITFA.

Due to high penetrating ability of radiation, in particular, of gamma-quanta and neutrons, it is possible to transfer the information in form of radiation signal through walls of production equipment, pipelines, capacities etc.

Development of methods and equipment for gamma and neutron radiography, self radiation radiography, radiometric flaw detection, radiation methods of the determination of such parameters, as level, density, throughput rate of materials, thickness of walls and coverings, granulometric composition of powders, distribution of material, the presence of interstices and cracks etc., allows to solve set of problems taking place in industries.

Use of packaged systems and typical structures, unification and normalization of elements of the developed equipment, wide application of computers and new types of radiation sources allow to create the equipment of the non-destructive testing and technical diagnostics at a level of the best world model.

Choice of detectors with the maximal "signal - noise" ratio, PC-processing of the radiographic and radiometric information allow to increase volume of information, to receive the volumetric and level-by-level images of controllable products. These investigations were used as a base of industrial computer tomography methods and devices, developed in institute.

Fig. 4 Radionuclide computer tomography device
For realization of radiography at small focal distances the special acute-angled focal sources are created at the basis of isotope $^{192}$Ir. The testing of thin-walled products will be carried out with use of sources low energy gamma radiation at the base of radionuclide $^{75}$Se etc.

On the basis of researches carried out by scientists of VNIITFA in cooperation with the experts of an industry it was created the line of gamma radiography flaw detectors for general application and the line of radiometric flaw detectors, allowing to investigate the complex shape products, objects located in out-of-the way places, details and units in process of their manufacturing, welded seams of main pipelines, power equipment etc.

Fig. 5 The tomogram of the nuclear fuel elements assembly for NPP, received with help of industrial radionuclide computer tomography device

Fig. 6 Gamma radiography flaw detector RID-Se/4 with a source $^{75}$Se

Fig. 7 Gamma-radiography flaw detector RID-K/100 using a $^{60}$Co source for transmission of products with a wall thickness up to 200 mm on steel
Development of nuclear power in our country, necessity of increase of its efficiency and the safety define the increased requirements to quality of the NPP equipment, including nuclear fuel elements. The high emphasis is placed on development at institute the means for non-destructive testing of objects in nuclear power engineering.

The development of the set of means for non-destructive testing in process the manufacture of nuclear fuel elements for power reactors VVER-1000, RBMK, BN will be carried out by VNIITFA in cooperation with a number of other institutes and enterprises. The purpose of this work - to supply the enterprises, producing the nuclear fuel elements, with the automated means for testing the fuel elements at all stages of their manufacturing, starting with nuclear fuel powders up to ready nuclear fuel assemblies.

For study both not irradiated, and irradiated fuel elements the methods neutron radiography can be applied. This method is used also for study of many other objects. The investigations on application of neutrons for non-destructive testing were carried out in VNIITFA within many years. Neutron radiography allows obtain the information on parameters of products located behind shields from heavier materials, to reveal inclusions of hydrogen containing materials, of substances with the large neutron-absorption cross-sections etc. In VNIITFA there were created some installations for neutron radiography with use of high active radionuclide $^{252}$Cf neutron sources, and also beams of neutrons produced by nuclear reactor.

A lot of industrial technological parameters can be determined with the help of radioisotope instruments.

The instruments of this type developed in institute, are used for measurement of humidity of coke in metallurgy, soils and spoils in construction works, size grading of the crushed ores and powders, inspection of filling the bins of feeding systems of blast-furnace burden, control by anti-icer systems of civil aircrafts etc.
4. **Gas-discharged radiation detectors**

The gas-discharged detectors of ionizing radiations are the major element of the instruments for radiation measurements. The gas-discharged detectors are simple on a design, are convenient in work, provide reliable measurements of various kinds of radiation in a wide range of the external influencing factors.

The fields of application of discharge detectors are wide and are various. They are used in inspection and protection systems of nuclear power units, in systems of anti-nuclear weapons protection of industrial and civil objects, instruments of the radiation inspection of the nuclear power plants personnel, personnel of a nuclear industry enterprises and population of the country, and also in the geophysical instruments for minerals investigation, devices for space researches etc.

There were created the essentially new types of gas-discharged detectors (such, as corona counters and integral-pulse chambers), which are used for the solving of a number of problems in reactor engineering.

The development of nuclear industry and nuclear power engineering has required the creation of neutron ionization chambers for registration of the neutron fluxes in control and protection systems of nuclear power plants and research reactors.

The ionization neutron chambers with solid boron radiator and ionization fission chambers containing as a radiator fissionable isotopes of uranium are widely applied in nuclear industry. They possess the high stability of intensive neutron fluxes registration.

The nomenclature of the gas discharge detectors of nuclear radiation created and produced in institute, numbers more than 60 types. Among them there are, for example, neutron counters both with solid boron and with gaseous $^3$He radiators.
Fig. 11 Ionization chambers of a various type

Besides of gas discharge detectors of radiations VNIITFA develops and produces also the suspension brackets of ionization chambers - the vacuum and gas-filled communication lines of ionization chambers with the electronic recording equipment.

The cited above review shows, that the radiation engineering becomes an advanced sphere of a science and engineering, the areas of its application are wide - from nuclear power and metallurgy up to agriculture and medicine.

5. Radiation technology equipment

The radiation technology is based on physical, physics and chemical and biological processes taking place in substances as result of interaction with ionizing radiations and causing the appropriate technological effect.

To the present time some dozens of various radiation technological processes are developed and they are at various stages of trial and industrial realization on the following directions of radiation technology: to synthesis of new chemical compounds and initiation of chemical reactions, polymerization, vulcanization and cross-linking of polymers, clearing of waste waters and gases, sterilization of materials, irradiation of foodstuff for increase of terms of their storage, giving of new properties to solids, in particular, to semi-conductor materials etc.

The significant contribution was brought by the researchers of VNIITFA to creation of scientific and technical bases of radiation-technological process and equipment. It was developed the complex of accounts the basic parameters of radiation technological installations, methods of irradiator optimization, definition of the rational circuits of management, control, protection and automation of the radiation equipment. The methods of definition of dose field topography, operating ratio of radiation for various configurations of irradiated objects were developed also. The large attention was given to monitoring of powerful fields of radiation.

The most highly developed area of radiation technology is the radiation sterilization, which is especially effective at processing of heat labile materials, for example, medical products made of polymeric materials (syringes-ampoules of single application, catheters, catgut, devices of a taking and transfusion of blood etc.), dressings etc. A radiation sterilization is carried out with use of both isotope sources of radiation, and electron accelerators. The plants, developed in VNIITFA with $^{60}$Co radiation sources of activity up to 1 MCi for sterilization of blood systems, operate at the factories in St.-Petersburg and in Belgorod-Dnestrovsky. The installation with two linear electron accelerators ЛУЭ-8/5В type for sterilization of medical production operates in Kurgan. The modern enterprise for manufacturing obstetrical packages equipped with gamma-installation for their sterilization has been constructed in Kondrovo in cooperation with companies from Finland. A number of other installations for the enterprises of medical products are created.

The radiation treatment is used also for processing of food and agricultural products. It is known, that for the various reasons (rotting, germination, the damage by insects etc.) lose a plenty of the foodstuffs and agricultural products (raw material, seeds etc.). VNIITFA conducts also works in this very important direction of radiation technology. For many kinds of products the optimum modes of radiation processing are established, the long-term researches of their suitability and harmlessness of use are carried out. The irradiation installation for these purposes are created, a part of them was supplied for foreign customers.
Fig. 12 Radiation installation in Republic of Peru has been constructed with the help of the VNIITFA experts

The irradiation exerts stimulating influence on course of a number of chemical reactions. The radiation-chemical reactions in comparison with reactions initiated by a heat or other kinds of energy, characterized by weak dependence of velocity of initiation from temperature, opportunity of realization without application of the additives and catalysts, dependence of velocity of initiation on a dose rate. Radionuclide sources of radiation ($^{60}$Co) as well as the powerful electron accelerators are used in plants for realization of radiation-chemical processes. The release of a lot of unique products and materials, such, as self-adhesive electric insulating tape and rubber glass cloth, thermo-shrinking polyethylene isolation, cotton fabric with anti-microbe, haemostatic, anti-putrefactive properties, synthetic fabrics with the improved consumer properties, is under production conditions adjusted.

The great importance is attached now to protection of an environment. Many problems in this important direction can be solved by radiation processing of gaseous and liquid industrial, household and agricultural wastes. The installations developed by VNIITFA are used at chemical and medical instruments plants and can be used for prevention of pollution of an air and natural waters.

Fig. 13 The circuit of installation for electronic processing of drinking and waste water
1 - electron accelerator, 2 – reactive chamber, A1 - air, A2 - air in an atmosphere, P – collection of ef-fluents, W - treated water
The further development of works in the field of radiation technology equipment is directed on creation of the specialized industrial installations, increase of their technological and economic data, reliability and convenience in operation.

6. Radionuclide power engineering

Energy, released as a heat following a radioactive decay, can be then transformed into electrical energy. Radionuclide sources of thermal and electrical energy have essential advantages at operation of the equipment with the such sources, mounted in remote regions of Globe, which are difficult to access, due their high specific energy capacity (thousand W·hour/kg), long service life (10 years and more), high reliability, keeping of serviceability by short circuit.

Therefore radionuclide power units (radionuclide thermoelectric generators (RTEG), the radionuclide power devices (RPD) etc.), developed and produced in VNIITFA ("Бета-С", "Бета-М", "Эфир - МА" etc.), find rather wide application for power supply of automatic hydro meteorological stations, various means of navigation - sea lights and radio beacons. The radio beacons, located on a line of Northern Sea Route, promote significant increase of navigation safety. More powerful radionuclide power installations are applied to the power supply of light beacons. Such installation, created in institute, was equipped, for example, Tallinn beacon by the Baltic Sea. In Antarctic Region RTEGs are used for a power supply of magnetic variation stations.

The radionuclide power engineering is on a junction of several sciences: nuclear physics, radiochemistry, thermal physics, the electrical engineering etc. Therefore, the experts of a various structure many scientific establishments and industrial enterprises of our country in creation of RPD elements take part.

The scientific and technical bases of RPD creation, principles of designing, mathematical modeling interconnected to nuclear physical, thermal physical and electrical processes are developed in Russian National Institute of Technical Physics and Automation with the purposes of optimization the parameters of separate parts of RPD and device as a whole.

As the basic fuel for radionuclide thermoelectric generators $^{90}\text{Sr}$ is mostly used, and $^{238}\text{Pu}$ is used for highly power intensive RPD.

In RPD, developed in VNIITFA, the thermo electric batteries are most widely used for transformation of energy, released by radioactive decay, to electrical power. The current researches are directed on rational use and other methods of transformation.

In institute the works on creation of the radioisotope power supply for the pump of artificial heart were carried out. Such source can be implanted directly in the body of patient together with the mechanism of artificial heart. However these works were bounded only by experimental stage, and in the near future, probably, the external systems of artificial blood circulation will be in using.
7. Radiation therapy equipment

The research and development in the field of radiation medicine are carried out at VNIITFA from the moment of its foundation and they are directed on creation of the equipment for beam therapy (mainly malignant neoplasm). The works were carried out in close collaboration with leading medical institutes and clinics of the country.

The complex of researches, executed in VNIITFA, has allowed proving the main radiation and physical data of remote and contact therapy equipment with radionuclide sources of radiation, to develop scientific and technical bases of its designing, to create methods and means of formation of radiation fields with the given data, to ensure their stability during beam treatment. The algorithms and computer programs were developed for account the dose distributions from sources of gamma and neutron radiation, which are handed to medical institutions for clinical application.

The modern radiological equipment for intracavity and intratissue gamma-therapy with high active radiation sources for application in oncogynaecology, proctology, urology, teratology (АГАТ-В1, АГАТ-В5, АГАТ-БТ), developed in VNIITFA, is a great scientific and technical achievement in area of radiation therapy.

Fig. 16 The device АГАТ-БТ for contact gamma-therapy

In institute were developed and continue to be improved remote gamma-therapy devices, static and rotational types (such, as АГАТ-С and АГАТ-Р, АГАТ-РМ, АГАТ-Р1). The devices with programmed control systems were developed on the basis of modern microcomputer. This equipment allows passing from automation the separate stages of beam treatment to complex automation the process of pre-irradiation preparation, to development of the individual optimum programs of beam treatment and their realization that considerably raises treatment efficiency of the patients.

Fig. 17 The device АГАТ-Р1 for remote gamma-therapy
VNIITFA developed also the device for intracavity neutron therapy with use of a radionuclide source of neutrons made of Californium-252. To the present time oncological health centers, clinics, the research institutes of Russia are equipped with several hundreds radiotherapy devices most of them was developed in VNIITFA. Other major direction in modern medicine is effective diagnostics of some serious diseases. In our institute is offered radionuclide X-ray fluorescence method and is realized in equipment for diagnostics of thyroid glands diseases and for control of its treatment. This equipment allows in tens time to reduce a dose load on organism of the patient in comparison with well known radioisotope methods of the iodine control in thyroid gland. This method gives qualitatively new opportunities for diagnostics and to carry out the control of process of treatment.

Fig. 18 The equipment for diagnostics of thyroid glands diseases and control of their treatment

It is already more than 40 years as VNIITFA is the leading scientific establishment on creation of new methods and devices applying radionuclide sources of radiation and accelerators. It seems a further use of radiation engineering will develop rapidly.

References

Poster Presentations

Session III:
Medical Applications
LONG TERM RETENTION OF D- AND L- $[^{123}\text{I}]-2$-IODOPHENYLALANINE IN R1M TUMOUR BEARING RATS: A POTENTIAL FOR RADIONUCLIDE THERAPY

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ABSTRACT

The aim of this study was to compare in vivo $[^{123}\text{I}]-2$-I-D-phenylalanine and $[^{123}\text{I}]-2$-I-D-tyrosine with their respective L-analogues $[^{123}\text{I}]-2$-I-L-phenylalanine and $[^{123}\text{I}]-2$-I-L-tyrosine, while using $[^{123}\text{I}]-\alpha$-Methyl-L-tyrosine as a reference. The tumour uptake and biodistribution for all tracers was measured in the same set of R1M rat rhabdomyosarcoma tumour bearing Wag/Rij rats by dynamic planar imaging and by static planar imaging at 24 and 48 hours. All tracers showed a favourable tumour uptake at 30 minutes p.i., with tumour DUR values ranging from 1.7 to 2.4. After 24 hours both tyrosine analogues but also IMT were almost completely cleared. There was a high retention however of both $[^{123}\text{I}]-2$-Iodo-D-Phe and $[^{123}\text{I}]-2$-Iodo-L-Phe activity in the tumour and body, resulting in an increased tumour/background contrast over time. In conclusion, both the D and L analogues of $[^{123}\text{I}]-2$-Iodo-Phe have a favourable tumour uptake and biodistribution, but moreover a very long retention in the tumour allowing radionuclidetherapy.

1. Introduction

The LAT transport system is an important key in tumour imaging using radiolabeled amino acids. This transporter is a major nutrient transport system responsible for Na$^+$ - independent transport of large neutral amino acids including synthetic amino acids by an obligatory exchange mechanism coupled to an anti-port system [1-4], and has several subtypes such as LAT1 and LAT2. LAT1 expression was scarcely detected in non-tumour areas [5-6] but highly expressed (up-regulated) in proliferating tissues, in particular malignant tumours, as it plays a critical role in cell growth and proliferation. A remarkable characteristic of the LAT-1 amino acid transport system is its broad substrate selectivity, which enables the transporter to accept amino acid related compounds, such as D-amino acids and cancer drugs like Melphalan [3,6]. LAT2 on the other hand has a high level of expression in small intestine, kidney, placenta, brain and in epithelia and blood-tissue barriers [4,7]. It transports all of the isomers of neutral alpha-amino acids by facilitated diffusion; however it does not transport D-amino acids.

Kersemans et al [8] have shown in tumour bearing athymic mice that D and L $[^{123}\text{I}]-2$-I-phenylalanine ($[^{123}\text{I}]-2$-I-Phe) were taken up in different types of tumour cell lines. They also noticed that while the tumour clearance of $[^{123}\text{I}]-2$-I-D-Phe was faster than of the L analogue, at 24 hours p.i. the tumour-background contrast of the D analogue was 3 times higher than of the L analogue. Recently, Tsukada et al. demonstrated in tumour bearing mice the advantages of D-isomers of O-$^{13}$C-methyl-tyrosine and O-$^{18}$F-fluoromethyl-tyrosine as Tumour-Imaging Agents as the tumour to blood ratio was higher for the D-isomers compared to the L-isomers [9].
2-I-L-tyrosine (2-I-L-Tyr) and 2-I-L-Phe [10-13] both show a very high tumour selectivity when compared to [18F]-FDG which shows a considerable uptake in brain and inflammatory tissue. Here we compare the pharmacokinetics of [123I]-2-I-L-Phe, [123I]-2-I-D-Phe, [123I]-2-I-L-Tyr, [123I]-2-I-D-Tyr and [123I]-α-Methyl-L-Tyr in a R1M tumour bearing Wag/Rij rat model. The biodistribution, the tumour uptake and tumour retention of these tracers are investigated via dynamic and static planar imaging, both at short time p.i. (0-40 minutes) and long time (24 and 48 hours p.i.). Dissection experiments are used to confirm and elaborate biodistribution data of [123I]-2-I-L-Phe and [123I]-2-I-D-Phe.

2. Material and methods

Precursor synthesis and labelling
The synthesis of 2-I-D-Phe / 2-I-L-Phe and the nucleophilic radioiodination of 2-I-D-Phe, 2-I-L-Phe, 2-I-L-Tyr and 2-I-D-Tyr were achieved by Cu+1 assisted nucleophilic exchange as earlier described [11]. [123I]-α-Methyl-L-Tyr labelling was achieved via the iodogen method [14]. The labelling yields for all tracers were at least 98%. After passing through a sterile 0.22 µm Ag-membrane filter (Millipore) a radiochemical purity of > 99% was achieved.

Laboratory Animals
Food and water was ad libidum during the experimental period. For the tumour model, male Wag/Rij rats (Bioservices, The Netherlands) were injected subcutaneously in the right flank (armpit region) with 15.10⁶ R1M rhabdomyosarcoma cells. All imaging experiments and all dissection experiments were performed 6 weeks after injection of the R1M cells. All tracers were injected intravenously (IV) in the penis vein. The study protocol was approved by the ethical committee for animal studies of our institution. Guidelines of the National Institute of Health principles of laboratory animal care (NIH publication 86-23, revised 1985) were followed.

Dynamic Planar Imaging (DPI)
DPI was performed in a crossed (two by two) experiment with a 2-day interval using four R1M bearing Wag/Rij rats. DPI was started immediately after i.v. injection of 18.5 MBq [123I]-2-Iodo-D-Phe or [123I]-2-Iodo-L-Phe and continued up to 40 min for all rats (day 0). Twenty-four hours and forty-eight hours p.i. a static image was acquired. This set-up was repeated in the same rats for [123I]-2-I-L-Tyr and [123I]-2-I-D-Tyr (day 7-11), after which [123I]-IMT imaging was performed (day 14-16). All rats were also injected with 37 MBq [99mTc]-Pyrophosphate ([99mTc]-PyP) ([99mTc] from a generator, GE) for blood pool estimation (day 16). The imaging experiment with [123I]-2-Iodo-D-Phe and [123I]-2-Iodo-L-Phe was repeated twice, each time with 4 rats.

For dissection experiments, 3.7MBq [125I]-2-Iodo-L-Phe or [125I]-2-Iodo-D-Phe was injected i.v. For each tracer, 5 animals were euthanised 30 min p.i. and 5 animals at 24 hours p.i.

3. RESULTS

Dynamic planar imaging
The uptake of all tracers reached its maximum between 10 and 30 minutes p.i.. The DUR value of the tumour at 20 minutes p. i. of [123I]-2-Iodo-L-Phe, [123I]-2-Iodo-D-Phe, [123I]-2-Iodo-L-Tyr, [123I]-2-Iodo-D-Tyr and [123I]-IMT is respectively 2.06 ± 0.43; 2.26 ± 0.81; 2.46 ± 0.77; 1.71 ± 0.61, 2.43 ± 0.56 and 2.43 ± 0.56 (data: mean ± SD (n=4)). The [123I]-2-Iodo-D-Tyr uptake is lower and yet the contrast (tumour/tissue ratio) versus heart (blood), muscle and brain is comparable with the values of [123I]-2-Iodo-L-Tyr and the phenylalanine analogues. The urinary excretion of [123I] activity of [123I]-2-Iodo-D-Tyr is much faster than of it’s L-isomer and represents 1/8 of the total body activity after 30 minutes. This phenomenon was also observed, but to a much lesser extent, for [123I]-2-Iodo-D-Phe. The tumour/heart ratio of all tracers was at least 5 times higher than the value obtained for [99mTc]-Pyrophosphate blood pool activity proving a specific tumour uptake.
Long term imaging
Twenty-four hours p.i. the tumour is clearly visible for \([^{123}\text{I}]-2\text{-I-L-Phe}\) and \([^{123}\text{I}]-2\text{-I-D-Phe}\), while for \([^{123}\text{I}]-2\text{-I-L-Tyr}\) and \([^{123}\text{I}]-2\text{-I-D-Tyr}\) tumour uptake is lower. In the case of \([^{123}\text{I}]-\text{IMT}\), the tumour can hardly be distinguished from the background. The tyrosine analogues show a clear thyroid uptake of free radiiodide which is not the case for the 2-I-Phe analogues. This indicates that the phenylalanine analogues are less susceptible to deiodination in these rats. The DUR value of the tumour at 24 hours p.i. of \([^{123}\text{I}]-2\text{-Iodo-L-Phe}\), \([^{123}\text{I}]-2\text{-Iodo-D-Phe}\), \([^{123}\text{I}]-2\text{-Iodo-L-Tyr}\) and \([^{123}\text{I}]-2\text{-Iodo-D-Tyr}\) is respectively 2.25 ± 0.85, 2.16 ± 1.12, 1.66 ± 0.50 and 1.54 ± 0.56 (data: mean ± SD (n=4)), while on the images of \([^{123}\text{I}]-\text{IMT}\) the tumour is not clear enough to allow an accurate calculation of the DUR value. At 48 hours p.i., the DUR value of \([^{123}\text{I}]-2\text{-Iodo-L-Phe}\) and \([^{123}\text{I}]-2\text{-Iodo-D-Phe}\) is 2.89 ± 0.83 and 2.69 ± 0.87, while those of the other tracers can no longer be determined.

Retention
The retention is expressed as the ratio of counts per pixel in the organ at the appropriate time to the counts per pixel in the organ at 30 minutes p.i. (decay corrected) and is represented for the tumour and the total body in table 1 (n = 12 for \([^{123}\text{I}]-2\text{-I-L-Phe}\) and \([^{123}\text{I}]-2\text{-I-D-Phe}\); n = 4 for \([^{123}\text{I}]-2\text{-I-L-Tyr}\) and \([^{123}\text{I}]-2\text{-I-D-Tyr}\)). The retention at 24 hours of the tracer in the total body is around 20% for \([^{123}\text{I}]-\text{IMT}\), but the majority of that activity is concentrated in the thyroid so the images of \([^{123}\text{I}]-\text{IMT}\) hardly yield recognizable organs or tumours anymore. The data clearly show that the phenylalanine analogues have a much higher retention, both in tumour and total body, when compared to the tyrosine analogues. Dissection data confirmed the imaging data of 2-I-L-Phe and 2-I-D-Phe.

<table>
<thead>
<tr>
<th>% Retention</th>
<th>([^{123}\text{I}]-2\text{-Iodo-L-Phe})</th>
<th>([^{123}\text{I}]-2\text{-Iodo-D-Phe})</th>
<th>([^{123}\text{I}]-2\text{-Iodo-L-Tyr})</th>
<th>([^{123}\text{I}]-2\text{-Iodo-D-Tyr})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tumour</td>
<td>24h 91 ± 10</td>
<td>92 ± 10</td>
<td>10 ± 3</td>
<td>6 ± 3</td>
</tr>
<tr>
<td></td>
<td>48h 81 ± 14</td>
<td>79 ± 11</td>
<td>ND</td>
<td>ND</td>
</tr>
<tr>
<td>Total Body</td>
<td>24h 84 ± 10</td>
<td>81 ± 9</td>
<td>15 ± 4</td>
<td>8 ± 2</td>
</tr>
<tr>
<td></td>
<td>48h 68 ± 11</td>
<td>63 ± 9</td>
<td>ND</td>
<td>ND</td>
</tr>
</tbody>
</table>

Table 1: Relative retention of radioactive tracer in the tumour, heart and total body at 24h and 48h p.i. when compared to 30 min p.i.. \([^{123}\text{I}]-\text{IMT}\) showed no measurable retention in the tumour. (ND: Not detectable)

4. Discussion
All tracers (\([^{123}\text{I}]-2\text{-I-L-phenylalanine}\), \([^{123}\text{I}]-2\text{-I-D-phenylalanine}\), \([^{123}\text{I}]-2\text{-I-L-tyrosine}\), \([^{123}\text{I}]-2\text{-I-D-tyrosine}\) and \([^{123}\text{I}]-\alpha\text{-Methyl-L-tyrosine}\)) show a fast tumour uptake and a favourable biodistribution at short times. We showed that D isomers are equivalent to L isomers with regard to tumour contrast and biodistribution, with a significantly faster clearance to the bladder in the first 30 minutes by the D-isomers.

At longer times however, there is a different pattern for the retention of radioactivity in the tumour and the body. For the iodinated tyrosine analogues the retention in the tumour was only about 10%, while \([^{123}\text{I}]-\alpha\text{-Methyl-Tyrosine}\) did not yield recognizable organs anymore, as the tracer was almost fully cleared 24 hours p.i.. A considerable deiodination could be noticed for the tyrosine analogues represented by the significant amount of radioactivity in the thyroid. No significant difference was noticed between the L and D isomers of both \([^{123}\text{I}]-2\text{-I-Phe}\) and \([^{123}\text{I}]-2\text{-I-Tyr}\).

Only the iodinated phenylalanine analogues showed a good tumour visualization after longer times. After 24 and 48 hours there was a similarly high retention of activity for both \([^{123}\text{I}]-2\text{-Iodo-D-Phe}\) and \([^{123}\text{I}]-2\text{-Iodo-L-Phe}\) in the tumour and in the body. Since the uptake in the tumour via the LAT transport system is reversible and dependent on the amount of radioactivity in the blood, the high total body and blood retention ensures that also the tumour retention is significantly higher for the Phe-analogues compared to the Tyr-analogues. This high blood retention can be due to a higher re-uptake
in the kidney coupled to a high tracer stability as no thyroid uptake could be observed for the radioiodinated phenylalanine analogues. It is important to notice that the retention in the tumour is higher compared to the retention in the total body, indicating a relative enrichment of activity in the tumour. Kersemans V. et al. already described the potential of long term imaging in a mouse model using \( ^{123}\text{I}\)-2-I-L-Phenylalanine and \( ^{123}\text{I}\)-2-I-D-Phenylalanine: at longer times the DUR values of the tumour were much higher compared to the DUR values at short times. The clearance from both the tumour and body in the athymic nude mice was much faster compared to our rat model. Samnick et al. also noticed tumour retention in human, they demonstrated that \( ^{123}\text{I}\)-4-I-L-phenylalanine shows an intense uptake in gliomas in patients up to 24 hours p.i.

The high retention of these iodinated phenylalanine analogues allows not only late imaging (due to the increasing tumour/background ratio), but it also points to the possibility of radionuclide therapy.

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5. **References**


OPTIMISATION OF PRODUCTION OF MEDICALLY RADIOISOTOPES THROUGH CROSS SECTION DETERMINATION.

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ABSTRACT

Diagnostic and therapeutic applications of radioisotopes are gaining in importance. A large fraction of the used radioisotopes are produced by charged particle reactions. Improving, optimising or innovating production pathways for efficient and reliable supply is an ongoing challenge for physicists around an accelerator.

The standard approach for gathering information allowing prediction of production yields and contamination rates is measurement of excitation functions with stacked foil irradiations and accurate gamma spectrometry.

We discuss three types of applications for production optimisation:
- comparison of pathways: which of proton or deuteron reactions is preferable for production of \textsuperscript{103}Pd from Rh targets?
- possibility to produce emerging radioisotopes: is the reaction \textsuperscript{141}Pr(d,3n)\textsuperscript{140}Nd an efficient choice?
- alternative production approaches: can fission \textsuperscript{99}Mo be replaced by accelerator routes like \textsuperscript{100}Mo(d,x)\textsuperscript{99}Mo or \textsuperscript{100}Mo(p,pn)\textsuperscript{99}Mo?

A comparison of experimental results with the reaction model based code ALICE-IPPE and thick target yields derived from excitation curves are discussed.

1. Introduction

Neutron deficient radioisotopes are increasingly used in diagnostic and therapeutic nuclear medicine, in industry and in research projects. Their production needs charged particle irradiations (CP) at accelerator sites. The CP radioisotopes are mostly produced in proton induced reactions exploiting the rather high cross sections of the processes involved and low stopping power of protons. Dedicated high beam power, cost effective and simple to operate (H, p) cyclotrons were commercially developed. Nevertheless several of these isotopes can be produced by other pathways: either other charged particle reactions (deuteron or alpha induced) or reactor production starting from other target material are possible. The choice of pathway will influence the overall batch yield and the radionuclide purity/specific activity of the desired end product. One of the ongoing tasks and challenges for a physicist or radio chemist working around an accelerator used for production of medically relevant radioisotopes is to guarantee an efficient and reliable supply to the hospital by improving, optimizing or innovating production pathways. The standard approach to allow prediction and optimization of batch yields and to choose routes that assure controlled contamination rates is accurate knowledge of the excitation curves of all (or at least most) of the nuclear reactions involved in the production process for given target/beam combinations. At present computer codes for calculation energy dependence of reaction cross sections, do not provide enough accuracy in shape and absolute values to allow production optimization. The preferred way is experimental determination of excitation functions by irradiation of stacked targets, analysis of activation products, mostly by gamma spectrometry, and calculation of the cross section at each energy point by using the classic activation formula. A comparison with results of model codes can allow better understanding of the relative contribution of individual reactions, in case of use of targets with multiple isotopes (targets of natural composition), or indicate improvements to be made to the theoretical model.

For optimization of production in itself, knowledge of excitation functions can have three main purposes: choose the most efficient charged particle to use on a given target material for a given end product,
investigate the possibility to use irradiation techniques to obtain new or poorly studied radioisotopes, explore the possibility to replace reactor based production by charged particle reactions. An example of these three applications will be discussed in detail, based on our experimental work in the past years, and a discussion of the predictive value of the ALICE-IPPE code in comparison with these excitation curves will be given.

2. Experimental methodology

Nearly all results discussed here were published earlier by our group [1-5] and were obtained in irradiations using the external proton or deuteron beams of the cyclotrons of Sendai (Japan), Atomki-Debrecen (Hungary), UCL-Louvain la Neuve or VUB-Brussels (both in Belgium). For all cross section determinations the established stacked foil technique of stacked foil, with monitoring of the beam energy and current with standard reactions for which recommended excitation functions exist [6, 7], was used.

2.1 Targets, Irradiation, measurements

Stacked foil targets consist of an assembly of thin (10 -100mg/cm²) either natural metal foils (commercially purchased) or supporting foils on which isotopically enriched material or oxides of the element to be studies are deposited electrochemically or by other techniques. The electronic stopping of the mono-energetic bombarding particles in the foils results in a gradual decrease of effective particle energy, yielding information at multiple energies for each irradiation. Insertion of monitor foils giving rise to activation products with well documented and evaluated standard cross sections allows checking and/or correction of the energy and intensity of the incoming beam, after comparison of the remeasured cross section of the monitor reaction with the recommended cross sections [6, 7].

The irradiations took place with incident energies between 15 and 80 MeV for protons and 12 and 40 MeV for deuterons, beam intensities were kept constant over the irradiation time (between 1 and 2 h) at a level of 50 to 250 nA.

The target holders acted as Faraday cups and were equipped with collimators and negatively polarised guard rings to suppress secondary electron emitted from the target material. The irradiated stacks were allowed to cool for a period of 1 to 2 h to after EOB (End of Bombardment) allow decay of the high activities (and dose rates to experimentators) of short lived radioisotopes. This procedure implies that excitation functions of isotopes with half-life shorter than 30 minutes are not determined as decay also occurs during the time needed to measure the 10-15 foils of a stack.

For isotope identification high purity Ge detectors, coupled with acquisition/analysis software, were used. Using standard calibrated sources the detector efficiencies were accurately determined over the domain of energies concerned. The measurements were started shortly after EOB and were repeated for several weeks. The activity was recorded at different detector-sample distances (5-70 cm) in order to obtain low dead times and to eliminate other possible interferences. The contribution of background signals at the different energy peaks was corrected for. No chemical separations took place.

2.2 Data processing

So called elemental cross-sections are determined from the measured activities at EOB, the number of incoming particles (derived from integrated charge) and the number of target atoms present considering the target material as mono-isotopic. The NUDAT data base was used for decay characteristics of the identified radioisotopes [8].

The incident beam energy, evaluated from the accelerator settings, as well as the initial current beam, measured with a Faraday cup, were adapted taking into account the excitation functions of the monitor reactions (\(^{nat}\)Ti(p,xn)\(^{48}\)V, \(^{nat}\)Ti(d,xn)\(^{48}\)V, Al(p,x)\(^{22,24}\)Na) measured over the whole energy domain. For energy degradation in the stack foils the codes like SRIM, based on polynomial approximations for the stopping power of the elements present, was used [9].

This technique allows determining the energy in every foil with high precision. The incident energy for the different beams was estimated with an uncertainty of ±0.1 MeV.

2.3 Uncertainty

Uncertainty on the median energy in each foil is increasing due to the cumulative effect of energy spread and variations in foil thickness and reaches a maximum of 1.5 to 2.5 MeV for the last foil of long stacks.
The uncertainties regarding cross section values were estimated according to the prescriptions of [10] and are obtained by quadratic summation of the individual contributions. The following individual uncertainties are taken into account: absolute abundance of the used gamma rays (1-5%), determination of the peak areas including statistical errors (1-5%), the number of target nuclei including non-uniformity (5%) and incident particle intensity (5%). The total uncertainty is evaluated at 8-15%. The strongly non-linear effect of the possible uncertainty of the half-lives for samples measured shortly after EOB (small T_{1/2}) was not taken into account.

3. Computer codes

The comparison of experimental results with a-priori model calculations without optimization of the parameters for individual reactions allows increasing the confidence in the predictive value of these numerical tools. In this report we show for some cases the cross sections of the relevant (p,x) or (d,x) on all stable isotopes of the target material calculated in the whole energy domain using the ALICE-IPPE code. This code is a version of ALICE-91 originally proposed by Blann [11] and last modified by the Obninsk group [12]. It is based on the hybrid, geometry-dependent hybrid or (HMS) pre-equilibrium models and the Weisskopf-Ewing evaporation formalism. The level density formalism includes both collective and non-collective effects, and excitation energy-dependent shell effects. The individual results for the reaction products of interest in a study are weighted and summed according to the abundance of target isotopes. Here we present only two calculations on mono-isotopic targets in figures 2 and 5 and are discussed together with the experimental results in the appropriate sections.

4. Excitation functions

4.1 Production of $^{103}$Pd from Rh targets

This radioisotope (0.022% γ-line at 357keV and strong low energy X-lines around 20keV, 64%), used in seeds for permanent interstitial brachytherapy, is presently produced in dedicated high power accelerators using $^{103}$Rh (100%) targets and 18 MeV protons. Older data, needing an update, existed for the (p,n) reaction and we decided to also measure for the first time the cross sections for the deuteron induced reaction. Thin, pure Rh foils (26µm for proton irradiation, 12.3 and 6.8 µm thick for deuterons) were irradiated together with Ti monitors in a total of 9 stacks. Analysis of activity showed a discrepancy between the results obtained from γ-lines and from X-rays even if self-attenuation of the X-rays is corrected for. The results are published in [1] and [2] and showed cross sections for the deuteron reaction that are up to 50% higher than for the proton induced reaction (see Fig. 1 and 2). The thick target yields derived from the fitted experimental excitation function (based on the X-ray results) agree well with those measured earlier and are nearly double at 20 MeV incident particle energy compared to those found for a proton irradiation (Fig. 3). Fig. 2 shows that the results of the theoretical calculations agree well with the experiments, which is certainly not always true for deuteron induced reactions, especially when stripping is involved.

4.2 Production of $^{140}$Nd from $^{141}$Pr

The radioisotope $^{140}$Nd decays by pure electron capture and without emission of measurable γ-rays to the β-emitter $^{140}$Pr. The $^{140}$Nd/$^{140}$Pr pair is gaining interesting for possible combination of therapeutic and imaging properties in a single radioisotopically labelled compound. Production by the $^{141}$Pr(p,2n)$^{140}$Nd route was studied earlier by Steyn et al. [13] In order to evaluate the possible advantage of the $^{141}$Pr(d,3n) reaction several stacks of Pr$_6$O$_{11}$ targets, deposited on Al or Ni backings (Julich) were irradiated in 20 and 40MeV deuteron beams (see also [3]). The monitoring relied on the standard natTi(d,x)$^{48}$V reaction. Assessment of induced $^{140}$Nd activity was done through measurement of the annihilation radiation of the daughter $^{140}$Pr. This approach asks, in the experimental condition used, for corrections for 511 keV signal from $^{140}$Nd, $^{48}$V, $^{56}$Co. Excitation functions for $^{139}$m$^{+}$Nd, $^{141}$Nd, $^{140}$Nd, $^{142}$Pr, $^{139}$Ce (high cross sections, Fig. 4) and $^{137}$Ce, $^{135}$Ce, $^{140}$La were determined. Presence of $^{139}$Pr was detected but too low counting statistics prohibits reliable cross section calculation. No earlier data are available for these reactions.

$^{140}$Nd batch production could be interesting with “dedicated” 30-40 MeV deuteron machines, radionuclidic purity has to be achieved by decay of shorter lived co-produced $^{138,139m,141}$Nd. The thick target yield derived from the excitation function is 250-300MBq/µAh (comparable to the p-induced route: overview in Steyn et al. [13]: optimal range 30-15 MeV, practical TTY 275 MBq/µAh).
4.3. Production of $^{99}$Mo from $^{100}$Mo

A vast majority of nuclear medicine investigations is presently relying on the $^{99}$Mo-$^{99m}$Tc generator where $^{99}$Mo is a fission product recovered from research reactors. In the 90’s progressive closure of these reactors was foreseen and possible accelerator production was studied. In [4, 5] we presented our results for proton and deuteron induced reactions on $^{100}$Mo resulting in $^{99}$Mo (figures 5, 6).

The advantages of the (d,p2n) over (p,pn) reaction can clearly be seen on figs. 5 and 6 as the cross sections for d induced are more than double compared to p induced but the maximum occurs at higher energy. As both excitation functions remain high over a broad energy range higher energy deuteron machines are needed to take advantage of the increased TTY (fig. 7). An upper limit is imposed by possible contaminants production, especially $^{96}$Tc reached by (d,6n) with $Q = -35.6$ MeV. In optimal conditions TTY could be 16 MBq/µAh resulting in daily batches (20 hours irradiation) at a dedicated high power machine (1 mA beam current) of $3.2 \times 10^{11}$ Bq to be compared with the weekly needs of $8 \times 10^{12}$ Bq in Belgium. Direct distribution of $^{99m}$Tc obtained by (p,2n) and (d,3n) on $^{100}$Mo or by $^{98}$Mo(d,n) could be other alternative. This would not require recycling of the expensive enriched target material but needs rethinking of the distribution pathways and introducing new radiopharmaceutical registration files.

Fig. 5 shows that, although the shape of the excitation function is well modeled by ALICE-IPPE calculations (data stored in MENDL2 database), the absolute values are underestimated by a factor 3.
Fig. 5 Excitation function for $^{100}\text{Mo}(p,x)^{99}\text{Mo}$ (figure taken from [4])

Fig. 6 Excitation function for $^{100}\text{Mo}(d,x)^{99}\text{Mo}$ (figure taken from [5])

5. Conclusions
Measurement of excitation functions allows to prove that for several reactions leading to production of medically relevant radionuclides, deuteron induced channels have higher thick target yields for the same incident energy than proton reactions. This should be an incentive for developing dedicated high power commercial 30 MeV deuteron accelerators. The planned medium energy, high intensity deuteron accelerator dedicated to neutron production (IFMIF) could hence be very effective in medical radioisotope production.

The agreement with theoretical calculations shows often large discrepancies especially for stripping reactions. Upgrades and improvements are being developed.

References
OPTIMIZING INJECTED DOSE IN EQUILIBRIUM GATED RADIONUCLIDE VENTRICULOGRAPHY BY USING A THREE CLUSTER MODEL AND LINEAR DISCRIMINANT ANALYSIS

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ABSTRACT

A mathematical model of three clusters is used to analyse differences in Equilibrium Gated Radionuclide Ventriculography image quality. The objective dependent variable, used to characterize image quality was Image Contrast. It was obtained from the Count ratios between Heart and Background in some Regions of Interest. Noise and Spatial Resolution were also analysed as part of Image quality evaluation. The aim of the present research was the determination of the minimum 99mTc-red cells activity which is able to guaranty enough diagnostic image quality in a traditional gamma camera. The method was applied in 30 patient’s studies, injected with one of the following activities: 300, 440, 620, 800 or 1020 MBq, using the "in vivo" labelled method of red cells previously stimulated with pyrophosphate. The statistical weight of each image quality determinant variable was obtained using linear discriminant analysis. The included variables were: the radionuclidic activity, the labelling yield, the weight of the patient, the acquisition time and the patient pathology. The cluster with optimum image quality was selected. The study showed that 800 MBq is a optimised activity for the technology used and Cuban patient characteristics.

1. Introduction

Patient exposure to ionizing radiations in Nuclear Medicine studies can not be indefinitely reduced without affecting image quality. The above problem is the cause of the necessity to establish a compromise relationship between image quality for diagnosis and radiation protection for the patient. It means that the activity should not exceed the minimum required to provide the indispensable clinic information. [1].

IAEA recommends some reference values for each Nuclear Medicine study [2]. Nevertheless, IAEA subjects to adjust these values to the practical reality of each country (equipment, radiopharmaceuticals, patient characteristics).

Gated radionuclide ventriculography [3] presents many advantages for the diagnostic of several cardiac diseases [4]. Therefore, there is not good consensus among doctors and medical physicists respect to the best activity value for this purpose [5-12].

A model of two clusters combined with linear discriminant analysis of image quality has been previously published as optimisation method in Nuclear Medicine [13]. The present work proposes the use of a three cluster model looking for a best precise analysis of image quality as well as best case classification inside clusters, in order to justify the adequate radionuclidic dose selection according to the radiopharmaceutical and equipment used and patient characteristics.

2. Material and Methods
2.1 Experimental Procedure
An injection of 10 mg of pyrophosphate was administered to 30 patients (16 women and 14 men, age between 28 and 80 years, mean 55 ± 3 years). Twenty minutes after they received a dose of Tc$^{99m}$ (divided in 6 groups, Group A: 303±6 MBq, Group B: 444±6 MBq, Group C: 617±7, Group D: 803±10 and Group E: 1020±5 MBq). The labelling of patient red cells was “in vivo” [14]. The main labelling yield was verified by laboratory method [15]. Ten minutes after each patient is placed under gamma camera detector in LAO position, (45° detector inclination). Radioactivity is synchronised to the R wave of electrocardiography signal obtained in CM5 derivation. Sixteen images were acquired for each cardiac cycle. Cycles with more than 10 % of difference with the mean cardiac frequency were rejected. A Toshiba digital gamma camera (single head, model 500A, with RDC-44A parallel holes general purpose collimator was used. The acquisition matrix was 64 x 64 pixels and the energy window was cantered in 140 keV ± 10 %.

2.2 Variables
Region of Interests (ROI) 5 x 5 pixels were traced over left ventricle, liver, lung, spleen and a background zone under the left ventricle and upon the liver in diastolic image. (Fig. 1). ROIs were processed by the method of the second derivative [16]. Background was automatically corrected [16].

![Diastolic image](image)

The variables used to characterize image quality in terms of Image Contrast were Count ratios between Useful signal in Heart respect to Organ or Background activity. They were Heart/Lung H/L, Heart/Background H/B, Heart/Spleen H/S and Heart/Liver H/L.

Other (Xi) variables were also analysed as: cardiac frequency, left ejection fraction, Weight of the patient (W), height, age, ejection time, accepted and rejected cycles, labelling efficiency (EM) and the acquisition time, looking for if they had influence over image quality. The subjective opinion about image quality of an observer, blinded to the activity used, was also taken into account.

2.3 Mathematical Procedure
We follow the k means cluster model (order 3) and linear discriminant analysis [17]. If there are significant differences among variables belonging to the constructed clusters, then, we can affirm that each cluster owns different image quality according to the Xi variables measured [18,19].

As optimisation criterion was taken the lowest activity which permits the inclusion of cases in the cluster with the best image quality [18, 19]. Others regression and correlation analysis were also applied in order to corroborate the results obtained.

3. Results and Discussion
Image quality is usually analysed in terms of physical parameters as: Image Contrast, Statistical Noise and Spatial Resolution [20]. The same gamma camera with a fixed distance patient-detector and the same acquisition matrix 64 x 64 pixels was used for all the cases included. For this reason we consider fixed the Spatial Resolution in our experiment.

Image Contrast was defined as: (Counts in Useful Signal – Background Counts) / Background Counts x 100 %, using ROIs while Signal/Noise ratios were taken as (Useful Signal ROI)$^{1/2}$. Then, as both variables are strongly correlated we took Signal/Background ratios as quantitative criterion of image quality to conform the clusters. This variable is dependent on the administered activity and the study stopping time (taken at 300 kcounts in our experiment). As specific indexes of image contrast we took H/B, H/S, H/L and H/P.
The expert observer graded all the images as good image quality, without distinction among activities used. Nevertheless, mathematically and taken into account the Xi variable measured, 3 different clusters were constructed after 3 iterations, instead of only one, as the observer describe. After linear discriminant analysis we obtained that Image Contrast was only determined in clusters by: EM, Weight, Systolic counts, Activity and Acquisition time, with 96.7 % of cases correctly classified into the clusters. The selected variables were significant taking into account their $\lambda$ Wilks’s values, higher than 0.9. EM p=0.642, W p=0.903, Syst Counts p=0.555, A p=0.155 and Acq t p=0.195.

Two linear image quality discriminant functions were constructed. The first one detects changes between Cluster 1 and Cluster 2 and the second one between clusters 2 and 3. The canonical correlation values are $r=0.454$ and $r=0.256$ respectively.

$$H/B = 0.16 \text{EM} - 0.41 W + 0.466 \text{Syst Counts} + 0.975 A - 0.042 \text{Acq t}$$  \hspace{1cm} (1)

$$H/B = 0.416 \text{EM} - 0.183 W + 0.810 \text{Syst Counts} - 0.255 A + 0.056 \text{Acq t}$$  \hspace{1cm} (2)

Both functions were significant. $\lambda$ Wilks _1 = 0.742 p = 0.709 and $\lambda$ Wilks _2 = 0.934 p = 0.504.

The only variable dependent on patient disease introduced in the functions was the Systolic Counts. The rest were eliminated due to multicolineality (High correlation coefficients with the selected variables) [17]. Fig. 2 shows the clusters in relation with the two functions.

![Canonical Discriminant Functions](image)

**Fig. 2. Cases selection by clusters and functions**

The cluster with the best image quality was cluster 3 (include the cases with the best H/B ratios). Table 1 shows the correlation coefficients of the selected variables in relation to H/B. The importance of these values is that each coefficient establishes the statistical weight of each variable to distinguish clusters with differentiated image quality [17-19].

<table>
<thead>
<tr>
<th>Variable</th>
<th>Correlation Coefficient with function 1</th>
<th>Correlation Coefficient with function 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>0.698</td>
<td>0.628</td>
</tr>
<tr>
<td>Acq t</td>
<td>0.658</td>
<td>0.550</td>
</tr>
<tr>
<td>EM</td>
<td>0.330</td>
<td>0.300</td>
</tr>
<tr>
<td>Syst counts</td>
<td>0.062</td>
<td>0.803</td>
</tr>
<tr>
<td>W</td>
<td>0.145</td>
<td>0.186</td>
</tr>
</tbody>
</table>

Tab. 1 Correlation coefficients with image quality discriminant functions

The above table expresses clearly that in this type of study the activity is the most important variable for image quality, following by the Acquisition Time and the Labelling Efficiency. These results show similarity with others discriminant studies of image quality in Nuclear Medicine [21-23]. Nevertheless, although the weight of the patient reveals some influence over image quality, this
variable was not very important for image quality discrimination, which has different behaviour respect to others Nuclear Medicine Studies [24].

Fig. 3. provides information about the variables behaviour to determine H/B ratios. A high dispersion is observed around the linear behaviour for almost all the variables, except for the weight, whose range included in this experiment was very similar and typical of Cuban population. Systolic counts neither had significant variations for the sample analysed. This variable was less determinant for image quality than the rest of the analysed variables.

\[
y = 118.03x + 359.36 \\
R^2 = 0.1205
\]

\[
y = -45.136x + 436.19 \\
R^2 = 0.0925
\]

\[
y = 0.7354x + 90.534 \\
R^2 = 0.0561
\]

\[
y = 1.303x + 64.731 \\
R^2 = 0.0195
\]

\[
y = -344.7x + 20976 \\
R^2 = 0.0039
\]

Fig. 3. Variables behaviour in relation to Image quality

Table 2 shows the case distribution by clusters and the correspondence with the activity administered. The activity with the best results from the point of view of H/B, H/L, H/P and H/S was 803 MBq (most of the cases are included in cluster 3). This result subjects that activities higher than 800 MBq saturates the detector for the technology used, diminishing its sensitivity and consequently Signal/Background ratios.

<table>
<thead>
<tr>
<th>Cluster 1</th>
<th>Cluster 2</th>
<th>Cluster 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cases 1, 2 and 5 (303c MBq), Case 7 (444MBq) and Cases 13, 14, 16 and 18 (617 MBq)</td>
<td>Cases 3, 4 and 6 (303 MBq), Cases 8,9,1 and 12 (444 MBq), Cases 15 and 17 (617 MBq), Cases 21 and 24 (803 MBq) and Cases 26 and 29 (1020 MBq)</td>
<td>Cases 19, 20, 22 and 23 (803 MBq), Case 10 (444MBq) and case 27 (1020 MBq)</td>
</tr>
</tbody>
</table>

Tab 2. Case distribution by clusters

Fig. 4 shows the Heart/Background ratio relative to Heart/Background maximum vs. the normalised activity respect to the activity that permits the highest H/B ratio. This result corroborates the results of table 2, after the three cluster model application. The value of 800 MBq is obtained as the optimised activity for the acquisition technology and radiopharmaceutical used, and the patients characteristics sampled.

Fig. 4. Curvas de respuesta relativa para 30 estudios de ventriculografía nuclear gatillada en reposo.
The activity obtained as optimum was coincident with previous results for this type of Nuclear Medicine study in Cuba [13]. In the referenced study was used a model with lower clusterisation dimension. The method of two cluster combined with discriminant analysis to optimise radionuclidic activity has been successfully also proved in other studies of Nuclear Medicine [24] and it has been validated by comparison with the well-known ROC analysis [25]. The present work shows how the increment in the cluster number increases the classification precision but did not change the optimised activity value selected. Special attention is required respect to the optimisation of the clusterisation dimension for futures works.

4. Conclusions

The use of a combination of three clusters and linear discriminant for mathematical image quality determination is a useful tool to optimise activity in Gated Radionuclide Ventriculography. The method is sensible to small variations in Image Contrast, not detectable for expert observers. The method is more precise than the model of two clusters. The value of 800 MBq was reported as the minimum radionuclidic activity tested for obtaining good image quality with the technology and radiopharmaceutical used.

5. References

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THERMAL FLUX ANALYSIS OF THE PFMA-1 DEVICE

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ABSTRACT

This work presents experimental results pertaining to the heating by plasma-wall interaction of the upper flange of the vacuum chamber of the PFMA-1 Plasma Focus at 18 kV and at 1 Torr of $^4$He. The 2D temperature distributions over the flange have been obtained using an infrared camera. From these distributions average temperature profiles are obtained and used to estimate the heat imparted to the flange by plasma-wall interaction. It is found that at least 15% of the input energy is converted into thermal energy of the flange. The heat flux has been estimated too; it is found that its average value is comparable to that for ITER in the cases of plasma disruptions. It is therefore proposed that a powerful PF can be used similarly to plasma guns and e.m. railguns to test materials for fusion reactors. Other studies are under way to correlate the model presented to the experimental results obtained with an aluminium disk which resulted ablated due to the interaction with the plasma.

1. Introduction

One of the various issues that has to be overcome along the pathway to the reliable, industrial applicability of the Plasma Focus (PF) technology in the repetitive regime is the management of the thermal loads to the components of a given device. Order of magnitude estimates predict that about 98% of the input energy imparted to a device in a shot is, at equilibrium, directly converted into thermal content of its parts. The largest fraction of the thermal loads is most probably generated and deposited in the vacuum chamber region, where the plasma is formed and pinched. Experimental and/or theoretical data about the mechanisms by which energy is deposited into the materials of PFs are very scanty, as well data about the cooling of these devices [1]. Small scale PFs, devices with input energy below about 1 kJ, have been produced with shot repetition frequencies up to 10 Hz and operated for some minutes without the need of cooling apparatuses [2]. Many nuclear reactions that are triggered in the pinched plasma of PFs are known to scale with a given power law of the input energy, most often the exponent being $\approx 2$. This means that a careful optimization design has to be carried out in choosing between a low input energy device with high repetition rates, or a high input energy device with rather low repetition frequencies. Since the nuclear yields scale with a more-than-linear power law and since it is quite difficult to raise the repetition frequency of powerful devices due to problems in the switching technology, it is often convenient to increase, for output efficiency purposes, the input energy rather than the repetition rate. An increase by a factor four in the output can be obtained either by enhancing the input energy by a factor two, or by enhancing the repetition frequency by a factor four; this second option however implies an increase by a factor four in the heat loads, whereas in the first instance the heat loads are increased by a factor two only. The most important problem related to the thermal analysis of repetitive PFs is the distribution of the heat loads; a previous experimental study [1] indicated that about 10% of the input energy is imparted to the inner electrode in a positive polarity device; in this study the energy released to the upper flange of the vacuum chamber of PFMA-1 is evaluated and reported from experimental evidence.
2. PFMA-1 and experimental setup

PFMA-1 [3,4] is a 150 kJ Mather-type PF designed for a repetition frequency up to 1 Hz and dedicated to the neutron-free endogenous production [5] of $^{18}$F, a short-lived radioisotope used in the preparation of FDG drugs used in PET medical examinations. A photograph of the device is given in Fig. 1. Theoretical estimates predict a yield of about 1 Ci of $^{18}$F in two consecutive hours of repetitive operation at 1 Hz. The device has a total equivalent inductance of about 40 nH, included the contribution of the cylindrical electrodes; its input energy is stored in a 350 μF capacitor bank charged at 30 kV. The vacuum chamber is filled with $^{16}$O and $^3$He at a total pressure of about 10 Torr. Peak total current is predicted by 2-D Snowplow calculations to be about 1.5 MA. Active cooling of all the system is achieved with closed-circuit SF6 flow, open-circuit dry air, closed-circuit deionized water and partially closed-circuit demineralized water refrigerated by a 150 kW evaporative cooling tower. The temperature distributions over the external surface of the upper flange, Fig. 2, of the vacuum chamber have been obtained with a Flir Thermacam PM 675 infrared camera. The flange is entirely made of AISI 304L stainless steel and has some ports that connect the vacuum chamber to various diagnostics as well as to the vacuum system. A special 3M black tape was used to cover entirely the area to be investigated, so that no reflections in the visible spectrum due to the very flat surface could affect the measurement; a specific cross-calibration for obtaining the emissivity coefficient of the black tape was done with the use of a contact thermocouple; error in the temperature measurements is estimated to be +/- 0.3 °C. The infrared images were taken with about 25-30 s delay after each shot, which is about 4-6 times the temperature relaxation time in the thickness of the flange (estimated at first approximation as $s^2 \rho c_p / \pi^2 k$, $s = 25$ mm being the thickness, $\rho$ the mass density, $k$ the thermal conductivity and $c_p$ the specific heat); this is the time the temperature gradient along the thickness of the flange takes to become so small that the measured external surface temperature $\tilde{T}(r)$ can be practically considered coincident with the average temperature:

$$\tilde{T}(r) \equiv \frac{1}{s} \int_0^s T(r, z) dz$$

(1)

$T(r,z)$ being the real temperature distribution after a shot. The cooling of the flange can be neglected because the experimental evidence showed that 25-30 s are not enough to modify substantially the radial profiles.

The radius of the flange is 17.5 cm. Its mass is estimated as being that of a full and perfectly circular plate flange without holes for ports, to compensate for the extra mass of the ports themselves; it results 18.9 kg.
The distance between the pinch point and the flange is about 14 cm. The experiments were carried out at 18 kV charging voltage, which corresponds to an input energy of 54 kJ, and 1 Torr pressure of $^4$He. The time to plasma-pinch is about 2 $\mu$s, and the whole half-period of the discharge lasts about 10 $\mu$s. Shots were performed every 4-5 minutes and no external cooling apparatuses were connected.

3. Experimental results and discussion

Fig. 3 shows a typical postprocessed image taken with the infrared camera; the point of maximum heating is at the center of the flange. This is due to two mechanisms: plasma-wall interaction and ion impinging from the post-pinch MHD particle emission in the forward direction (this being the direction toward the geometrical center of the flange). The contribution of the first mechanism is probably the most significant for the energy deposition, as will be shown below. Fig. 4 is the plot of the isothermal lines from Fig. 3; it shows rather uniform gradients in the radial direction. Both figures are plotted against the matrix indices by which spatial information is stored in the infrared camera.

Fig. 5 shows in the upper row the temperature profiles $\tilde{T}_n(r)$ along the dashed line drawn in Fig. 3, after

Fig 3. Infrared camera picture.                                 Fig 4. Isothermal lines from Fig 3.

Fig 5. Upper: temperature profile sequence. Lower: average incremental temperature profile for a single shot.

Fig 6. Internal side of flange, with circular aluminium plate before and after (upper left corner) 40 shots.
each shot of a series of 11 shots; it is clearly seen that a difference of about 5-6 °C exists between the center of the flange and its borders. In the lower row is plotted \( \Delta T(r) = (\bar{T}_{n+1}(r) - \bar{T}_n(r)) \), the average profile of temperature increase between two consecutive shots, as can be determined from the profiles of the upper row; ± 1σ error bars are also shown. It was also determined that a series of 20 shots is capable of generating a temperature increase of about 18 °C above room temperature in the center of the flange. The amount of energy \( Q \) deposited in the flange per shot can therefore be estimated as

\[
Q = \frac{mc_p}{2\pi \sigma^2} \int_0^R \Delta T(r) 2\pi r dr
\]

\( R \) being the radius of the flange, \( m \) its mass and \( c_p \) the specific heat of stainless steel. It results \( Q \approx 8 \) kJ.

This corresponds roughly to 15% of the whole input energy stored in the capacitor bank; such a large amount of energy can only be transmitted by the direct contact with the plasma. After about 200 shots it had also been found that a large circular area of the inner surface of the flange resulted eroded or ablated. This area is rather well defined; Fig. 6 shows the ablated area delimited by a white circle. It was therefore decided to investigate more deeply this phenomenon. An aluminium circular plate or disk (diameter 15.1 cm, thickness 1.6 mm) was installed in the peripheral part of the flange and exposed to 40 shots. After exposure it was found to be heavily eroded or ablated in an area that roughly corresponded to that of the stainless steel (inset in the upper left corner of Fig. 6). Gaussian shapes in \( r \) and \( t \) are assumed for the heat flux:

\[
F(r,t) = Ne^{-\left(\frac{r}{\sigma}\right)^2} e^{-\left(\frac{t}{\alpha}\right)^2}
\]

where \( N \) is a normalization constant, \( \sigma \) is taken to be \( R/3 \) and \( \alpha \) is taken to be 0.1 \( \mu \)s (0.6 \( \mu \)s is assumed to be the duration of the flange heating phase by plasma interaction). The normalization constant can be determined by:

\[
Q = \int_{r=0}^{R} \int_{t=-\infty}^{\infty} F(r,t) 2\pi r dr dt
\]

It results \( N \approx 4\times10^{12} \) W/m². The average heat flux is found to be:

\[
F = \frac{1}{6\pi \sigma \alpha} \int_{r=0}^{R} \int_{t=-\infty}^{\infty} F(r,t) 2\pi r dr dt = 1.4\times10^{11} \) W/m². This value is of the same order of magnitude as that expected in ITER in the case of plasma disruption [6]. Therefore it can be suggested that a powerful PF can be used, likewise plasma-guns and electromagnetic railguns [7], to test fusion reactor components and materials. In Fig. 7 it is plotted the ablation threshold of aluminium as well as the profile

![Fig 7. Q''(r) over the flange and Q_{th}'' for aluminium.](image_url)
\[ Q''(r) = \int F(r, t) dt \] along the radial dimension of the flange. It is seen that the local heat flux results higher than the threshold in an area that corresponds roughly to that seen in the experiments (see Fig. 6). The ablation threshold has been calculated as \[ Q''_{\text{th}} = \Omega \sqrt{\rho k / c_p} \sqrt{\tau} \approx 22.2 \text{ J/cm}^2, \] \( \Omega \) being the vaporization specific enthalpy, \( \rho \) the mass density, \( k \) the thermal conductivity, \( c_p \) the specific heat and \( \tau \) the time-duration of the heat flux (taken as 6\( \alpha \)) [8].

This formula is valid for the nanosecond or longer energy deposition-times, this regime being typical of PFs. Moreover Vorobyev and coworkers [9] have shown that energy deposition to aluminium with ambient pressures higher than 0.1 Torr (the case of PFMA-1) is independent from the specific ambient gas, and that the energy coupling is maximal, with residual energy coefficient between 0.8 and 1, so that the estimated value of \( Q \) is not only the residual heat in the flange, but is also very close to the whole heat imparted to the flange before ablation itself.

Further studies are currently on the way to correlate the ablation rate of aluminium with the observed change in mass of the disk and with the energy deposited per shot.

4. References

NEW PERSPECTIVES FOR MEDICAL APPLICATIONS USING THE TRIGA MARK I IPR-1 RESEARCH REACTOR

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ABSTRACT
This work presents the more recent enhancements of the TRIGA MARK I IPR-R1 research reactor located at Nuclear Technology Development Centre/Brazilian Commission for Nuclear Energy (CDTN/CNEN) focusing medical applications. It is presented the preliminary results of the preparation of some specific molecules of platinum compounds to be used as radiotracer in pharmacokinetical studies as well for therapeutic purposes as antitumoral or antibiotic.

1. Introduction
Research reactors have contributed to the development of the nuclear science and technology for the past 50 years, but the new discoveries and innovations need newer tools and more powerful reactor with special attributes. Even though many research reactors worldwide are underutilized and many older ones will be shut down next years, the need for research reactors in not waning. According to a recent diagnostic of a survey performed by IAEA, many research reactors worldwide, as is the case of TRIGA, are underutilized due to several factors, mainly due to the inefficient communication owner-client of the reactor [1,2]. The starting up of the TRIGA MARK I IPR-R1 research reactor in 1960 marked the beginning of the activities of the Nuclear Technology Development Center (CDTN) one of the five institutes of the National Commission of Nuclear Energy, (CNEN)\(^3\). Since then, the reactor has been used mainly for neutron activation analysis and training of the reactor operators. The TRIGA IPR-R1 CDTN’s research reactor is of the type MARK I, where the core is below the floor level, as it is shown in Figure 1 [3,4].

Figure 1 - Views of the well (left) and the core of the TRIGA IPR - MARK 1 reactor [5].
More recently, new research projects were initiated to enhance the reactor utilization. Since 2001, special labelled molecules, to be used in biodistribution experiments for the investigation of new drugs design, have been obtained [6]. Brazilian gemstones, specially topaz, are being irradiated to improve the color [7,8] and neutron radiography images a limited to the samples of small size have been taken [9]. The reactor is operating at 100kW but the actual configuration of the core allows the increasing of the power up to 250kW.

Specially during the last five years, results of researches involving new drugs or new strategies of administration of drugs already in the market opened a very interesting and important field of application for the TRIGA reactor: the labelling of special molecules to be used in biodistribution and for pharmacokinetical studies in vitro and in vivo. The first compound irradiated in the TRIGA with this purpose was the CDDP, cis-dichlorodiammineplatinum (II), Pt(NH$_3$)$_2$Cl$_2$. CDDP is an effective chemotherapeutic largely used to treat systemic tumours in several organs: testicles, ovary, head, neck, bladder. However, its side effects are serious principally the nephrotoxicity [10-14]. Investigation of new formulations containing CDDP to minimise or eliminate these effects is extremely relevant. A detailed description of irradiation of CDDP by CDTN and its applications are described elsewhere [6].

Besides CDDP, in the last few years new compounds of platinum and other metals with antitumoral and/or antibiotic activity has being investigated as, tetracycline-platinum II (Tc), Pt(C$_{22}$H$_{24}$N$_2$O$_8$)Cl$_2$ complex. Tetracycline is one of most important antimicrobial agents with broad spectrum but its low toxicity, low cost and oral administration led to and indiscriminate use and appearance of bacterial resistance. Previous studies performed with Tc, showed a more pronounced antibacterial effect compared to tetracycline [15]. More recent results obtained by our group showed also and enhancement of the antitumoral activity of the radiolabelled Tc. Further in vivo studies must be performed to confirm this important potential use of Tc. Once a chemotherapeutic agent is radiolabelled it is interesting to investigate the possible synergic effect of its chemotherapy and radiotherapy actions. The preliminary results obtained using the radiolabelled platinum compounds of in vitro studies showed very interesting results [16].

2. Material and Methods

The development of new medicines or a new formulation of those already existent for clinical use requires many stages of assessment such as biodistribution, pharmacokinetics and toxicity studies. The evaluation of in vivo biodistribution using radiolabelled compounds has many advantages compared with others available analytical methods, such as, spectrophotometry of atomic absorption which requires laborious sample treatment, and consequently, longer time for the analysis. In the case of platinum compounds it may allows, in the future, to investigate its potential clinical use in postoperative treatment using gamma imaging systems already. The radionuclides produced by irradiation of Pt compounds CDDP and Tc are showed in Table 1 [6].

Table 1. Main radionuclides produced by platinum compounds after irradiation and nuclear data.

<table>
<thead>
<tr>
<th>Stable Nuclide</th>
<th>Natural abundance (%)</th>
<th>Nuclide Produced</th>
<th>Half-life</th>
<th>γ energy in keV, (abundance %)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{100}$Pt</td>
<td>0.01</td>
<td>$^{191}$Pt</td>
<td>2.96d</td>
<td>359.93 (6), 409.48 (8), 538.9 (13.7)</td>
</tr>
<tr>
<td>$^{100}$Pt</td>
<td>0.79</td>
<td>$^{193m}$Pt</td>
<td>4.33d</td>
<td>-----</td>
</tr>
<tr>
<td>$^{194}$Pt</td>
<td>32.9</td>
<td>$^{195m}$Pt</td>
<td>4.02d</td>
<td>30.8 (2.3), 98.8(11.4), 129.7 (2.8)</td>
</tr>
<tr>
<td>$^{196}$Pt</td>
<td>25.3</td>
<td>$^{197}$Pt</td>
<td>18.3h</td>
<td>279.11(2.3), 191.36(3.7)</td>
</tr>
<tr>
<td>$^{198}$Pt</td>
<td>7.2</td>
<td>$^{199}$Pt</td>
<td>30.8m</td>
<td>317.06 (4.87), 493.74 (5.7), 542.9 (14.8)</td>
</tr>
<tr>
<td>$^{37}$Cl</td>
<td>24.2</td>
<td>$^{38}$Cl</td>
<td>37.24m</td>
<td>1642.69(31.0), 2167.68(42.0)</td>
</tr>
</tbody>
</table>
The direct irradiation of Pt compounds is undoubtedly the most simple procedure in spite of its limitations related to maximum specific activity, chemical and radiochemical purity to be reached. It occurs due to the breaking of Pt-NH₃ and Pt-Cl bonds as it showed in Figure 1, by the recoil of the excited nuclei of Pt and Cl, - the Szillard-Chalmers effect - and by the collision with high energy neutrons [17,19]. Thus, the preparation of Pt compounds showing an high specific activity, good chemical and radiochemical purity by the direct irradiation process is important in order to optimise the in vivo biodistribution studies, avoiding eventual corrections of decay time in the activity for each sample and also allowing the assessment of its potential clinical use with gamma imaging systems.

Figure 1. Molecular structures of CDDP (left) and tetracycline-platinum (right) [15]

2.1 Irradiation and Gamma Spectrometry
CDDP was initially irradiated under the flux of average thermal neutrons of 6.4 x 10¹¹ n.cm⁻².s⁻¹ at 100 kW for 2, 3, 4, 6 and 8 hours. After 4 hours of irradiation the sample turned from yellow, its original colour, to grey, the colour of elemental platinum, confirming the molecule degradation, as already expected. As some platinum isotopes have high integrals of resonance related to the thermal cross sections, the CDDP samples covered by a Cd capsule of 1 mm thickness were irradiated in order to obtain an higher specific activity with acceptable chemical and radiochemical purity. Tc sample was initially irradiated by 8h covered by Cd capsule but the best results of antitumoral activity performed using cells of leukaemia was obtained with bare sample of Tc irradiated by 2h. Any evidence of disruption of the Tc molecule was verified for both conditions of irradiation. The determination of radiochemical purity for CDDP and Tc are described elsewhere [6, 20] Gamma spectrometry was applied in a HPGe detector with 15% nominal efficiency and 1.85 keV FWHM for 1332 keV peak of ⁶⁰Co GENIE 2000, CANBERRA software was used for data preparation and spectrum analysis.

3. Results and Discussion
3.1 Irradiation of Pt compounds
In the Figure 2, it is showed the gamma spectra of bare samples and Cd-covered samples irradiated for 2 hours, after the decay time of 30 min. It can be observed that the spectra are similar up to 300 keV, but above this limit, photpeaks of ¹⁹¹Pt of 359.6 keV, 409.4 keV and 538.9 keV were detected in the bare sample but not in the spectrum of the sample irradiated with cadmium. This result can be expected according to the values of $(\sigma_{th})$ and RI of $^{191}$Pt [6].

Figure 2. Pt-compounds spectra of 2 h of irradiation: upper: bare sample; lower, Cd-covered sample.
3.2 Radiation counting in mice
Mice were used to evaluate the biodistribution of CDDP after its administration. The results of detection of radiation of CDDP* in liver are shown in the Figure 3. The liver was collected and submitted to the gamma spectrometry using the well-type NaI detector, of 20% efficiency. The measurement time was of 2 min. The irradiation time of 8 hours allowed to a great level of detection.

![Gamma counting of CDDP in the liver of mice samples](image)

Figure 3. Gamma counting of CDDP in the liver of mice samples [21].

The specific activity was of 11.5 kBq.mg⁻¹ for the bare sample irradiated during 2 hours and approximately 57 kBq.mg⁻¹ for the Cd-covered sample during 8 hours.

3.3 In vitro-effects of radiolabelled CDDP
Figure 4 shows the comparative results of the antitumoral effect against GH3 tumoral cells between irradiated and non irradiated CDDP. This result is very important and open new interesting perspectives for the TRIGA IPR1 research reactor.

![Effect of the irradiated and irradiated CDDP over the metabolism of tumoral GH3 cells](image)

Figure 4. Effect of the irradiated and irradiated CDDP over the metabolism of tumoral GH3 cells.

Preliminary investigations of citotoxicity of the radiolabelled Tc against cells of human leukaemia K5562 showed an enhancement of 22 times if compared with similar results obtained with non irradiated Tc. With the same purpose, new antitumoral metallic complex of Cu and Sn has been investigated and will be published briefly.

4. Conclusions
Results obtained through irradiation in the TRIGA reactor operating at 100kW open new and promising prospects to enhance the use of the reactor for research and development of new drugs or new formulation of those existent ones. Optimistic prospects for radiolabeled drugs production open new possibilities for its use in biological research at CDTN/CNEN and partners confirming the
importance of nuclear area as an indispensable research tool.

Acknowledgements
The authors would like to thank the FAPEMIG, CNPq for their financial support and also colleagues of the Reactor and the Irradiation Service of CDTN: F.M. Júnior, A. Z. Mesquita, P.F.de Oliveira, L.O.I.S. Câmara and A. Amaral, for their valuable technical co-operation.

5. References
Persons, working in ionising radiation (IR) environment are exposed to continuous radiation with low doses and low dose rate. The assessment method of the Probability of Causation (PC) provides possibility to determine the causative connection between the occupational exposure and the consequent radiation-dependant cancer. An analysis has been carried out to 27 people. The probability for such a connection depends on the duration of the employment period in an IR environment, on the cumulative dose for that period, age and sex and the latent period for the corresponding localization. Data have been processed with a software product “Survrad”.

For 24 of the cases there has been established no causative connecting between cancer disease and a foregoing occupational radiation. In 3 of them a 1% (0.0-0.02) PC is found. The present study does not find out a causative connection between cancer and a foregoing occupational radiation by normal work. The results show that the doses and the dose rates obtained by the workers of the Nuclear Power Plant are low.

INTRODUCTION

Epidemiological studies among workers in the nuclear industry are being preferred because of that they offer a possibility for a direct assessment of the effect of the chronically expose with low doses by a human being.

The purpose of this study is to determine the probability of the occupational radiation to be a reason for the diagnosed newly found malignant disease with the group of workers from NPP.

MATERIALS AND METHODS

The concept of Probability of Causation (PC) determines what is the probability for the foregoing radiation to be the cause for the cancer disease with a given individual. The PC was defined by the United States National Institutes of Health Ad Hoc Committee as the fraction of the risk at the age of occurrence for the given cancer that is attributable to the exposure, i.e.

\[
PC = \frac{\Delta r(D,t,e,s)}{\Delta r(D,t,e,s) + r_0(a,s)}
\]

where \( r_0(a,s) \) is the cancer rate for age \( a \) and sex \( s \) for the particular cancer type under consideration and \( \Delta r(d,t,e,s) \) is the excess cancer rate due to a dose of radiation \( D \) at age \( e \) and time since exposure \( t (= a - e) \). The rate for a given cancer is the probability per unit time for a person of sex \( s \) and age \( a \) to develop the cancer (9).

The probability of a causative connection is accepted to be expressed by excess relative risk (ERR) and the dependence looks like the following:

\[
PC = \frac{ERR(D,e,s)}{1 + ERR(D,e,s)}
\]

All calculations have been conducted with the software product "Survrad", with which it can be determined the cancer risk and the probability reason for 17 radiogenic malignant neoplasm.

For a period of 10 years 67 cases with cancer have been diagnosed altogether. Data for occupational radiation of 27 workers with malignant illnesses have been submitted by Division “Safety”
in the NPP. 40 workers are not under control – they are not included in the system for individual dosimetric control due to lack of necessity. As a risk factor for the cancer emergency only the occupational exposure has been analyzed, as data for the rest radiation and non-radiation harms is incomplete.

Necessary data to conduct the analyses is year of birth, sex, year of first employment in IR environment and inclusion in the system for individual dosimetric control, occupational experience in an IR environment with individual dosimetric control, annual dose (for each year of employment, including null values of the dose), cumulative dose for the whole employment period in the IR environment, diagnosis, year of diagnosis.

RESULTS

The distribution of all cases (number of cases and their percentage proportion) according tumor localization by organs and systems is presented on Tab. 1.

Tab. 1: Number and percentage of cancer cases diagnosed among worker studied.

<table>
<thead>
<tr>
<th>Type of cancer</th>
<th>Number of cases</th>
<th>Percentage</th>
</tr>
</thead>
<tbody>
<tr>
<td>Male</td>
<td>43</td>
<td>64</td>
</tr>
<tr>
<td>Female</td>
<td>24</td>
<td>36</td>
</tr>
<tr>
<td>Total</td>
<td>67</td>
<td>100</td>
</tr>
</tbody>
</table>

On table 2 are shown the main characteristics of diagnosed cancer cases, for which full dosimetric data is available and PC is calculated. Analysis of the data (Table 2) shows that the dose loading of the examined employees of NPP is low, below 200 mSv. Only one case (No. 18) is an exception when the cumulated dose for the period prior to the diagnosed disease is approximately 300 mSv.

With 24 out of 27 workers the probability their occupational radiation to be the reason for the cancer is zero. There is a probability that the additional exposure to radiation could be the reason for carcinogenic illnesses in 3 employees (Case No. 6, 14, 18).

DISCUSSION

The studies of the health status of persons, professionally occupied in an IR environment can give us a direct assessment of the correlation between incidence and chronically radiation with relatively low doses, as interpretation of the results should be careful (it is very often when no other carcinogens from the professional and living environment are not taken into consideration et ct).

Basically the NPP workers are men because of which in the present study prevalent are cancer cases by men. The distribution of the malignances by localizations in the studied cohort is in accordance with their distribution characteristic for the whole country (10).

Cancer cases are late health radiation effects which mean that between the radiation and the appearance of the disease there passes a certain period of time. The duration of that latent period is different for the different cancer localizations.
Tab. 2: Assessment of probability of causation between occupational exposure and cancer diseases among workers in NPP.

<table>
<thead>
<tr>
<th>№</th>
<th>Sex</th>
<th>Diagnosis</th>
<th>Age at cancer diagnosed</th>
<th>Employment period before diagnosed cancer</th>
<th>Cumulative dose in mSv for the whole period before diagnosis</th>
<th>Probability of causation and 95% confidence interval</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>М</td>
<td>Skin cancer</td>
<td>49</td>
<td>8</td>
<td>4.02</td>
<td>0.00</td>
</tr>
<tr>
<td>2</td>
<td>М</td>
<td>Lung cancer</td>
<td>61</td>
<td>1</td>
<td>0.1</td>
<td>0.00</td>
</tr>
<tr>
<td>3</td>
<td>М</td>
<td>Lung cancer</td>
<td>47</td>
<td>9</td>
<td>133.49</td>
<td>0.00</td>
</tr>
<tr>
<td>4</td>
<td>М</td>
<td>Lung cancer</td>
<td>41</td>
<td>4</td>
<td>3.88</td>
<td>0.00</td>
</tr>
<tr>
<td>5</td>
<td>М</td>
<td>Lung cancer</td>
<td>45</td>
<td>24</td>
<td>14.3</td>
<td>0.00</td>
</tr>
<tr>
<td>6</td>
<td>М</td>
<td>Lung cancer</td>
<td>50</td>
<td>12</td>
<td>153.17</td>
<td>1% (0.0-0.02)</td>
</tr>
<tr>
<td>7</td>
<td>Ж</td>
<td>Breast cancer</td>
<td>49</td>
<td>5</td>
<td>0.91</td>
<td>0.00</td>
</tr>
<tr>
<td>8</td>
<td>Ж</td>
<td>Breast cancer</td>
<td>56</td>
<td>6</td>
<td>0.49</td>
<td>0.00</td>
</tr>
<tr>
<td>9</td>
<td>Ж</td>
<td>Breast cancer</td>
<td>50</td>
<td>16</td>
<td>4.38</td>
<td>0.00</td>
</tr>
<tr>
<td>10</td>
<td>Ж</td>
<td>Mastitis carcinomatosis</td>
<td>57</td>
<td>12</td>
<td>0.26</td>
<td>0.00</td>
</tr>
<tr>
<td>11</td>
<td>М</td>
<td>Laryngeal cancer</td>
<td>51</td>
<td>25</td>
<td>16.3</td>
<td>0.00</td>
</tr>
<tr>
<td>12</td>
<td>М</td>
<td>Esophageal cancer</td>
<td>34</td>
<td>13</td>
<td>34.06</td>
<td>0.00</td>
</tr>
<tr>
<td>13</td>
<td>Ж</td>
<td>Stomach cancer</td>
<td>54</td>
<td>11</td>
<td>3.15</td>
<td>0.00</td>
</tr>
<tr>
<td>14</td>
<td>М</td>
<td>Pancreatic cancer</td>
<td>43</td>
<td>14</td>
<td>181.15</td>
<td>1% (0.0-0.02)</td>
</tr>
<tr>
<td>15</td>
<td>М</td>
<td>Rectal cancer</td>
<td>50</td>
<td>4</td>
<td>1.47</td>
<td>0.00</td>
</tr>
<tr>
<td>16</td>
<td>Ж</td>
<td>Rectal cancer</td>
<td>56</td>
<td>11</td>
<td>5.03</td>
<td>0.00</td>
</tr>
<tr>
<td>17</td>
<td>М</td>
<td>Sigmoid cancer</td>
<td>53</td>
<td>28</td>
<td>118.49</td>
<td>0.00</td>
</tr>
<tr>
<td>18</td>
<td>М</td>
<td>Sigmoid cancer</td>
<td>48</td>
<td>29</td>
<td>281.87</td>
<td>1% (0.0-0.02)</td>
</tr>
<tr>
<td>19</td>
<td>М</td>
<td>Sigmoid cancer</td>
<td>54</td>
<td>22</td>
<td>0.7</td>
<td>0.00</td>
</tr>
<tr>
<td>20</td>
<td>М</td>
<td>Kidney cancer</td>
<td>42</td>
<td>7</td>
<td>2.93</td>
<td>0.00</td>
</tr>
<tr>
<td>21</td>
<td>Ж</td>
<td>Kidney cancer</td>
<td>50</td>
<td>8</td>
<td>1.25</td>
<td>0.00</td>
</tr>
<tr>
<td>22</td>
<td>М</td>
<td>Bladder cancer</td>
<td>51</td>
<td>14</td>
<td>0.61</td>
<td>0.00</td>
</tr>
<tr>
<td>23</td>
<td>Ж</td>
<td>Ovarian cancer</td>
<td>51</td>
<td>15</td>
<td>4.76</td>
<td>0.00</td>
</tr>
<tr>
<td>24</td>
<td>М</td>
<td>Cancer cerebri</td>
<td>48</td>
<td>13</td>
<td>200.88</td>
<td>0.00</td>
</tr>
<tr>
<td>25</td>
<td>М</td>
<td>Meningeoma</td>
<td>57</td>
<td>9</td>
<td>211.5</td>
<td>0.00</td>
</tr>
<tr>
<td>26</td>
<td>М</td>
<td>Oligoastrocitoma</td>
<td>44</td>
<td>8</td>
<td>48.28</td>
<td>0.00</td>
</tr>
<tr>
<td>27</td>
<td>М</td>
<td>Meta hepatic</td>
<td>50</td>
<td>24</td>
<td>22.06</td>
<td>0.00</td>
</tr>
</tbody>
</table>

NPP – Nuclear Power Plant; М - male; Ж - female;
The comparison of latent period continuity including different cancer cases in this present research involving the relevant localizations as per specialized data shows that the first option is shorter for a great deal of the cases. This comparison made on the ground of occupational exposure evaluation about examined cancer cases is rather unlikely.

Research aiming the analysis of the healthy effects of low dose and capacity exposure among occupational exposed persons has been restrictedly performed in Bulgaria. The observation of the healthy status of NPP workers did not show any registered cases of occupational exposure so far. This is due to low dose of occupational exposure (average annual effective dose for the NPP crew in the last few years is from 0.5 to 3.0 mSv per capita) as well as to emergency exposure of the crew (11, 12).

The analysis confirms that the dose load of the examined workers is low. 90% of all examined PC is practically 0 as expected, having in mind the low values of registered occupational exposure. These are malignant tumors that had spontaneously apperead and which would have been developed in a life time period of the said person regardless his working place.

There is only 1% probability in every 3 cases as a connection between a previous occupational exposure and a disease diagnostically confirmed. The main etiologic factor for lung cancer is smoking (13). The lack of detailed information about harmful habits (smoking) (case No. 6) of smokers in this research limits the particular participation of the said factors. The most affected age for pancreatic cancer is above 65 years old compared to the present case (case No. 14, the diagnosis has been confirmed in an earlier age). The most affected age groups of colon cancer are between 60 - 80 years (case No. 18) The confirmation of the genetic factors has been proved as a determined importance of the nutritive regime (food rich in animal fat increases the risk of cancer) (13).

Meantime, the evaluations of inductive-radiation cancer corresponding to the probability of reasonable connection are rather unlikely. They are bounded to inevitable extrapolations, dosimetry as well as of the participation of other cancerous professional and domestic conditions.

PC calculation offers the best method of systematically quantifying the probability that a particular cancer may have been induced by radiation in an individual (9).

CONCLUSION

The present research is one among the fewest making an attempt to clarify the healthy effects of occupational exposure by IR among workers in the nuclear industry. The lack of radiogenic solid tumors and leukaemia cases in appliance of modern research methods manifests the good level of existing radiation preventive measures in NPP – “Kozloduy”.

REFERENCES

Poster Presentations

WS I:
Education, Training and Knowledge Management
VELLA PROJECT: AN INITIATIVE TO CREATE A COMMON EUROPEAN RESEARCH AREA ON LEAD TECHNOLOGIES FOR NUCLEAR APPLICATIONS

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ABSTRACT

VELLA (Virtual European Lead Laboratory) is a Euratom FP6 project which emerged from the idea to create a common research area among the European Union and its associate countries in the field of lead technologies. A wide use of lead and lead alloys is foreseen in several nuclear-related fields, i.e. it is studied as coolant for critical and sub-critical nuclear reactors, as spallation target for neutron generation and for tritium generation in fusion systems. Given this possible future extensive use of lead in nuclear systems, which require a deep understanding of its physical properties and engineering applications, large efforts are dedicated to lead technologies. In particular, the EU has launched large R&D programmes, strongly interconnected. Among those programs VELLA, has the ambitious aim to homogenize the European research area in the field of lead technologies by exploiting the tools of Networking Activities, Transnational Accesses activities and Joint Research Activities.

1. Introduction
A wide use of pure lead, as well as its alloys (such as lead-bismuth, lead-lithium), thanks to its favourable properties, is foreseen in several nuclear-related fields, i.e. it is studied as coolant for
critical and sub-critical nuclear reactors, as spallation target for neutron generation and for tritium generation in fusion systems.

Given this possible future extensive use of lead in nuclear systems, a deep understanding of its physical properties and engineering applications is mandatory. As a consequence, given the quite limited nowadays experience, large efforts both at national level as well as within the European Commission are dedicated to the development and understanding of heavy liquid metal technologies. In particular, the European Commission has launched several large R&D programmes, strongly interconnected, such as TECLA (*TEChnologies for Lead Alloys*), MEGAPIE-TEST (*MEGawatt PiPilot Experiment*), EUROTRANS-DEMETRA (*EUROpean research programme for the TRANsmutation of high level nuclear waste in Accelerator Driven Systems-DEvelopment and assessment of structural materials and heavy liquid METal technologies for TRAnsmutation systems*), ELSY (*European Lead-Cooled SYstem*), and VELLA (*Virtual European Lead Laboratory*).

VELLA is an FP6 project which has the ambitious intent to create a virtual laboratory for lead technologies. More in detail, the driving idea of VELLA is to homogenize the European research area in the field of lead technologies in order to produce a common platform of work which will continue also beyond the VELLA initiative. Above all, VELLA has the ambitious intent to both create a network of all the principal laboratories and to strongly connect the different groups of experts, in order to have a common definition of the good operational practices and to promote the exchange of the scientific results by means of appropriate and innovative tools and procedures. It also has the important objectives to promote the access to the main existing facilities in the EU to different specialist groups. This will allow supporting the technological development and the qualification activities as well as the formation of a European “scientific community”, organized to meet all the technological challenges and the necessary research requirements.

In this framework, detailing the abovementioned goals, VELLA is articulated in Networking Activities (NA), Transnational Accesses activities (TA) and Joint Research Activities (JRA).

### 2. Networking Activities

The Networking Activities have as main objectives to create a virtual community of researchers, to define common standards and protocols for the use of the facilities and to interact with the programmes and the institutions operating in this field.

#### 2.1 Networking Activity 1: “Management of the consortium activities”

VELLA is a Consortium gathering thirteen European Research Institutions and Universities, from nine Countries, established in 2006, with the support of the European Commission, under a three-years contract.

The initiative is coordinated by ENEA (Italy). The other partners, at the moment, are: Le Commissariat à l’Energie Atomique (CEA), CIEMAT, Consiglio Nazionale delle Ricerche, sezione di Genova (CNR-IENI), CNRS, Forschungszentrum Karlsruhe GmbH (FZK), Forschungszentrum Dresden -Rossendorf e. V. (FZD), Institut Quimic de Sarria -Universitat Ramon Llull (IQS), Kungliga Tekniska Högskolan (KTH), Nuclear Research Institute Rez (NRI), Paul Scherrer Institut (PSI), The Belgian Nuclear Research Centre (SCK-CEN), and Institute of Physics, University of Latvia (IPUL).

The management structure consists of: an Executive Board (EB), formed of representatives from each participant institution, which is the Consortium decision-making and arbitration body. A Technical Advisor Committee (TAC) has the responsibility for scientific advices and recommendations covering the topics of the Project. A Scientific Access Panel (SAP) has the duty of selecting the accesses to the infrastructures during the whole duration of the Project, selecting the Human Mobility (HM) applications, monitoring the accepted actions. A Scientific and Technical Panel (STP) has the responsibility to manage the Joint Research Activities within the project execution. Finally, the Coordinator is the single point of contact between the EC and the Consortium and it is responsible for the project management and the Project Office (PO) is the body which manage the administrative, legal, financial and other non-technical aspects of the Project and assist the abovementioned bodies and the Coordinator in their activities.
2.2 Networking Activity 2: “Virtual community creation and results dissemination”

The activity has the main objective to create a real “virtual” community of researchers in the field of heavy liquid metal (HLM) technologies for nuclear applications through:

- the creation of an interactive website where it is possible to access all the publications related to the project, to have direct contact with researchers for further information and to visit the “virtual laboratory”; 
- the realization of a “dedicated chat line” (and/or programmed teleconferences) for discussion among researchers; 
- the edition of a VELLA electronic newsletter reporting the most important news; 
- the organization of dedicated workshops on specific issues related to the HLM technology.

2.3 Networking Activity 3: “Harmonization of knowledge and establishment of good practices”

The activity has a twofold objective:

- the improvement and the harmonization of the scientific knowledge of the present generation of scientists and engineers coming from different areas of the nuclear R&D projects related to HLM; 
- the preparation of the new generation of scientists, by giving them a common platform of knowledge and developing their skills in all aspects of HLM technologies and related areas.

For attaining these objectives, two types of actions are considered:

- thematic workshops on HLM technologies to be organized by the VELLA partners during the three-years duration of the initiative; 
- ‘Good practices’ workshops to be organized for young researchers involved in the field of nuclear science coming from Universities, Agencies or Companies in the different research teams or laboratories of the Organizations grouped in VELLA.

2.4 Networking Activity 4: “Guidelines and procedures”

The activity is aimed at developing guidelines for relevant issues for future HLM systems, collecting the available information, analysing the experimental procedures used in different laboratories, evaluating the quality of the available data, identifying items not covered and realizing, if needed, some experiments to fill the existing gaps.

The expected impact of this NA, therefore, is to provide the research community with recommendations, procedures and data to be used for the design and construction of new HLM systems. It is intended to represent a forum for discussion and understanding of the most relevant topics. The recommendations and procedures issued, as results of this activity, are intended to be used by the labs involved in HLM investigations. If possible, standard proposal will be written to be submitted to ISO.

2.5 Networking Activity 5: “Collaborations with other programmes and future R&D”

The main goal of the NA5 is to promote a real integration among the EU activities on HLM already going on within Europe in several fields and to establish regular and coherent links among national and international programmes in the field of HLM technologies in order to increase the possible cooperation.

This objective will be pursued setting up a group of experts, including the major EU specialists in the field of HLM, establishing links with other teams working on programmes related to the HLM technologies in order to have a complete overview of the different research actions going on and promoting collaboration and exchange of knowledge.

3. Transnational Access Activities
The TA are activities aimed to promote the access of researchers, universities and companies to the existing infrastructures and knowledge, in order to increase the competitiveness of the European industry, to train the researchers in using the EU infrastructures during the three years duration of the project and to help the human mobility between and towards the laboratories. These objectives will be realized using two different tools:

- access to the infrastructures, granted to the users interested in technological development and/or basic research;
- Human Mobility, to support researchers inside and outside the VELLA Consortium.

For both the access to the infrastructures (Transnational Access) and the Human Mobility, a call for proposals will be published on the VELLA website (www.3i-vella.eu) every year, soliciting applications from interested candidates.

### 3.1 Access to the Infrastructures

The infrastructures made available within VELLA are the two virtual structures MATLAB and CHEMLAB, and the large devices considered reference laboratories themselves. These infrastructures can give access to researchers and users creating the unique opportunity of the accessibility to a large European common laboratory.

MATLAB *(MAterial LABoratory)* is a virtual laboratory where it is possible to perform compatibility tests among different structural materials and heavy liquid metals. The possible tests include tensile, fatigue, fracture toughness, creep and corrosion investigations on irradiated and unirradiated materials. The corrosion science laboratory comprises both facilities for flowing liquid metal and facilities for stagnant tests.

Parts of MATLAB, in fact, are LECOR & CHOEPE III (ENEA, Italy), COLONRI I&II (NRI, Czech Republic), LINCE (CIEMAT, Spain), CORRIDA (FZK, Germany) and CICLANL (CEA, France) for corrosion tests of unirradiated materials in flowing Lead and Lead-Bismuth. Also LIMETS I&II (SCK.CEN, Belgium), COSTA 1-6 (FZK, Germany) and COLIMESTRA (CEA, France) belong to MATLAB. They are stagnant devices for irradiated materials (the first one) and for unirradiated materials (the last two devices).

CHEMLAB *(CHEMistry LABoratory)* is a virtual laboratory devoted to the chemistry control related studies of Lead-Bismuth eutectic systems, such as the oxygen control and monitoring, as well as the other impurities control in both the liquid and the gas phases. It comprises ELEFANT (FZD, Germany) STELLA (CEA, France) OCEAN & THESYS (FZK, Germany) and CHEOPE II (ENEA, Italy).

Finally, also three large facilities offer access in the framework of the VELLA TA: “CIRCE”, pool type facility for the study of thermal – hydraulics issues, “TALL”, originally designed to investigate thermal-hydraulic phenomena for the ADS normal and transient conditions, and “THEADES”, a loop type facility having relevant dimension for thermal-hydraulic studies and large components testing.

### 4. Joint Research Activities

The JRA are technical activities with the objective to: develop and the technologies needed for the operation of large facilities for future Gen IV reactors and ADS cooled by HLM; develop the needed components and instrumentation; study the liquid metal thermal-hydraulics and analyse the effects of irradiation in presence of LBE. Finally, to homogenise and complete the results obtained in the other research programmes related to the HLM technologies for nuclear applications.

#### 4.1 Joint Research Activity 1: “Lead technology”

The objective of this JRA is the development and validation of the needed technologies and the required know-how for the future Gen-IV reactors and Accelerator Driven Systems, providing, above all, data on:
the oxygen control in large lead pool-type configurations, including the development of oxygen sensors for these applications;
the corrosion behaviour of potential materials in stagnant lead;
the influence of lead on the mechanical properties (tensile, creep rupture).

4.2 Joint Research Activity 2: “HLM components, instrumentation development and system operation”

The JRA2 has the objectives of:
- defining and developing optimised systems and dedicated components for future experiments characterizing the features and the potentialities of the existing dedicated instrumentation;
- developing new instrumentation (e.g. pumps, flowmeters, coolant handling systems, etc.) for optimised experimental facilities;
- defining and validating experimentally, if necessary, the basic operational procedures;
- developing a standardized In Service Inspection and Repair methodology.

4.3 Joint Research Activity 3: “Liquid metal thermal-hydraulics”

Three experiments are proposed in this JRA, which will be conducted by at least by two associations with a good experience in liquid metals and are followed by the whole HLM community.
- single phase turbulent heat transfer from highly heat loaded surfaces;
- single phase buoyant and mixed convection in loop systems;
- free surface shaping in heavy liquid metal flows in dependence of the geometric and fluid dynamic boundary conditions.

4.4 Joint Research Activity 4: “Irradiation in presence of LBE”

The aim of this JRA is to cover a major lack in the capabilities of the existing facilities to perform experiments by combining HLM and neutron irradiation. Hence a high temperature lead alloy loop will be developed of, to be installed in the MTR reactor BR2. The in-pile HLM loop should be designed in such a way that it can be used for the purposes that are common to the development of GEN-IV LFR and Pb(-Bi) ADS, whereby some of these points are as well covered in LiSoR.

5. Acknowledgments

This work is supported by the European Commission under the Contract VELLA number 036469.

6. References

[2] Vella official website: www.3i-vella.eu
Poster Presentations

WK II:
Safeguards and terrorism
ABSTRACT

Pulsed Fast Neutron Analysis (PFNA) has long been suggested as a potential means for the non-destructive detection of hidden explosives and contraband. The premise for the technique is to use fast neutrons to stimulate $\gamma$-ray emissions that are characteristic of the target isotope. However, it has often proved difficult to 1) separate the response of the material under detection from materials that have a similar isotopic composition, 2) distinguish the radiation response from scattered neutrons and 3) to process the data sufficiently quickly for real-time processing. In this paper we report on a novel approach to PFNA that exploits the digital discrimination of neutrons and $\gamma$ rays to provide information on the shape and composition of suspicious objects in mass transport systems.

1. Introduction

Ionising radiation has long been used for the non-destructive screening of opaque objects ever since x-ray use was pioneered by Roentgen. The recent global escalation in the threat level associated with terrorist attacks, and horrific events such as the Madrid bombings and the July 7th 2005 attacks in London, have renewed research interest in a variety of potential screening capabilities, and especially the use of neutrons. Neutrons are very useful because of they have great penetrating ability, as a result of their having no electrostatic charge, and because they are easily detected by relatively inexpensive detector systems that are now widely available. Furthermore, inelastic interactions between the incident neutron and the atomic nuclei making up the object under scrutiny often result in the emission of prompt $\gamma$ rays. The energies of these photons are characteristic of the isotope that emitted them and, since most explosives comprise of carbon, nitrogen, oxygen and/or hydrogen, it is thus possible to identify the substance emitting them. A variety of possibilities based on this concept have been studied over the past twenty years and a few are now deployed commercially.

However, the measurement of neutron energy is notoriously difficult, predominantly because all indication of the energy of the neutron is often lost via the indirect basis on which neutrons are usually detected, in order for them to be converted to electronic signals. Limited spectroscopic information can be gleaned by unfolding the neutron response from several detectors with
complementary spectral sensitivities, but this is sophisticated and often results in too crude a response to extract detailed analytical information. Alternatively, the time taken for neutrons to traverse a given distance, the time-of-flight (TOF), can be used to extract the neutron energy and/or to determine the origin of the neutron but then it is necessary to also know the time at which the primary neutron was emitted. This requires the use of neutron sources that are pulsed very rapidly indeed. Interestingly, for most inelastic reactions relevant to freight and baggage screening, neutron energies in excess of 10 MeV are required. Whilst 14 MeV neutron generators are available, based on the deuterium-tritium (D-T) reaction, the traverse-time of a standard candidate container by a neutron with such energies will be of the order of tens of nanoseconds.

Fast neutrons are often detected with scintillation detectors. Instruments using proprietary organic liquid scintillation compounds respond with typical pulse-lengths of the order of a few nanoseconds. Whilst high-end commercial electronic systems have been available for some time that are able to digitise γ-ray events arising from semiconductor detectors (see for example DESCOVICH et al. (2005)), it has only been possible to sample at a rate compatible with digitisers and until recently digitisers were not fast enough for scintillators. Thus, neutron detection and measurement has been limited by the capabilities of analogue data acquisition systems, many of which were developed thirty years ago. The discrimination and rejection of contaminant γ-ray events (to which organic scintillators also respond) has long been performed in the analogue domain. Amongst the principal restrictions of such systems are the inability to post-process the pulse data and the requirement of very careful skilled optimisation. These result in difficulties in the automation of such processing and the inherent incompatibility with computer-based control. Most importantly, all record of the continuous signal that describes each event is lost and only the salient features, such as the pulse amplitude or rise time, can be recorded.

In this paper we report on the digital processing of neutron events for use in counter-terrorism applications. The potential flexibility of this advance is described in the context of improved neutron/γ discrimination and the digital measurement of TOF. Although much of the data presented here has been acquired with a standalone digital data acquisition system more consistent with a laboratory experiment, the performance requirements are compatible with the burgeoning capability of field-programmable embedded systems.

2. Pulse-shape discrimination

2.1 Experimental details

The experimental set-up for this aspect of the research consisted of a 4.5 ml cylindrical cell scintillation detector filled with EJ-301 organic liquid (John Caunt Scientific Ltd., UK), optically-coupled to a Hamamatsu R5611 photomultiplier tube (PMT). The PMT was operated with a negative high-tension (HT) supply voltage of −840 V dc. An 8 m length of 50 Ω coaxial cable was used to transport the signal from the PMT to an Infinium digital oscilloscope (Agilent Tech.). Data were recorded digitally with a sampling frequency of 4 GHz and 12 bit amplitude resolution. Data acquisition was automated by driving the oscilloscope remotely via a Transmission Control Protocol/Internet Protocol (TCP/IP) connection to a personal computer running a bespoke Matlab program. The personal computer used for this system was a Dell Latitude D400 laptop.

Events were acquired from an americium-beryllium (Am-Be) neutron source at the Physics Department, University of Liverpool, UK. This is suspended in a moderating water tank and externally shielded on three sides with 30 mm thick of plate cadmium, encased in sheet plywood. This source can be moved forwards and backwards to vary the degree of neutron moderation. The source was positioned 180 mm from the outside edge of the front of the tank; the detector was positioned in line with the source (360 mm above ground-level) at a distance of 800 mm from the front edge of the tank, as shown in Figure 1. A dataset comprising 5000 events was collected.

The Am-Be source has a low neutron/γ ratio and the high proportion of γ rays from background and scattering results in a flux consisting mainly of γ-ray events. By including a γ-ray absorber, a more even spread of neutron and γ-ray events can be obtained. A 50 mm thick lead wall, placed immediately in front of the detector was enough to significantly attenuate the γ-ray flux to a neutron/γ ratio of ~50:50. Presented in Figure 2 are twenty superimposed pulses acquired using the set-up described above.
2.2 Results and discussion

Pulse gradient analysis (PGA) (D’MELLOW et al. (2007)) is a simple and inherently fast method for the classification between γ-ray and neutron particle detection in organic liquid scintillators. In common with many analogue discrimination methods, it exploits the principle that the scintillation light function due to a neutron interaction decays more slowly than that for a γ-ray interaction. However, in the digital domain the elegance of the technique arises from the comparison of sample amplitudes early in a given pulse’s evolution.

Evidence of digital PGA is presented in Figure 3, as a scatter plot of peak amplitude versus discrimination sample amplitude, where the discrimination sample amplitude is a measure of the decrease of the pulse amplitude in a given time after the peak. Clearly, two characteristic plumes are observed. The data were sampled at 250 MHz, 8-bit amplitude resolution and the amplitude of the sample 10 ns after the peak was measured. Figure 4 suggests that the optimum sample to use for discrimination lies between 15 ns and 25 ns after the peak of the pulse for the EJ-301 liquid scintillator. In this time window the separation between pulse shapes is greatest. Whilst there is an optimum discrimination sample for synthetic pulse shapes (D’MELLOW (2006)), it is favourable to discriminate as early as reasonably possible to minimise the possibility of pile-up events corrupting the single pulse amplitudes.
3. Digital time-of-flight

3.1 Experimental details

The Van de Graaff accelerator at the National Physical Laboratory (NPL) Neutron Irradiation Facility was used to produce a 2.924 MeV proton beam which was incident on a thin 60 μg cm\(^{-2}\) lithium fluoride target. Through the \(^7\)Li\((p,n)\)^7Be reaction neutrons at 1.225 MeV and 0.745 MeV energies were produced corresponding to transitions to the ground and first excited states respectively of the product nucleus. The accelerator was operated in pulse mode, and a capacitive-type detector was positioned in the beam line at about 1.5 m from the lithium fluoride target in order to detect the arrival of the proton beam pulse immediately before they collided with the target. This provides the beam pickup signal which is used to provide timing information for the time-of-flight measurement and to provide information on the proton pulse duration and frequency. The transit time from the pickup to the target is constant because the proton beam is mono-energetic and the target itself is very thin. Therefore, there is a constant delay between the beam pickup signal and the emission of neutrons from the \(^7\)Li\((p,n)\)^7Be reaction. Thus the beam pickup is used to identify the start, pulse duration and period of the subsequent neutron pulses. The proton pulse in this experiment had a period of 400 ns and pulse width of 5 ns.

The detector set-up consisted of the same scintillation detector described in Section 2. In this case, the HT was connected to the PMT via a standard RG58 cable. The output signal from the scintillator was connected to channel 1 of an Infinium® digital oscilloscope, via a Huber+Suhner SX 07262 BD cable. This cable preserved pulse shape information whereas the standard RG58 did not. The beam pickup signal was coupled to the control room from the main bay via a 30 m length of RG58 cable. The beam pickup signal was then passed through a discriminator and connected to channel 3 of the digital oscilloscope via a further 4 m length of 50 Ω coaxial cable. The scintillator was taped onto the NPL plastic scintillator and was placed 54 mm vertically off-axis and 1613 mm horizontally from the face of the lithium fluoride target.

Scintillator pulses were used to trigger acquisition. Acquisition of the scintillator pulse and corresponding beam pickup pulse data were automated by driving the oscilloscope remotely via a TCP/IP connection to a personal computer running a bespoke Matlab® program.

3.2 A comparison of analogue time pick-off and digital leading-edge triggering

The plastic scintillation detector in the experimental set-up at NPL has a constant-fraction discriminator built into the base of the detector’s photo-multiplier. The signal from this detector is attenuated by a long 50 Ω cable from the experimental area to the control room; therefore, an Ortec 436 100 MHz discriminator is used to boost the signal. The output from this discriminator provides a time-to-amplitude converter (TAC) unit with its start pulse. The beam pickup pulse acts as the TAC stop pulse. The TAC data are presented in Figure 5(a).

The leading-edge triggering method is the simplest time pick-off method. It is based on the identification of the time that the detector signal crosses a fixed trigger-level. Leading-edge triggering is a commonly used timing method and can be quite effective provided the dynamic range of the input pulses is small. In the present context it is the simplest method to replicate digitally in order for comparison to be made with the established analogue method. Amplitude walk can be a source of uncertainties with leading-edge timing methods and arises as a result of variations in amplitude and pulse shape. Timing uncertainty resulting from amplitude walk is unacceptable in situations where accurate timing is required over a wide amplitude range. Detectors that have variable charge collection times, such as germanium counters, will be subject to amplitude walk irrespective of input pulses of constant amplitude.

Amplitude walk uncertainties are reduced by setting the trigger-level as low as possible. However, the trigger-level should be in a region of fast rise time to minimise uncertainties due to jitter. An optimum trigger-level is usually between 10% – 20% of the average pulse amplitude. For the TOF spectra presented in Figure 5(b) the trigger-level was set to 20% of the average pulse height.

For the TOF data collected, the purpose was to use the TOF information to provide n/γ discrimination rather than to achieve the best timing. The TOF spectra presented in this work do not have the best timing resolution possible, but rather demonstrate the importance of digital TOF data.
Some points which should be clarified are, (1) the NPL TOF system is under development working towards optimisation of the timing; (2) the Lancaster scintillator may have better timing characteristics because it is smaller than the NPL scintillator; and; (3) background scatter has not been subtracted from either spectrum. Nevertheless, the significance of Figure 5(b) with respect to Figure 5(a) is a sharper spectral line for the 0.745 MeV neutrons and a lower baseline is achieved.

Figure 5: Time-of-flight (TOF) spectra for the lithium-7 reaction over a flight path of 1.614 m using the Neutron Irradiation Facility at the National Physical Laboratory.

4. Conclusions

In this paper we have described two digital methods for the processing of neutron events that are fundamental to advanced PFNA methods of screening in counter-terrorism. The digital capture of neutron data enables post-processing and optimisation of entire data sets because each complete pulse is stored. In particular, mixed radiation fields can be sorted in terms of their neutron and γ-ray components and this discrimination process is carried out in software; therefore the efficiency of the method is only reliant upon the algorithm used. By replicating analogue timing methods in the digital domain it is possible to time-stamp neutron events in real-time. Once again, the opportunity exists to post-process the data to optimise the confidence with which the timing information is held.

5. References


6. Acknowledgements

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MEGAPORTS: DETECTION OF NUCLEAR SMUGGLING IN THE PORT OF ANTWERP

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ABSTRACT
Since the beginning of 2007 Belgian customs performs nuclear inspections with the widely used radiation portals. The detection of nuclear smuggling with these plastic scintillator detectors is not as simple as it seems. In this paper we will present the results of a 6 month measurement period of container traffic in the port of Antwerp. The main conclusion is that almost all alarms on radiation portals are due to natural occurring radioactive materials (NORM). The combination of a scanning device and radiation portals in the inspection is an important aid to distinct between NORM alarms and the few alarms that are due to radiological contaminations or artificial radioactive sources.

1. Introduction
The Megaports Initiative is a worldwide effort to prevent nuclear smuggling in sea containers and was started after 9/11. In the port of Antwerp the installation of a nuclear detection system is co-funded by the American and Belgian governments. Starting from 2007 Belgian customs inspects container traffic on nuclear smuggling in the port of Antwerp, the third biggest port in the world. The container traffic is permanently inspected with radiation portals. The portals are equipped with 4 helium tubes for neutron detection and 4 plastic scintillators for gamma detection.
Belgian customs officers make a decision on releasing or holding a container after it alarmed on a radiation portal. This is not an easy or straightforward task and therefore Belgian customs asked NuTeC to be the scientific partner in the inspections during the first year.

In 2006 NuTeC performed a survey of the container traffic with a radiation portal. These measurements were needed to estimate the effect of the inspections on the workings of the port and to have a first glimpse of the kind of radioactive materials that are shipped through the port of Antwerp. The survey was performed at the scan site of Antwerp customs. At this site containers are scanned for smuggling. The container traffic at the scan site is selected from all over the port and therefore can be estimated as a site with an average container traffic. In figure 1 the set-up for the survey is presented. It consisted of two radiation portals and one x-ray scan facility or scan tunnel (2 linear accelerators).
2. **Results and discussion**

Measurements were carried out during a 6 month period in 2006-2007. In general the results are the following:

<table>
<thead>
<tr>
<th>Number of scanned containers</th>
<th>9145</th>
</tr>
</thead>
<tbody>
<tr>
<td>Number of alarming containers</td>
<td>624 (6.8%)</td>
</tr>
<tr>
<td>Number of containers with radiation levels above legal inspection limit</td>
<td>27 (0.3%)</td>
</tr>
<tr>
<td>Number of measurements with handheld equipment</td>
<td>37 (0.4%)</td>
</tr>
<tr>
<td>Number of containers opened for physical inspection</td>
<td>3 (&gt;0.1%)</td>
</tr>
<tr>
<td>Number of contaminated materials found</td>
<td>1</td>
</tr>
<tr>
<td>Number of unlicensed radioactive sources found</td>
<td>0</td>
</tr>
</tbody>
</table>

**Tab 1: General results**

Portals give an alarm when a radioactive load passes through them. During the survey almost all alarms were caused by natural occurring radioactive materials (NORM). All measured activities were below exemption levels for radioactive materials as stated in the Belgian and European legislation. Therefore no containers were blocked during the survey period.

The materials causing alarms can be quite divers, the reason for the radioactive nature of these materials is always the same: NORM. Only one case of a Co-60 contamination was detected in a period of 6 months.

<table>
<thead>
<tr>
<th>Type of material</th>
<th>% of alarms</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ceramics</td>
<td>38</td>
</tr>
<tr>
<td>Stone</td>
<td>16</td>
</tr>
<tr>
<td>Biological materials</td>
<td>7</td>
</tr>
<tr>
<td>CRT (televisions)</td>
<td>6</td>
</tr>
<tr>
<td>Ores</td>
<td>5</td>
</tr>
<tr>
<td>Kitty Litter</td>
<td>2</td>
</tr>
<tr>
<td>TENORM</td>
<td>3</td>
</tr>
<tr>
<td>Glass fibre and glass</td>
<td>4</td>
</tr>
<tr>
<td>Chemical products</td>
<td>5</td>
</tr>
<tr>
<td>Others</td>
<td>13</td>
</tr>
<tr>
<td>Unknown</td>
<td>2</td>
</tr>
</tbody>
</table>

**Tab 2: Type materials causing alarms during the survey period**
Alarms on radiation portals do not give information about the nature of the alarm. To distinct between NORM, contaminated materials, radioactive sources or nuclear smuggling measurements with handheld equipment are a possible technique.

During the survey very few inspections with handheld equipment were needed due to the presence of the x-ray scanner. The scanner allowed a comparison between the radiation alarm profile and the contents of the container. In almost all cases alarms could be sufficiently explained by this technique. For example if a container causes an alarm on a radiation portal and this is due to the presence of ceramics (NORM), the scan image clearly shows this.

Information about the contents of the container is also needed to verify if the radiation has a natural source. For example if steel rods cause an alarm this is not due to natural radiation, but a result of the presence of radioactive sources during the recycling of scrap metal.

3. Conclusion

In conclusion we can state that techniques and working protocols for nuclear inspections should be selected so that a distinction between NORM, contaminated materials, radioactive sources and nuclear smuggling is easy and straightforward. For example the combination of a radiation portal and a X-ray scanner is a strong and efficient technique. Furthermore information about the contents of containers should be easily accessible and reliable.