

~~ENS~~ ~~RRFM~~ 2005

TRANSACTIONS

Poster Session 4

th
9 International Topical Meeting

Research Reactor Fuel Management

European Nuclear Society (ENS)
in cooperation with the
International Atomic Energy Agency (IAEA)

Hilton WestEnd Hotel Budapest, Hungary

April 10-13, 2005

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STATUS OF THE VIND PROGRAM, SEPTEMBER 2004

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ABSTRACT

The Vinca Institute Nuclear Decommissioning (VIND) Program (earlier known as the "Green Vinca") was approved by the Government of the Republic of Serbia in July 2002. The main goal of the Program is to solve nuclear and radiation safety related problems currently existing in the Vinca Institute of nuclear sciences that were accumulated as a 'heritage' of various nuclear programs carried out in the country since 1960. This paper summarizes the achievements and status of the VIND Program, two years after its formal initialization. The Program is supported by technical assistance from the IAEA since 2003 and by a donation of an US NGO "Nuclear Treat Initiative", made in 2002. Four interrelated projects of the Program are supposed to be completed in next 10 years by about 40 experts and 40 technicians from the Vinca Institute. The total cost of the VIND Program is estimated at 40 million US\$.

1. Introduction

Nuclear and radiation safety conditions in the Vinča Institute of Nuclear Sciences, elaborated at the end of last century, were found as inappropriate. These problems were mainly arisen as results of various nuclear programs carried out from 1958 to 1990 at the former Yugoslavia and strong economic crisis during the last decade. The Vinca's and international experts have concluded that these radiation safety conditions had to be improved as soon as possible. The major problems, pointed out, were:

- Considerable amount of low and high-enriched uranium (HEU) spent nuclear fuel (SNF) accumulated during 25 years of operation of 6.5 MW heavy water RA research reactor. After storage period from 20 to 40 years in water, aluminium cladding some of the spent fuel elements is degraded and fission products (primarily ^{137}Cs nuclide) leak to water environment;
- RA research reactor was in the extended shut down since 1984, when its modernization was started, but never completed due to various technical, political and economic reasons;
- Different low level (LL) and medium level (ML) radioactive waste (RAW) was collected from whole ex-Yugoslavia and stored in two storage hangars at the Vinca site since 1960. Records on the waste description are inadequate or (in the most of the cases) are missing; the RAW is not properly stored and conditioned according to modern radiation safety standards, and,
- Significant amount of fresh HEU fuel elements were stored at premises of the RA research reactor.

To solve the problems mentioned above, new projects, latter known jointly as the "Green Vinca Program" were initiated firstly in the Vinča Institute in August 2001. The basics of these four projects (related to SNF shipment, RAW management, RA reactor decommissioning and RB reactor refurbishment) were announced at the International Atomic Energy Agency (IAEA) 2001 General Conference by the Yugoslav delegation. The projects were welcome by the IAEA officials. Initial program team was assembled from about 30 experts and the same number of technicians from the Institute and some relevant organisations. It was expected that the Program will be supported, besides++ the Government's funding, also by donations expected from foreign organizations and governments and by experts' help offered from the IAEA. The necessary equipment should be

obtained through the technical assistance from the IAEA and applications for few relevant projects were sent through the country official channels to the Technical Cooperation (TC) Department of the IAEA in late 2001. Close cooperation of the team members with experts and relevant companies from nuclear developed countries is expected. Preparation phase of these activities were started already in 2001.

Proposal for solutions of the radiation and nuclear safety problems at the Vinca Institute, mentioned above, prepared jointly by the Vinca's and international experts, engaged by the International Atomic Energy Agency (IAEA) in late nineties and at beginning of 21st century, was submitted to the Government of Serbia and to the Government of the Federal Republics of Yugoslavia, for consideration and support. Both Governments have had Expert Committees that have had evaluated the proposal and had promised support at various levels of organization and through appropriate decisions. Here are especially underlined the 'opinions' of the Government of the Republic Serbia about the 'Green Vinca Program', brought at July 2002:

- Research reactor RA shall be finally shut down and decommissioning process should be initiated;
- Fresh HEU fuel elements from both research reactors shall be sent back to Russian Federation (RF) for uranium down blending;
- Safety and storage conditions of SNF elements should be increased with the aim to ship safely all the spent fuel back to RF, as soon as possible;
- LL and ML RAW at the Vinca site should be properly managed and safely stored in new temporary storage at the site with the aim to ship it at the final disposal site, to be determined and constructed in future;
- RB critical assembly will continue operation with the Government's support, and
- The Vinca Institute of nuclear sciences was set in charge of organization and completion of the Program.

2. VIND Program activities in 2002 – 2003

Intensive negotiations on shipment fresh high-enriched uranium fuel elements from the Vinca research reactors back to Russian Federations were carried out in 2002. These negotiations have included different Ministries of the Government of the Republic Serbia and the Government of the Federal Republics of Yugoslavia, USA Department of the State and USA Department of the Energy, the MINATOM of the Russian Federation, representatives of the IAEA and an USA nongovernmental organization "Nuclear Treat Initiative" (NTI). Beside the Vinca Institute, expert organisations of other two involved countries and the Safeguards Department of the IAEA were involved too. As a result, the first shipment of fresh HEU fuel from the Vinca site back to the RF has occurred in August 2002 and, at the same time, the first considerably donation to the Program was obtained from the NTI.

Work with the IAEA experts had also resulted to establishment of three Vinca's projects supported generously by the IAEA TC Department from beginning of 2003. These projects are:

- Safe Removal of Spent Fuel of the Vinča RA Research Reactor, SCG/4/003 (ex-YUG/4/029),
- Safe Management of Waste in the Vinča Institute, SCG/4/005 (ex-YUG/4/031), and
- Decommissioning of the Vinča RA Research Reactor, SCG/4/004 (ex-YUG/4/030).

Further support to the Program was obtained though participation of the country in few IAEA TC regional projects.

The Federal Republics of Yugoslavia changed name to the Serbia and Montenegro (SCG) in March 2003, but general attitude to the Green Vinca Program had stayed unchanged. Ministry for Science, Technology and Development of the Republic of Serbia has appointed the Vinca Institute of nuclear sciences in charge of the Green Vinca Program and established an independent the Green Vinca Expert Committee with the main aim to monitor the program progress and to propose optimal attitude to solutions for the problems treated. Unfortunately, funding of the Program was at very low level, so the IAEA support, through the service contracts and requested equipment, was the main driving force for the program participants.

The project '**Safe Removal of Spent Fuel of the Vinča RA Research Reactor**' has a main task to prepare SNF of the RA reactor for shipment back to RF. Research heavy water reactor RA was operated in the Vinča Institute of Nuclear Sciences since 1959 to 1984 using 2 % enriched and 80 % enriched uranium fuel elements. The SNF, stored in the Vinča Institute of Nuclear Sciences, consist of about 8000 irradiated fuel elements – the total mass of about 2.5 tons of uranium. Both types of the fuel elements are known as the TVR-S type and are ex-USSR origin. Fuel elements have the same shape and dimensions and approximately the same initial mass of ^{235}U nuclide. Almost all SNF elements are stored in the spent nuclear fuel pool filled by ordinary water. About 500, last used, SNF elements are kept in the RA reactor core since 1984. Due to non-suitable chemical parameters of water in the spent fuel storage pool the corrosion processes penetrated 1 mm thick aluminium cladding and few millimetres thick aluminium walls of storage containers during storage period long from 20 years to 40 years. Activity of fission products (^{137}Cs nuclide) is detected in water samples since 1996. Measurements of water samples, made from 2000 to 2003 have found increased ^{137}Cs activity in about 1/3 of the storage containers.

Regular monitoring and maintenance of water quality in the spent fuel storage pool were established in 1996 and following actions to improve conditions in the existing temporary RA reactor spent fuel storage pool are finished or already in progress:

- Design and production of special equipment for underwater cutting of corroded iron steel construction in the basin 4 of the storage pool;
- Removal of the contaminated iron steel construction, its conditioning and storage at the temporary low and medium level radioactive waste storage at the Vinča site;
- Final removal of sludge from the spent fuel storage pool. Physical purification of pool water by mechanical filtering;
- Washing up corrosion deposits from all surfaces in contact with the pool water using technology and equipment already provided by Russian side;
- Chemical purification of pool water using the ion exchange resins;
- Increasing physical protection at the site, and
- A work on SCG – RF government-to-government agreement for repatriation of the SNF from the RA reactor is in progress since November 2003.

The project '**Decommissioning of the Vinča RA Research Reactor**' was initiated due to the fact that, for a number of both technical and political reasons, the RA reactor has not ever been restarted during long period of extended shutdown. All plans for the reactor RA refurbishment, initiated during last two decades, are abandoned. A proposal for decommissioning of the reactor RA in near future, based on economical, technical and legislation reasons, was approved by decisions on the final shutdown of the RA reactor and initiation of the decommissioning process brought by the Governments of the Federal Republics of Yugoslavia and of the Republic Serbia in summer 2002. Initial activities related to decommissioning of the RA reactor are started in 2003 within IAEA TC project with aim to establish in next 2-3 years detail decommissioning plan, including site characterisation, preparing and mastering with appropriate dismantling and decontamination techniques for waste management and to obtain all necessary licenses. At the same time, it is supposed that the spent nuclear fuel will be removed from the site and that appropriate waste management facilities and temporary storage will be established in the Institute with aim to allow uninterrupted decommissioning progress. These initial activities are followed after the establishing the decommissioning team and basic training of the personnel.

The project '**Safe Management of Waste in the Vinča Institute**' has to establish the appropriate organisation structure for initiation works and realisation of the forthcoming activities related to RAW management during transfer of the spent fuel and decommissioning of the RA research reactor. A new RAW processing facility and new temporary storage facility of large capacity ('hangar no. 3') are planned to be finished during first stage of the project. In next stage of the project, characterisation of large volume RAW stored at the hangar no. 1 and its safe packaging and storage at hangar no. 3 is

expected to be done in parallel with the RAW flow from the RA reactor decommissioning activities. Also, about 300 m³ of the liquid LL/ML RAW, stored at the Vinca site in two stainless steel underground storage tanks for almost 40 years, should be evaluated, appropriately treated and safely stored. High intensity radioactive orphan sources will be evaluated at the Vinca site too and properly stored in a well-protected bunker within the RAW storage area.

3. VIND Program activities in 2004

After country public elections in late 2003, new Government of the Republic Serbia was established in March 2004, but the previous Republic's Government has brought appropriate decisions related to the Green Vinca Program in February 2004:

- The only acceptable ('the final') solution for the Republic Serbia is shipment of the SNF from Vinca back to the RF for reprocessing, i.e., no long term storage of the SNF or high-radioactive waste at the country is foreseen in future;
- The 'immediate dismantling' is accepted as the decommissioning strategy to be applied at the RA research reactor;
- Government of the Republic Serbia will establish appropriate funding for the Green Vinca Program that will be managed by the Ministry for Science, Technology and Development, and
- The Vinca Institute of nuclear sciences will be in charge of realization of the Green Vinca Program.

New Government of the Republic Serbia has established a new Ministry of Science and Environmental Protection (MSEP) and had continue to support the previous Green Vinca Program through the new announced the Vinca Institute Nuclear Decommissioning (VIND) Program. The Green Vinca Program was restructured at the Vinca Institute and the VIND Program was proposed to the MSEP in July 2004 for further support. IAEA supported such approach of the MSEP in May and September 2004 and, also, by engaging two additional staff members with primary tasks related to monitoring progress of the TC projects and supporting the organisation structure of the VIND Program.

New VIND Program has included, beside three already existing projects, a new one related to radiation protection. The program was evaluated and accepted by the MSEP in September 2004 and funding of the VIND Program started October 1, 2004 for the following four projects:

- Shipment of the spent nuclear fuel of the RA research,
- Radioactive waste management at the VINCA Institute,
- Decommissioning of the RA research reactor, and
- Radiation protection.

To carry out efficiently activities specified within the projects mentioned above, the "Vinča Institute Nuclear Decommissioning Program" (VIND Program) established new integrated Program Team, assembled from about 40 experts from the Institute and about the same number of technicians. The Project team, lead by the Program manager and the Program Council assembled from the projects leaders and their deputies, works closely in cooperation with the Vinca Institute Director General, officials of the MSEP and the IAEA TC program manager, TC country officer and IAEA technical officers of each project. Close cooperation of the team members with experts and relevant companies from nuclear developed countries is stimulated within all projects. Currently, four interrelated projects of the VIND Program are supposed to be completed in next 10 years at the total cost estimated at 40 million US dollars.

Legal matters related to almost all activities of the VIND Program were identified at very beginning of the (Green Vinca) Program and pointed out to the both Governments to be properly solved in near future. It was recognized that only rudiments of the full-power Regulatory Body exist in the country. Especially, legal matters related to the safe transport of the SNF from the Vinca Institute to the RF through transit countries have to be addressed appropriately, with the help of the IAEA experts,

through bilateral or multilateral government-to-government agreements. Intermediate Regulatory Body of the Republic Serbia was established in the second half of 2004. Work on new integral country law on Nuclear Safety and Radiation Protection was initiated in 2003 and the draft, ready to be submitted to the public opinion and to Republic's Parliament for the approval, was finished in September 2004. This new law is expected to establish a solid ground for a legal introducing of the modern Regulatory Body in the country and to increase the culture level of the nuclear and radiation safety and regulative approach in all activities of the VIND Program.

4. Conclusion

As a general conclusion we can point out that solving the problem of safe disposal of research reactor spent nuclear fuel and decommissioning of a research reactor, including provision of adequate low-level and intermediate radioactive waste disposal site (even temporary one) is a difficult task for a country with no long-term nuclear power program and with limited potentials and resources. Factors that may cause delays or prevent implementation of the above projects are lack of manpower, lack of material resources and necessary equipment, lack of the appropriate legal structure and full and modern regulative in the field, as well as general economic difficulties in the country.

This paper describes process of establishment and initialisation of the Vinča Institute Nuclear Decommissioning (VIND) Program, known earlier as the "Green Vinča" Program. This Program, supported by Government of Republic Serbia, is supposed to solve all problems related to accumulated spent nuclear fuel, radioactive materials associated to the RA reactor facility and radioactive wastes from the research, industrial, medical and other applications generated in previous period which are stored in the Vinča Institute by proper repackaging and removal from the Vinča site to some other disposal site, to be decided yet. Beside that, a research and development program in modern nuclear technologies is carried out with the aim to preserve experts manpower and establish solid ground for new research in nuclear field.

5. Acknowledgements

This work was supported by the Ministry of Science and Environmental Protection of the Republic Serbia through the VIND Program.

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NEW CASK FOR IRRADIATED RODS

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The TNTM MIL will be a new B (U) F packaging, meeting AIEA 1996 regulations, and will be used for transport of at least 10 whole irradiated fuel (UO₂ and / or MOX), damaged or not, in capsules or not.

The fuel characteristics of the rods will be:

	MOX	UO ₂
Maximal burn up	90 000 MWd / t heavy metal	90 000 MWd / t heavy metal
Minimum cooling time	6 months	6 months
Maximal enrichment (in mass)	At least 11 % [Pu / (U+Pu)]	At least 5% (²³⁵ U / U)

Each fuel rod in the packaging may have these characteristics. Moreover, the TNTM MIL design will accept a mixture of MOX and UO₂ rods without any loading plan.

TNTM MIL packaging will enable dry or wet loading and unloading, either in vertical or horizontal position, in order to meet the needs or requirements of Research Reactors or Laboratories, and power plant. Its maximal weight will be under 24 tons (loaded packaging), in order to be adapted for the greater part of laboratories.

For transport purpose, the TNTM-MIL will be tied-down by 4 trunnions on a transport skid, and will be loaded into an ISO container or inside a tarpaulin.

Two shock absorbers will be fixed on the package in transport configuration.

The outer dimensions of the package will be:

- External diameter of the body less than 1000 mm excluding the trunnions, without the shock absorbing covers;
- Overall length will be less than 7000 mm with the shock absorbing covers, and less than 5500 mm without shock absorbing covers.

The internal diameter will be around 165 mm and will have the possibility to be adapted to 220 mm. The useful cavity length will be around 4 700 mm.

The loaded weight of the packaging will be less than 24 tons. The maximal weight of the internal arrangements loaded with the rods will be around 300 kg.

Principle terms of development are:

- Submittal of the safety analysis report to French Competent authorities: before the end of 2006.
- First transport: before the end of 2007.

STRONTIUM AND BARIUM PRECIPITATION AS CARBONATES IN MOLTEN EUTECTIC LiCl-KCl

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ABSTRACT

Precipitation of solution containing SrCl_2 and BaCl_2 in molten LiCl-KCl eutectic have been performed using as precursors carbonates of Li, K and Na. The effect of carbonate cation and temperature has been studied. The main results obtained indicates that the quantitative removal of these cations is not possible in this molten salt mixture, however, a partial precipitation is possible. The solid compound formed consists of a solid solution of carbonate of Sr and Ba. Precipitation is enhanced by the use of K_2CO_3 and the decrease of the temperature.

1. Introduction

Considering the very long term radiotoxicity associated to the storage of the actual spent nuclear and vitrified wastes, coming from the actual reprocessing process PUREX, the radiotoxicity inventory is dominated by the presence of transuranium elements such as Pu (0.9%) and minor actinides (Np, Am and Cm) (0.1%), since the long-lived isotopes such as iodine and technetium constitutes only the 0.1% and the short-lived radioactive elements decay to harmless species over few hundred years.

Nowadays it is being considering another strategy for the management of the nuclear wastes with the aim of resolving the potential problems that guarantee the safety of the repositories for long periods. This strategy that is called "Partitioning and Transmutation" (P&T), consists of separating all the hazard long-lived radionuclides to transmute them in stable or short-lived isotopes by means of neutronic capture or fission processes, reducing this way the mass and volume of high level nuclear waste and its long term radiotoxicity.

Due to the nature of the nuclear fuels that are being proposed to carry out the transmutation, which will have high content of Pu and minor actinides and will reach high burn-ups, the use of pyrometallurgical processes seems a promising option due to the high radiation stability of the molten salts media proposed. Molten salt media that are under consideration are mainly alkali and alkaline earth elements chlorides and fluorides salts [1].

One of the most developed pyrometallurgical process (IFR) consist of the electrorefining, for metallic fuels in molten LiCl-KCl, from the EBR-II reactor, or the electrolysis for other type of nuclear fuels, in order to recover Pu and the minor actinides (Np, Am and Cm) in a metallic form, remaining in the electrolyte the rest of the fission products. After several cycles of separation, the fission products build up in the molten bath, therefore, they have to be removed in order to recycle the molten salts. On the other hand, it is also necessary to transform these elements in a waste form suitable with the actual vitrification materials that are used in the nuclear waste disposal, since chlorides are not compatible

with them [12]. To achieve this goal several techniques are proposed, among them the precipitation of these elements in different forms such as oxides and oxychlorides [13, 14, 15, 16, 17], carbonates [18], phosphates [19, 20], electrochemical separation using liquid cathodes of Pb [21], Al [21, 22] or Zn [23] etc.

Some of the fission products that are considered of interest to study are the Sr and Ba, that although are non-transmutable can be safely enclosed using engineering barriers only. These elements are difficult to separate from the electrolyte since they have similar chemical properties to the metallic cations that constitute the molten salts [11].

The aim of the present work is to study the feasibility of precipitating the fission products Ba and Sr chlorides into carbonates, in the LiCl-KCl eutectic melt, in order to remove them from the salt and dispose them separately.

2. Experimental

The molten salt media used in this study was the LiCl-KCl (59-41% mol) eutectic. Reagents used were LiCl (Aldrich 99.5 %) and KCl (Merck 99.5%). Solutions of Sr and Ba chlorides were prepared from SrCl₂ (Aldrich, 99.99%) and BaCl₂ · 6H₂O (Merck, 99.9%). Concentration of both elements used in all experiments was 1.5 · 10⁻² mol kg⁻¹. As precipitating agents Na₂CO₃ (Aldrich 99.95%), Li₂CO₃ (Merck, 99.9%) and K₂CO₃ (Panreac, 99.9%) were used.

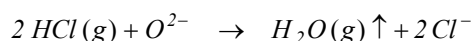
The analysis of the Sr and Ba concentration samples taken from the molten LiCl-KCl was performed by ICP-MS, previously dissolved into HNO₃ solutions (1%). The solid precipitates were washed with distilled water, filtered and dried in a stove at 80°C, then characterised by X Ray diffraction.

3. Procedure

The corresponding amount of LiCl and KCl was mixed and introduced in a crucible of vitreous carbon or Quartz. Quartz has been used in order to observe what happens inside the crucible. Then the Sr and Ba chlorides were added in order to obtain a concentration of 1.5 · 10⁻² mol kg⁻¹. The crucible containing the mixture of salts was placed into a cell made of Quartz, then hermetically closed and finally introduced into a tubular furnace. The temperature inside the molten salt is measured by a thermocouple, type K, sheathed in an Al₂O₃ tube and immersed into the salt.

Before performing the precipitation tests, a pre-treatment of the eutectic mixture was carried out in order to purify it from moisture and residual oxide ions. This treatment consists of a dehydration of the

salt by maintaining the temperature at to 200°C for 8-10, under Ar (g) atmosphere. Then, temperature was raised up to 450°C, temperature used in most of the experiments, also under argon atmosphere. Purification of the salt was performed according to the method proposed by H.A. Laitinen [24] that consists of HCl(g) bubbling (through an Al₂O₃ tube) for 1 hour and then Ar(g) bubbling for 45-60 min in order to remove the HCl dissolved into the molten mixture. This treatment allows to remove the O²⁻ ions content in the molten salt as they react with the HCl(g) according to the following reaction:



El HCl off gas was neutralized by a set of washing flasks containing NaOH solution (4-5 M). The schematic step up is shown in Figure 1.

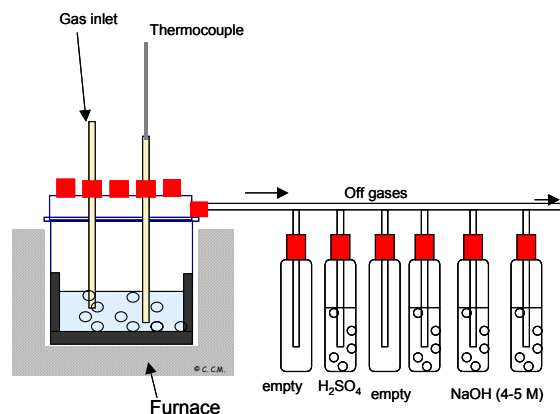


Fig 1. Schema of the experimental set up.

The precipitation was performed by adding successive known amounts of solid carbonate corresponding to a molar ratio $[\text{CO}_3^{2-}]/[\text{Sr}^{2+}+\text{Ba}^{2+}]=1.0$. After each carbonate addition Ar(g) was bubbled for 1.5-2 hours, in order to enhance the carbonate dissociation reaction, the melt was then left settle for about 10 minutes before taken a sample of the molten salt in order to analyse the Sr and Ba concentration in solution by ICP-MS. The used of CO₂ has also been tested and a worsening of the precipitation has been observed.

At the end of the experiment the melt was quenched, the solid precipitated, in those cases in which a precipitate was formed, was washed with distilled water, filtered and dry into a stove at 80°C, then characterised by X-Ray diffraction.

4. Results and discussion

As it was mentioned above three different carbonates were used as precursors, carbonate of Li, K and Na. Next the main results obtained with them are presented.

Na₂CO₃

With this carbonate several precipitation tests were performed at 450°C, the molar ratios carbonate/Sr+Ba tested varied from 0 to 5, 0-10, 0-15 and finally 0-27. For the two first experiments, additions of carbonate corresponded to molar ratios $[\text{CO}_3^{2-}]/[\text{Sr}^{2+}+\text{Ba}^{2+}]=1.0$. For the last two experiments each carbonate additions corresponded to molar ratios of 5 units. In none of these experiments a precipitate formation was observed; this results were confirmed by the analysis of Sr and Ba, that indicated that both cations remained in solution

Li₂CO₃

With lithium carbonate several precipitation test were also performed, the first one was performed adding amounts of Li_2CO_3 corresponding to a total molar ratio, $[\text{CO}_3^{2-}]/[\text{Sr}^{2+}+\text{Ba}^{2+}]$, from 0 to 10. The analysis of both cations in solution indicated that both remained in solution, which was confirmed by the lack of a precipitate formation. Then, additional experiments increasing the amount of carbonate added were performed, the final molar ratios varied from 0-15.5, 0-21.5. In none of them the formation of a precipitate was obtained. Confirmation of this result was carried out measuring the concentration of Sr(II) and Ba(II) in solution.

Finally, an experiment reaching final molar ratio $[\text{CO}_3^{2-}]/[\text{Sr}^{2+}+\text{Ba}^{2+}]$ of 27 was performed, in this case a white precipitate was observed at the bottom of the crucible. The analysis of the solid compound by X-ray diffraction indicated that the solid consisted of a solid solution of carbonate of Ba and Sr, with the following formula: $\text{Sr}_{0.5}\text{Ba}_{0.5}\text{CO}_3$, and Li_2CO_3 . The corresponding diffractogram is shown in Figure 2.

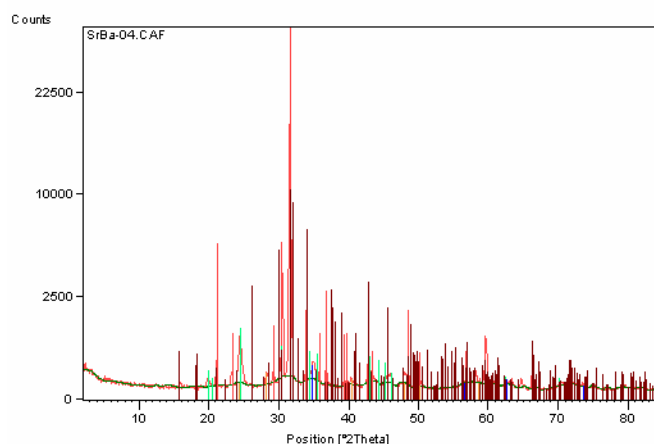


Fig 2. Diffractogram of the solid obtained by precipitation of Sr and Ba chlorides with Li_2CO_3 .

As mentioned before, during the precipitation samples of the molten salt were taken in order to analysed the Sr and Ba concentration. The concentration decrease of both cations in the molten LiCl-KCl is very slow, and at the end of the experiment, molar ratio 27, only a 5.6% of Sr and 14.04% of Ba had precipitated.

K_2CO_3

With K_2CO_3 two initial precipitation tests were performed, in the first one the variation of molar ratios carbonate/Sr+Ba varied from 0-5, with no precipitate formation. In the second one total molar ratio achieved was 11.3, in this case a white precipitate was formed. The solid obtained was characterised by X-Ray diffraction and as it happened with the use of Li_2CO_3 the compound identified was a solid solution of carbonate of Sr and Ba ($\text{Sr}_{0.5}\text{Ba}_{0.5}\text{CO}_3$) and Li_2CO_3 . The presence of Li_2CO_3 could be explained by an exchange of K for Li in excess of carbonate precursor, similar effect have been observed by V.A. Volkovich [20] and C. Caravaca [25] during phosphate precipitation in the same molten bath.

Variation of Sr(II) and Ba(II) in solution for the experiment in which a precipitate was obtained is shown in Figure 3, it can be observed that only a 13.5% of Sr and 16.75 % of Ba were precipitated, remaining the most part of these cations in solution.

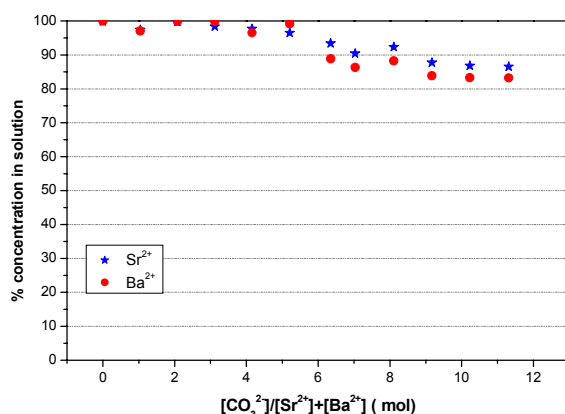


Fig 3. Variation of Sr(II) and Ba(II) concentration during precipitation with K_2CO_3 in LiCl-KCl at $450^\circ C$.

With the aim of reaching a quantitative precipitation of these cations additional tests, increasing the molar ratio, were performed. Results indicate that even if a higher amount of carbonate was added, the precipitation does not improve a lot, and in none of the tests performed a quantitative precipitation of Ba and Sr was achieved.

Effect of the temperature on precipitation

Based on the results obtained with the three carbonates a study of the temperature influence on the precipitation was carried out, for these experiments only K_2CO_3 was used. The temperature range studied varied from $400-500^\circ C$. Results obtained showed that as the temperature increases the molar ratio required to form the precipitate also increases, from values of 9.5 at $400^\circ C$ to ≈ 14 at $500^\circ C$.

5. Conclusions

It has to be mentioned that preliminary precipitation tests were performed using solutions with two cations separately, in this experiments the formation of individual carbonates was not observed.

The results obtained in this work indicate that is not possible to remove quantitatively the Sr and Ba as carbonates in molten LiCl-KCl. However, it is possible to remove part of them jointly using carbonates of Li and K. The analysis of the concentration of both cations in solution shows that the maximum percentage of removal achieved under this experimental conditions was $\approx 17\%$ of Ba and 14% of Sr.

The Sr and Ba precipitation as double carbonate is enhance by the used of K_2CO_3 instead of Li_2CO_3 . It was also observed that the used of Na_2CO_3 does not lead to a precipitate formation. Also the decrease of the temperature improves the precipitation in terms of decreasing the amount of solid carbonate addition, therefore, the volume of the final waste.

The compound formed, that was characterised by X-Ray diffraction, consists of a solid solution of carbonate of Sr and Ba with the formula $Sr_{0.5}Ba_{0.5}CO_3$.

6. Acknowledgements

The work presented here is part of the EC project EUROPART. Authors thanks ENRESA (Spain) for the financial support.

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IR-100 RESEARCH REACTOR AND PROPOSALS FOR SPENT NUCLEAR FUEL MANAGEMENT

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ABSTRACT

IR-100, a pool-type research reactor, is based in the city of Sevastopol, the Ukraine. The core is composed of 47 fuel assemblies (FA). The fuel assemblies use EK-10 fuel elements with a uranium dioxide meat in a magnesium matrix contained in an aluminum cladding. The external diameter of the aluminum cladding is 10 mm and the cladding thickness is 1.5 mm. There are no currently a reliable SNF storage facility and an agreed SNF handling program at IR-100. This report presents proposals and ways for addressing the SNF handling problem.

1. Introduction

The IR-100 research reactor is a unique installation in the Ukraine as there are only two research reactors in the country. One reactor is in Kiev and the other is in Sevastopol. At present time, IR-100 is used to train personnel for the Ukraine's nuclear power complex as part of the Sevastopol National Institute of Nuclear Energy and Industry (SNINEI). The reactor's first criticality was achieved on April 18, 1967 and the reactor was commissioned on October 18, 1967. The reactor designer is Research and Development Institute of Power Engineering (NIKIET), Moscow. Since its first criticality in 1967, the reactor has produced some 1 400 000 kWh of power.

2. IR-100 reactor and its nuclear fuel

IR-100 is heterogeneous, light water moderated and cooled pool-type research reactor. Data and characteristics of IR-100 reactor are presented in Table 1.

Reactor tank* diameter	1800 mm
Reactor tank height	4400 mm
Thermal power	200 kW
Maximum fuel cladding temperature	100°C
Maximum core outlet temperature	55°C
Reflector	Two layer graphite with the total thickness 400 mm
Core	Hexagonal prism with the height of 500 mm and the circumscribed diameter of 460 mm. Total weight of U-235 in the core is 2.66 Kg

* Reactor tank is installed in a well within the biological shielding.

Table 1. Data and characteristics of IR-100 reactor

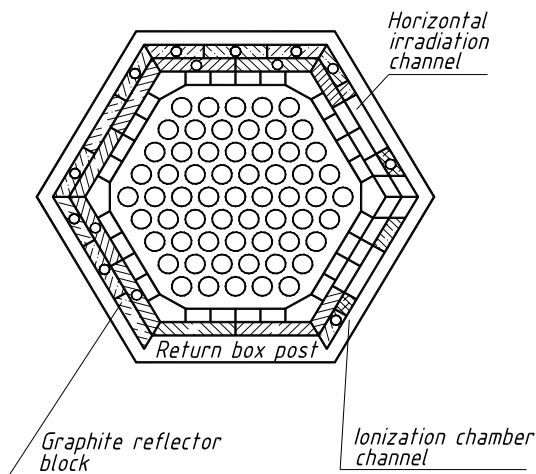


Fig.1. Core configuration of IR-100 reactor

The reactor core (Fig. 1) is composed of 47 fuel assemblies, 35 graphite displacers and one beryllium block. FA is a shroudless assembly of 7 EK-10 rod type fuel elements forming a triangular structure with a pitch of 17 mm. There is a tail nozzle in the FA for the FA positioning in the core grid and a head nozzle for FA gripping during loading and unloading. The main characteristics of fuel assembly and fuel element (FE) are presented in Table 2. At present time, IR-100 has a new core with S-36 fuel elements in fresh fuel storage facility. Fuel composition of S-36 fuel elements is uranium-aluminum alloy in aluminum matrix having enrichment of 36% uranium-235. Both types of fuel elements (EK-10 and S-36) have the same geometry and dimensions.

FA circumscribed diameter	44mm
Active part height	500mm
FA flow area	7,7 cm ²
Number of fuel elements (FE)	7
FE cladding material	Aluminum alloy
Fuel composition	UO ₂ in magnesium matrix
Enrichment in uranium-235	10%
FE diameter	10mm
FE cladding thickness	1,5mm

Table 2. FA and FE main characteristics

3. Handling of irradiated nuclear fuel in IR-100 operation

There is a special wet facility within the biological shielding for the spent fuel storage.

Within the storage facility, the biological shielding is:

- distilled water in the axial direction;
- concrete in the radial direction;
- a protective container during FA loading, unloading and transportation.

The main characteristics of spent fuel storage are presented in Table 3.

Storage facility diameter	1200 mm
Storage facility height	4500 mm
Number of cells	114 The cells make up a uniform triangular grid with a pitch of 100mm.
Water working level	4020 mm
Minimum shielding water layer during FA loading and unloading	2000 mm
Minimum shielding water layer during FA storage	3000 mm

Table 3. Wet storage facility characteristics

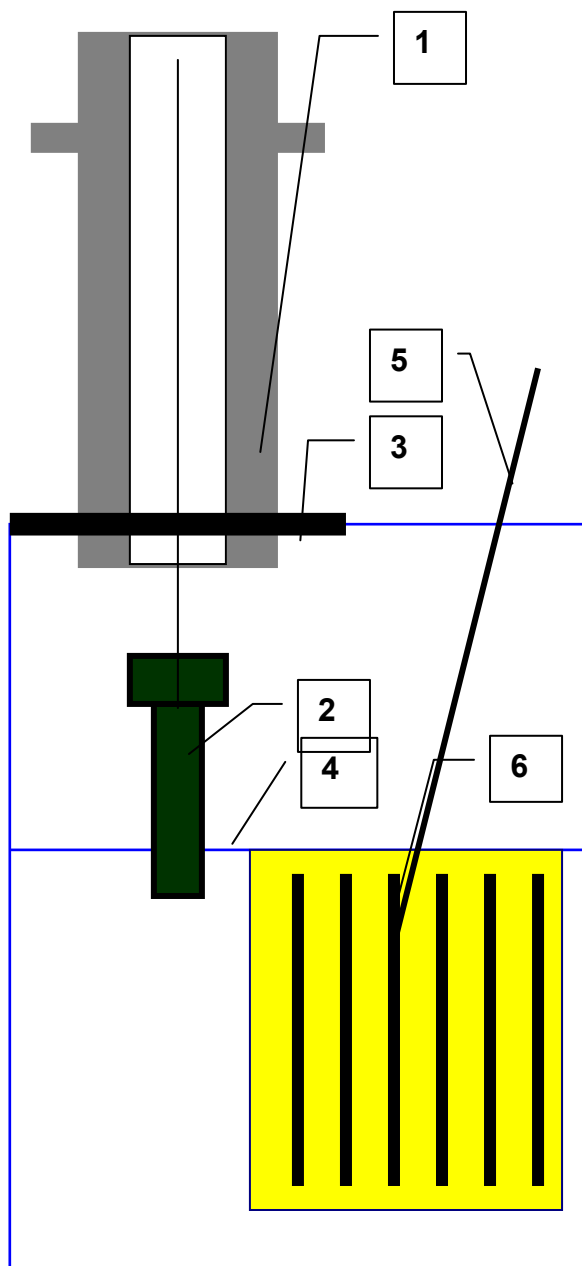


Fig. 2. Scheme of the FAs loading and unloading

The technology of FAs loading and unloading incorporates the following stages (Fig. 2):

The container (pos. 1) with a canister (pos. 2) is installed on the upper rotary plate (pos. 3).

The canister is lowered down to the reactor upper edge level (pos. 4).

A manual clamp grip (pos. 5) is introduced to inside of the reactor through a hole in the upper rotary plate.

The clamp grip transports the selected FA (pos. 6) and loads it into the canister.

The canister with the FA is further drawn into the protective container.

The latter is transported to the SFA storage facility. The FA is loaded into a storage cell using the same flowchart.

The storage facility body is a cylindrical welded vessel of AD1 aluminum alloy with the internal diameter of 1200 mm and the height of 4270 mm.

The grid for the FAs arrangement is made of 2 plates interconnected through tubes welded into the plate holes. To load an FA into the storage facility, the FA tail nozzle is fit in the grid tube so that to keep the FA spacing in storage and avoid the FA displacement.

According to calculations, K_{∞} does not exceed 0.423 ± 0.007 for an infinite number of the IR-100 FAs kept in water with a spacing of 100 mm over the triangular grid.

It should be noted that at present time there is not a single spent fuel assembly (SFA) loaded into the storage facility. The explanation is that the fuel burnup reached during operation is much below the permissible value.

4. Core leak-tightness monitoring

The radiation situation in working rooms during the reactor operation depends on the ionizing radiation penetrating through the biological shielding, the primary coolant, the air pumped by the special ventilation system and activated structural materials.

The coolant activity is caused by:

- the coolant and its impurities activation under neutron effect;
- activation of structural materials corrosion products.

The reactor core state is analyzed and predicted based on the magnitude and nature of the dry residue activity change, the total of the iodine isotopes in the coolant and the activity of radioactive gases in

the release through special ventilation. The coolant samples are taken during the reactor rated power operation (after not less than 4 hours).

The state of the reactor FA integrity is assessed based on the criteria presented in Table 4.

Core state	Major indices		Additional indices
	Specific activity of the coolant in terms of dry residue, Ci/l	Specific activity of iodine in the coolant, Ci/l	Volume activity of the air in the release system, Ci/l
Normal	$\leq 1.5 \cdot 10^{-7}$	$\leq 3 \cdot 10^{-8}$	$\leq 2 \cdot 10^{-7}$
Acceptable	$1.5 \cdot 10^{-7} - 1.5 \cdot 10^{-4}$	$3 \cdot 10^{-8} - 1 \cdot 10^{-5}$	$2 \cdot 10^{-7} - 5 \cdot 10^{-7}$
Inadmissible	$> 1.5 \cdot 10^{-4}$	$> 1 \cdot 10^{-5}$	$> 5 \cdot 10^{-7}$

Table 4. Analysis of the FA leak-tightness state (main parameters)

An additional criterion of the fuel cladding failure is an increase in the activity of gaseous fission products (largely Xe^{135}) in the coolant by not less than twice as compared to the previously recorded value with the same activity of the total iodine isotopes.

Design of IR-100 fuel elements proved to be very reliable and as a result not a single non-leak-tight FA has been found in the reactor core during 38 years of IR-100 operation.

5. Proposals developed to secure a safe and reliable SNF storage

As has been noted above, none of the SFAs has been loaded into the storage facility from the reactor. At the same time, it should be noted that the SFAs were loaded in the reactor about 40 years ago and in the case of SFAs transferring to the existing wet storage it would be a prolongation of exposed time of water – cladding direct contact.

As a consequence the possibility of fuel cladding failure because of corrosion processes should be taken in consideration.

In order to reduce the possibility of fuel elements failure, the nuclear fuel after being unloaded from the reactor should be transferred for safe interim “dry” storage enabling their following transfer for reprocessing. It is reasonable that the interim “dry” storage of SNF and its preparation for being transferred for reprocessing should be arranged for on the IR-100 complex site. As such, the SNF reprocessing may be successfully organized at NPO “MAYAK”, Russia.

The SNF should be brought to a safe condition to ensure nuclear and radiation safety during storage.

The criteria of safe SNF storage are as follows:

- the correct selection of the SFA arrangement geometry in the storage facility;
- the availability of a biological shielding for personnel;
- the availability of a system of organizational and technical measures to ensure the SNF state monitoring;
- a reliable system of physical safety at the SNF handling installation.

Two proposals have been considered to secure a safe and reliable SNF storage: storage in dry stationary storage facility and storage in dual-purpose containers.

5.1. SNF intermediate storage in a stationary facility

The storage facility is a metal tank installed on a concrete platform or in a well. The loading into the storage facility is performed through the upper plate into the cells for the SNF arrangement in the tank. The cell number corresponds to the number of the IR-100 SFAs. SFAs should be placed in special canisters to avoid the SNF contacts with water used as biological shielding. The canisters are installed in the storage cells directly prior to the SFAs unloading from the reactor for storage. The SFAs are loaded into canisters using a shipping cask based on a technology tried out by the reactor personnel. The SNF condition is monitored using special systems currently used as part of the IR-100 laboratory. The state of the SNF in long-term storage may be examined during the storage facility operation.

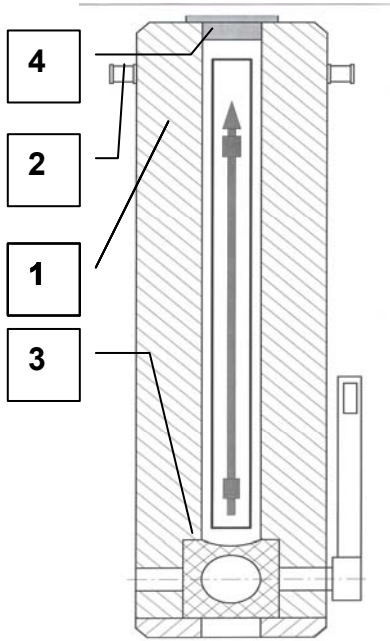


Fig.3. Scheme of the protective container

If it is required to remove the SNF for reprocessing, the special canisters with FA are sealed, loaded into special shipping casks and onto a vehicle.

5.2. SNF intermediate storage in dual-purpose containers

Dual-purpose containers (protective container) are used for interim storage and transportation of SNF to reprocessing points.

Described hereinafter is a potential design of a protective container (Fig. 3) with the SFA loading from below. This simplifies considerably the SFA handling technology. A protective container is a hollow thick-wall cylinder (pos. 1). A special crosspiece is used to transfer the container by a hoist. The container body has two pivots (pos. 2) in its upper portion to be gripped with the crosspiece.

The clamp grip for the canisters with SFA is led to the inside of the container from the upper end. The container lower portion has a rotary slide (pos. 3) used to seal the internal space after the SFA canister loading. A handle with two fixed positions of “OPEN” and “CLOSED” is used to operate the slide. The internal space is sealed from the above using a process lid (pos. 4). Pressure bolts are used to seal the process lid.

A receiving yard with special outfit is used for interim storage of containers with SFAs. The yard has access road and a hoist for the loading of containers onto vehicles.

6. Conclusion

In the course of IR – 100 reactor operation, 47 FAs with road-type FEs in aluminum cladding were used. During 38 years of reactor operation, not a single non-leak-tight FA has been found in the reactor core. In accordance with the design, the SFAs should be placed into a wet storage facility, but at present time there is not a single SFA loaded into existing storage facility.

The SFAs transferring to the wet storage (the prolongation of exposed time of water – cladding contact) may cause the corrosion failure of SFAs.

Two options have been considered to raise the reliability of storage:

- storage in dry stationary storage facility
- storage in dual – purpose containers.

The first option is more expensive, and due to a low number of SFAs, the preferable option is storage in dual – purpose containers.

In both options the intermediate storage of SFAs is the first step for further reprocessing.

~~EN~~ ~~PREM~~ 2005

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