

Production of Mo⁹⁹ in Europe :

Status and Perspectives

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NUCLEAR MEDECINE

- 10.000 hospitals in the world use radioelements
 - 90 % for diagnostic (cardiology, oncology, neurology, ...)
 - 10 % for therapy/palliative care (oncology, cardiology, ...)
- Radioelements are produced by fission of HEU (Mo^{99} , I^{131} , ...) in test reactors or by activation of targets in reactor and cyclotrons (I^{125} , I^{123} , ...)
- Fission $\text{Mo}^{99}/\text{Tc}^{99\text{m}}$ is used for 80 % of diagnostic procedures (\pm 25 million procedures per year). Fission I^{131} and $\text{Sr}^{90}/\text{Y}^{90}$ are currently used for therapy. Activation $\text{W}^{188}/\text{Re}^{188}$ is under qualification for therapy.
- Essential criteria for medical isotope supply :
 - High and proven quality
 - Continuous and reliable timely delivery
 - Economical supply
 - Compliance with all regulatory requirements



RADIOISOTOPES PRODUCTION

Selection of radioisotopes :

- Medical criteria : Diagnostic (low doses) (γ)
Therapy (long life) (β , $\beta\gamma$)
- Production criteria : Cyclotron (activation)
Short life
Reactor (activation or fission)
Logistic requires long life or generator process
- Chemical/physical criteria
 - carrier free single isotope/radioisotope (Sr-89 decay/Sr-90)
 - low level of impurities (use of HEU); high volumic activity
 - availability of reactor/cyclotron with specific characteristics (eg. Re-188, Pd-103, ...)

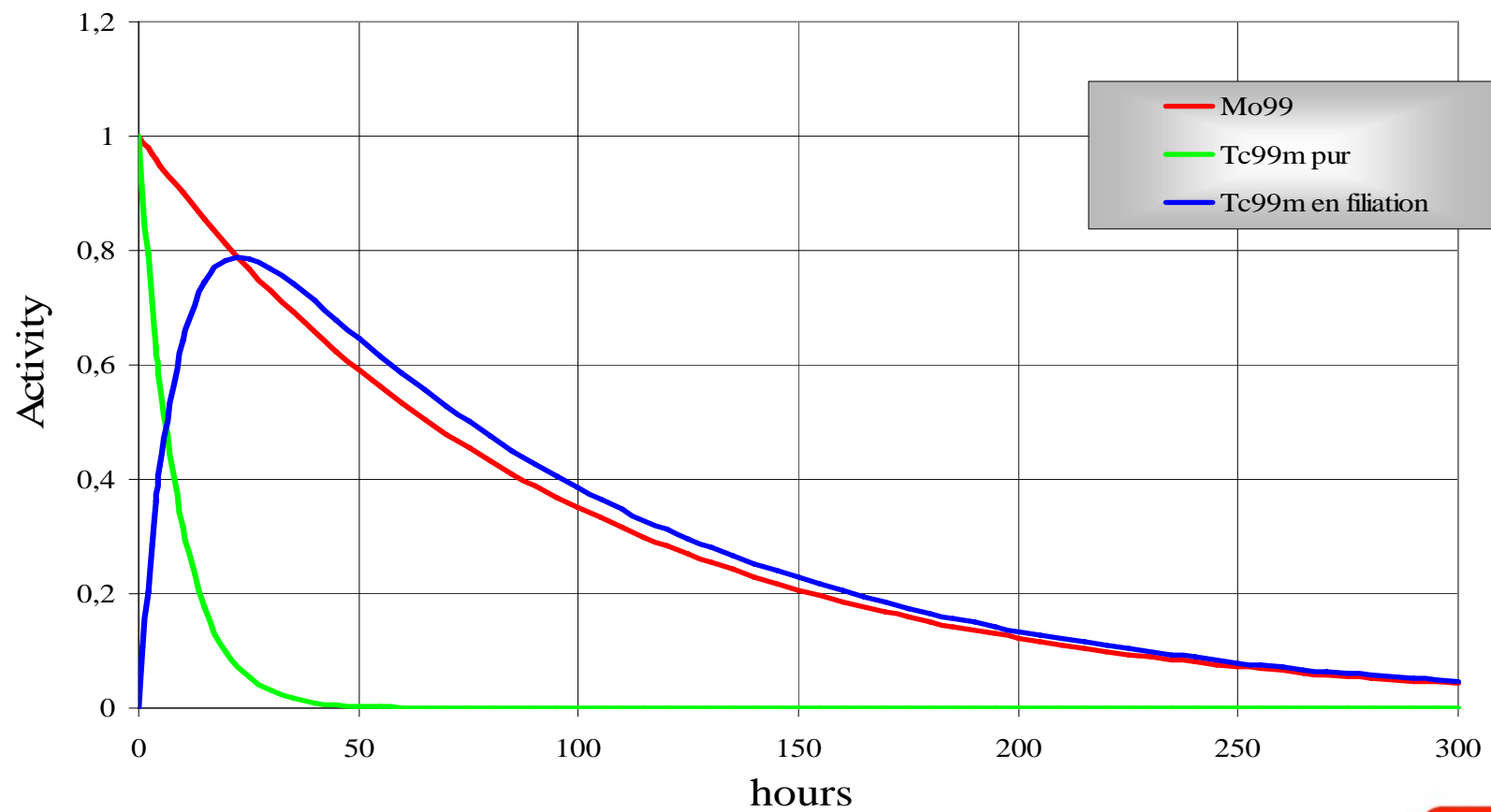


PRODUCTION OF RADIOELEMENTS FOR MEDICAL APPLICATION

| ORIGINE | DIAGNOSTIC | | THERAPY | |
|----------------------|-------------------|------------------|----------------|------------------|
| Cyclotrons | Isotope | Half life | Isotope | Half life |
| | Gallium-67 | 3,2 days | Palladium-103 | 17 days |
| | Indium-111 | 2,8 days | | |
| | Iode-123 | 13,3 hours | | |
| | Thallium-201 | 3,0 days | | |
| | Krypton-81 | 13 seconds | | |
| | Fluor-18 | 118 minutes | | |
| Test reactors | Isotope | Half life | Isotope | Half life |
| | Chrome-51 | 27,7 days | Césium-137 | 30,0 years |
| | Fer-59 | 44,5 days | Cobalt-60 | 5,3 years |
| | Iode-125 | 59,9 days | Iode-131 | 8,0 days |
| | Indium-113 | 1,6 hours | Iridium-192 | 74,0 days |
| | Molybdenum-99 | 66,0 hours | Palladium-103 | 17 days |
| | Technétium-99m | 6,2 hours | Phosphore-32 | 14,3 days |
| | Iode-131 | 8,0 days | Tungstène-188 | 69,0 days |
| | Xénon-133 | 5,2 days | Rhénium-186 | 3,7 days |
| | | | Rhénium-188 | 16,9 hours |
| | | | Strontium-85 | 64,8 days |
| | | | Strontium-89 | 50.5 days |
| | | | Strontium-90 | 28 years |
| | | | Yttrium-90 | 2,7 days |



RADIOACTIVE DECAY



REACTOR ISOTOPES PRODUCTION

- Irradiation specification :
 - Flux level (thermal – perturbed) : Mo^{99} ($\geq 1\text{E}14$)
(depend on σ_{eff}) W^{188} ($\geq 1\text{E}15$)
 - Duration (depend on $T^{1/2}$) $\text{Mo}^{99(66\text{h})}$ ($\geq 150\text{h}$)
 $\text{W}^{188(69\text{d})}$ ($\geq 1000\text{h}$)

Fission Products :

Production scheme for Mo^{99} :

- procurement of HEU targets (storage at reactor)
- irradiation in reactor 150 h 6750 Ci
- loading container/transport 30 h
- processing hot cells 12 h 4550 Ci
- shipment of Mo 12 h
- production generator (distribution) 12 h
- hospital use (6 days) 120 h 1000 Ci

Other fission products : I-131, Xe-133, Sr-90/Y90

Activation Products (± 40 isotopes) :

Sr-89*, I-125*, I-131, Ir-192*, Re-186*, Re-188, Lu-177*, Sm-153*,
Ho-166*, ...

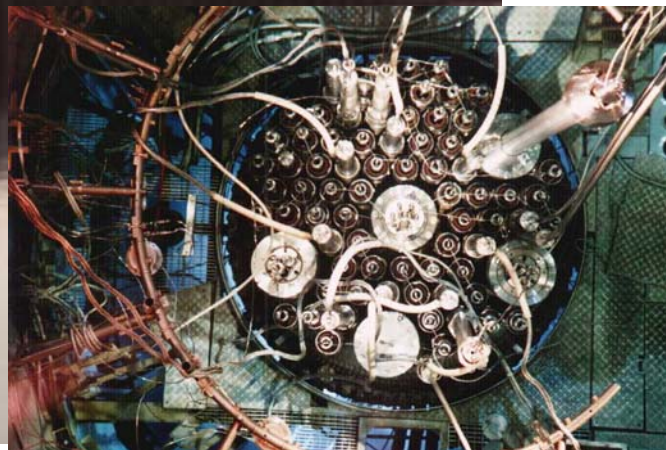
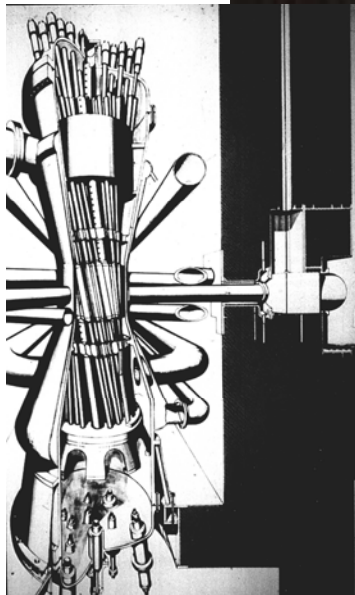
* typical isotopes produced at HFR



Table 4. Research reactors producing radioisotopes in 2002

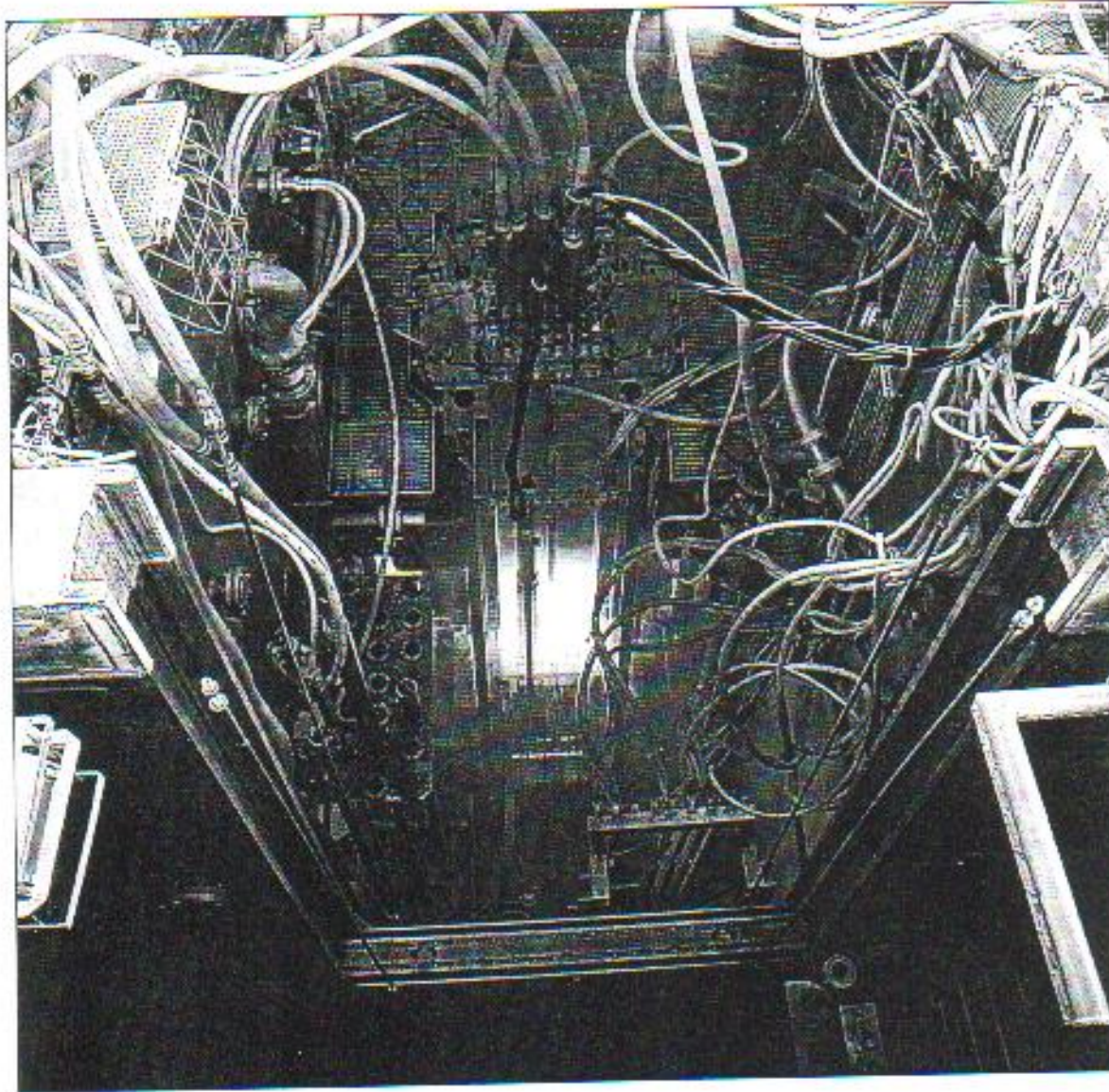
| Country | Reactor Name | Location | Start up year | Planned for S/D by 2008? | Power (MW) | Maximum thermal flux (n/cm ² -s) | Isotopes produced in 2002 | New isotopes planned for 2003-2005 |
|-----------------|--------------|-------------|---------------|--------------------------|------------|---|--|--------------------------------------|
| Australia | HIFAR | Menai | 1958 | Yes ¹ | 10 | 1.4×10^{14} | ⁵¹ Cr, ⁶⁰ Co, ⁹⁰ Y, ⁹⁹ Mo, ¹³¹ I, ¹⁹² Ir, ¹⁹² Yb | ¹⁷⁷ Lu |
| Bangladesh | TRIGA Mk II | Savar | 1986 | NR | 3 | 5.6×10^{13} | NR | NR |
| Belgium | BR2 | Mol | 1961 | No | 100 | 1×10^{15} | ⁹⁹ Mo, ¹³⁵ I, ¹³¹ I, ¹³² Xe, ¹⁵³ Sm, ¹⁷⁷ Lu, ¹⁸⁶ Re, ¹⁹² Ir, ²⁰³ Hg | NR |
| Brazil | IEA-R1 | Sao Paulo | 1957 | No | 5 | 1.17×10^{14} | ²⁵ S, ⁵¹ Cr, ⁶⁰ Co, ⁸² Br, ¹⁵³ Sm, ¹⁹² Ir, ²⁰³ Hg | ⁹⁹ Mo, ¹³¹ I |
| Canada* | NRU | Chalk River | 1957 | NR | 135 | 4×10^{14} | ¹⁴ C, ⁶⁰ Co, ⁹⁹ Mo, ¹²⁵ I | NR |
| Chile | RECH-1 | Santiago | 1974 | No | 5 | 7×10^{13} | ⁸² Br, ^{99m} Tc, ¹³¹ I, ¹⁵³ Sm, ¹⁹² Ir | NR |
| Chinese Taipei* | THOR | Hsinchu | 1961 | NR | 2 | 3.6×10^{15} | ¹³¹ I | NR |
| China* | HFR | Chengdu | 1979 | NR | 125 | 6.2×10^{14} | ⁶⁰ Co, ^{99m} Tc, ¹³¹ I, ¹⁵³ Sm, ¹⁹² Ir | NR |
| | MJTR | Chengdu | 1991 | NR | 5 | 8×10^{15} | | |
| Czech Republic | LWR-15 | Rez | 1957 | No | 10 | 1.5×10^{14} | ⁹⁰ Y, ¹⁵³ Sm, ¹⁶⁹ Tm, ¹⁹² Ir, ²⁰³ Hg | NR |
| Egypt | ETR-1 | Cairo | 1962 | Yes | 2 | 1.5×10^{13} | ³² P, ¹³¹ I | ¹⁵³ Sm, ¹⁸⁶ Re |
| | ETR-2 | Cairo | 2003 | No | 22 | 2.8×10^{14} | | ¹⁸⁸ W |
| France | ORPHEE | Saclay | 1980 | No | 14 | 3×10^{14} | ⁹⁰ Y, ⁹⁹ Mo, ¹³¹ I, ¹³² Xe, ¹⁴⁰ La, ¹⁵³ Sm, ¹⁶⁹ Er, ¹⁸⁶ Re | NR |
| | OSIRIS | Saclay | 1966 | No | 70 | 2.7×10^{14} | | |





BR2 REACTOR





HFR REACTOR



PRODUCTION OF FISSION RADIOISOTOPES IN THE WORLD

| Reactor | Producer | Operation d/year | Current production (%) | | Peak Capacity Production |
|---------------|---------------------|---------------------|------------------------|--------------|--------------------------|
| | | | <i>Mo-99</i> | <i>I-131</i> | <i>Mo-99</i> |
| NRU | MDS- NORDION | ± 315 | 40 | - | 80 |
| HFR | TYCO- HEALTHCARE | ± 290 | 20 | - | 30 |
| | IRE | | 10 | 30 | 20 |
| BR2 | TYCO- HEALTHCARE | ± 115 | 5 | - | 15 |
| | IRE | | 4 | 15 | 20 |
| OSIRIS | IRE | ± 220 | 3 | 15 | 20 |
| FRJ-2 | IRE | ± 210 | 3 | 15 | 10 |
| SAFARI | NTP | ± 315 | 10 | 25 | 45 |
| OTHERS | -- | -- | 5 | - | 10 |
| | | | 100 | 100 | 250 |

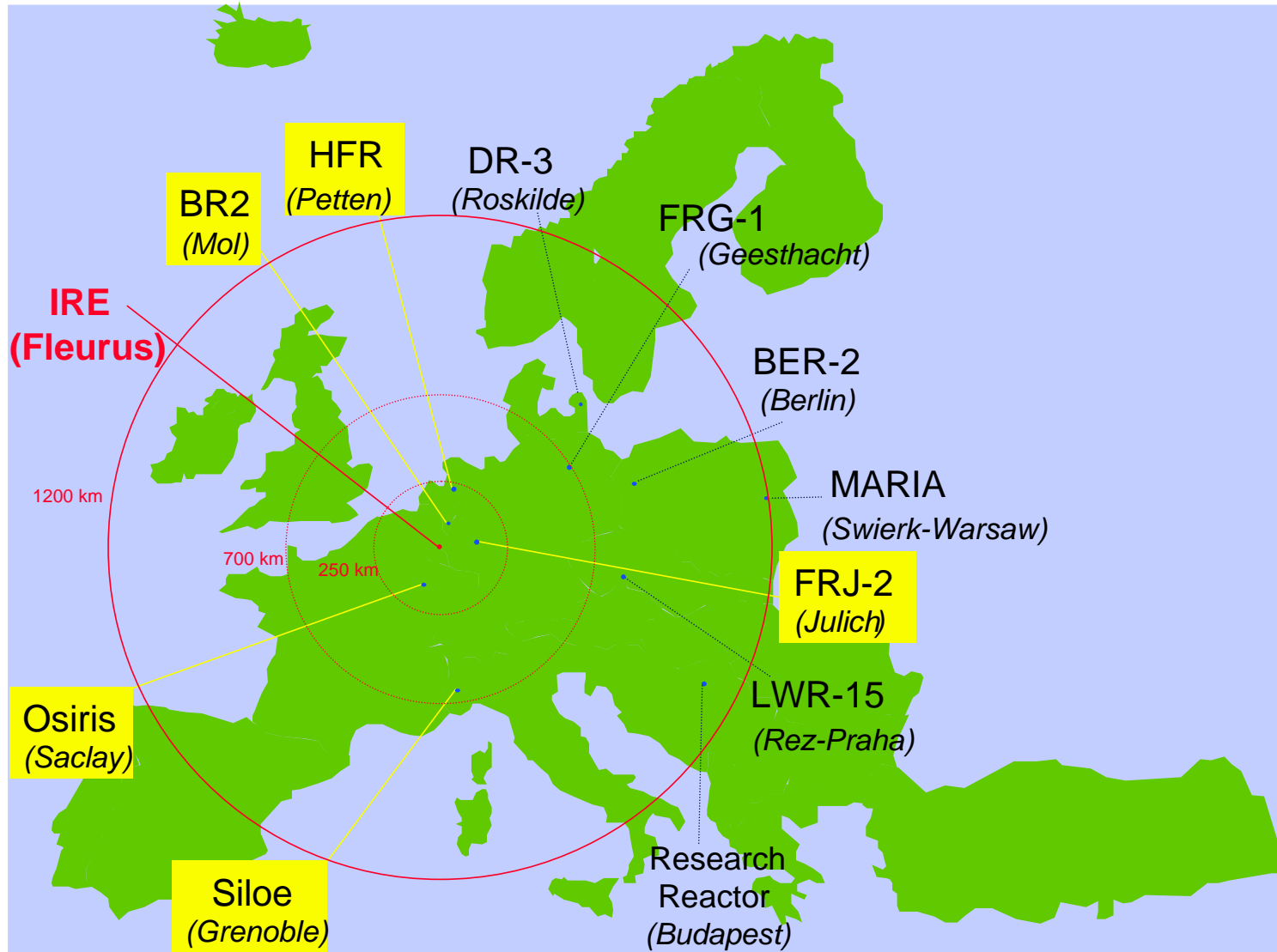


| Reactor | HFR | OSIRIS | FRJ-2 | BR2 | SAFARI |
|-------------------------|----------------|---------------|--------------|------------|---------------|
| Cycle length (days) | 25 | 26-27 | 21-35 | 21-28 | 17-24 |
| N° Cycle/year | 11-12 | 8-9 | 9-10 | 5 | 14 |
| Normal shutdown (days) | 3 | 8-10 | 7-14 | 21 | 4-5 |
| Special shutdown (days) | 1) 24 2) 24 | 80 | 21 35 | 35 | - |
| Cycle start | Friday | Wednesday | Monday | Tuesday | Thursday |
| Cycle end | Monday | Sunday | Sunday | Tuesday | Saturday |
| Max. Annual EFPD | 290 | 220 | 210 | 115 | 315 |

Reactor Operating Conditions



REACTORS SITES





TRANSPORT OF IRRADIATED TARGETS



| January 2003 | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | |
|--------------|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|--|--|----------|--------|-----------|
| | W | Th | F | Sa | Su | M | Tu | W | Th | F | Sa | Su | M | Tu | W | Th | F | Sa | Su | M | Tu | W | Th | F | Sa | Su | M | Tu | W | Th | F | | | | | |
| | 01 | 02 | 03 | 04 | 05 | 06 | 07 | 08 | 09 | 10 | 11 | 12 | 13 | 14 | 15 | 16 | 17 | 18 | 19 | 20 | 21 | 22 | 23 | 24 | 25 | 26 | 27 | 28 | 29 | 30 | 31 | | | | | |
| R-2 | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | Studsvik | | |
| HFR | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | Petten | |
| OSI | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | Saclay |
| BR2 | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | Mol |
| FRJ-2 | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | Jülich |
| Safari | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | Pelindaba |

| February 2003 | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | |
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| March 2003 | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | |
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| April 2003 | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | |
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| May 2003 | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | |
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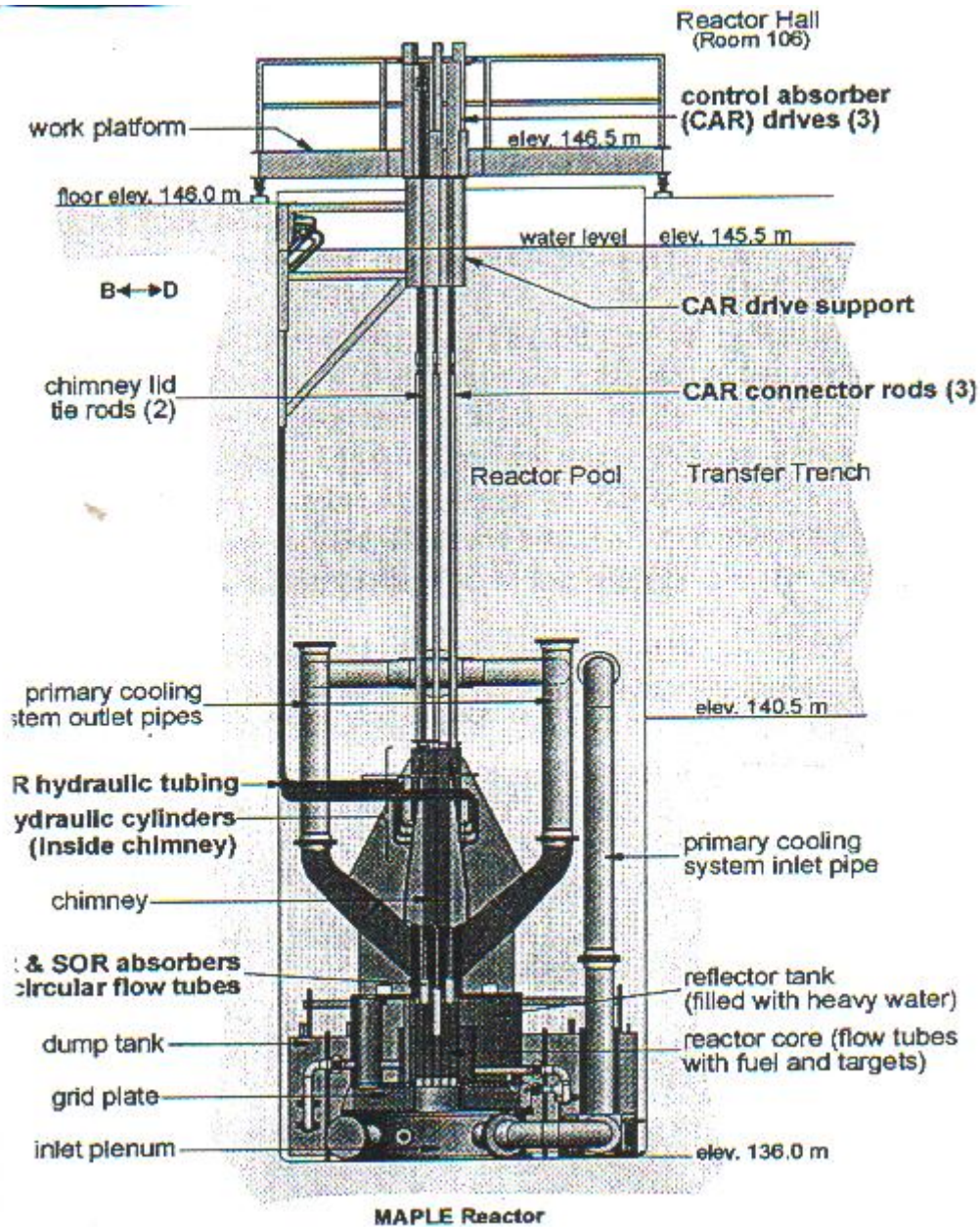
| June 2003 | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | |
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| REACTORS COUNTRY | MAPLE (2) CANADA | RRR AUSTRALIA | MPR EGYPT | FRM-2 GERMANY | JHR France |
|--------------------------------------|--|--|---|---------------------------------------|--|
| Start Project | 1985 | 1997 | ? | 1974 | 1995 |
| Start construction | 1996 | 2002 | ? | 1993 | ? |
| Power operation | 2006 ? | 2006 ? | 2003 | 2004 | 2014 ? |
| Initial Investment 10 ⁶ € | 40/30 | 180 | ? | 250 | ? |
| Revised Investment 10 ⁶ € | 100/75 | ? | ? | 350 | ? |
| Power (Mwth) | 10 | 20 | 22 | 20 | 100 |
| Thermal flux n/cm ² .sec | 2.10 ¹⁴ | 3.10 ¹⁴ | 2.710 ¹⁴ | 8.10 ¹⁴ | 7.10 ¹⁴ |
| Fuel | LEU rods | LEU plates | LEU plates | Plates Cyl. | Plates Cyl. |
| Reflector | D ₂ O | D ₂ O | Be | D ₂ O | Be |
| Operation (days) Power/Shutdown | 2 x 183/0 | 336/ | ? | 260/21 | ? |
| Testing facilities | 0 | Cold/Therm n T 19 conveyors 17 rigs | n beams T 1 fuel loop | Cold/Therm/fast n 9 n beam T | 10 fast n T 10 thermal n T |
| Applications | RI production Mo ⁹⁹ , I ¹³¹ , Xe ¹³³ I ¹²⁵ | RI production Physics BNCT Training | RI production Fuel test Physics Training | Physics Material test n Therapy | RI production Fuel test Material test Physics |

New Test Reactors





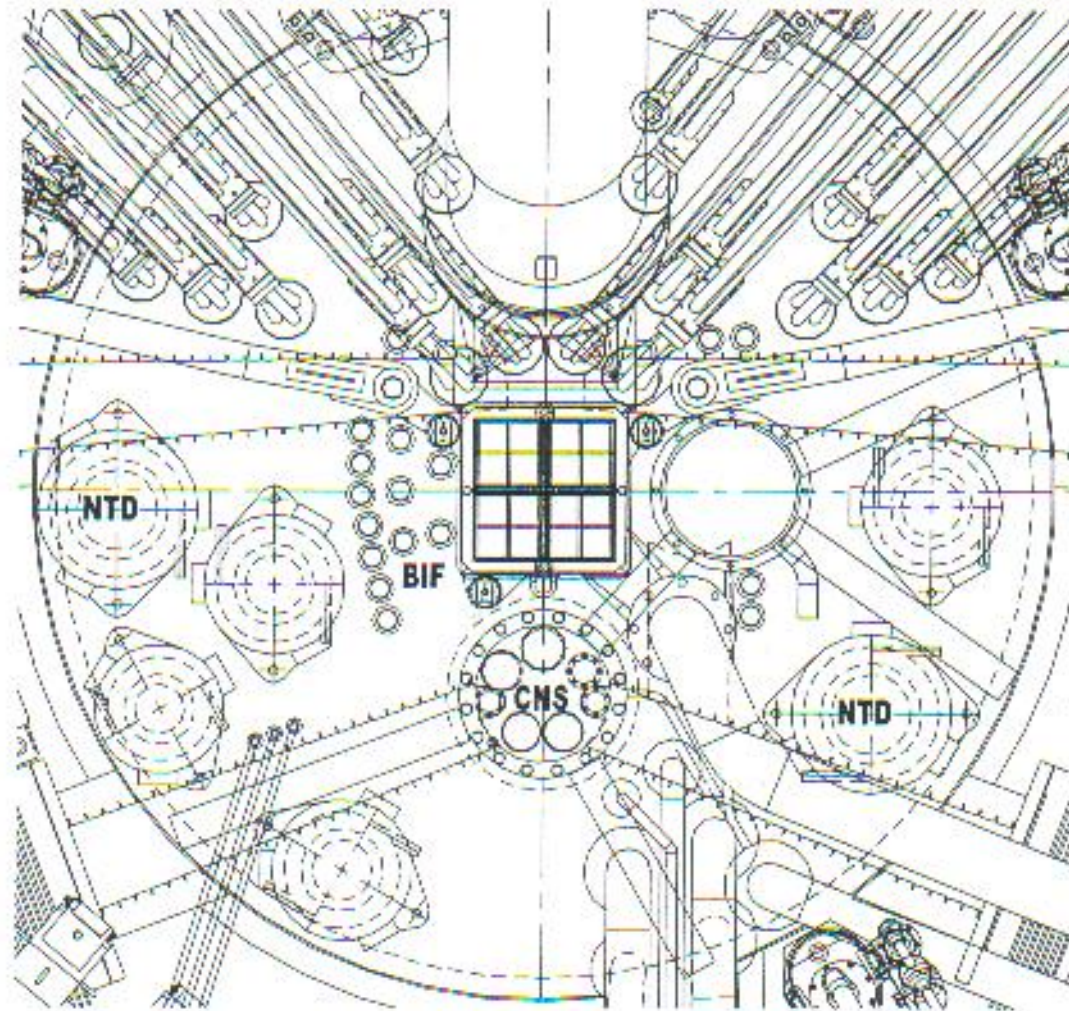
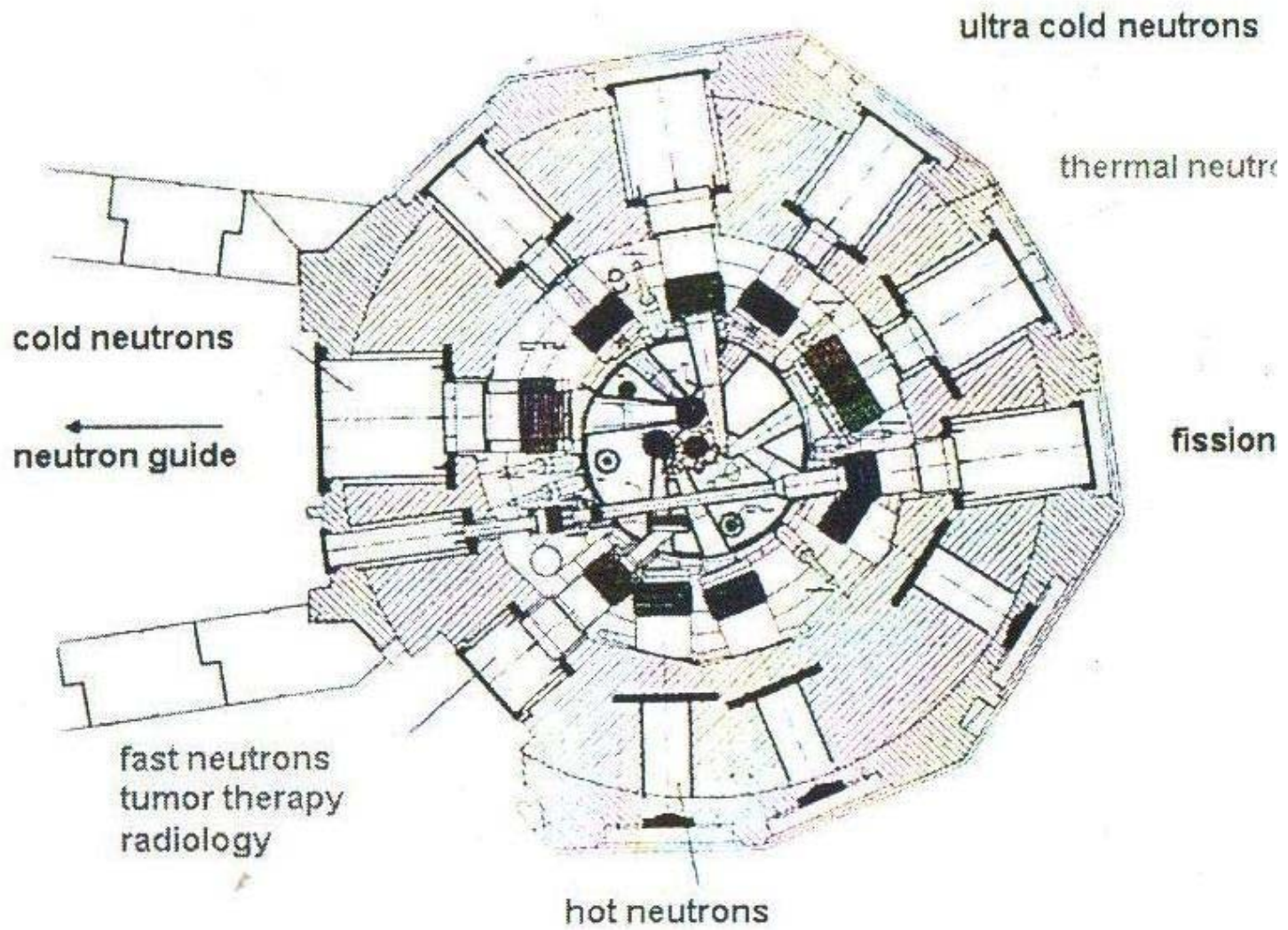


Figure 4. Upper View of the Reflector Tank and main Irradiation Facilities.

RRR REACTOR





FRM-2 REACTOR



| INSTALLATION COUNTRY PROJECT | MIPR USA/BELGIUM (B&W – IRE) | ARGUS RUSSIA/USA (Kurchatov/TCl) | ADONIS BELGIUM (SCK/IBA) | CONCEPT RUSSIA/USA (KHARKOV/ANL) |
|---|--|---|--|--|
| Start Project | 1995 | 1981-1999 | 1995 | 2004 |
| Concept | Solution Reactor UO ₂ (NO ₃) ₂ | Solution Reactor UO ₂ (SO ₄) | ADS P/n conversion Pb-Bi target | ADS e ⁻ /n conversion W or U target |
| Power/Flux | 200 KW | 20-50 KW | 5.10 ¹³ n/s.cm ² | 1.10 ¹³ n/s.cm ² |
| Accelerator | | | 150 Mev | 150-200 Mev |
| Fuel | HEU | HEU-LEU | HEU | HEU-LEU |
| Reflector | - | - | - | Be |
| Keff | 1.0 | 1.0 | 0.98 | 0.98 |
| Mo ⁹⁹ production Ci6d/W | 1500 | 250 | 1000-3000 ? | 200-600 ? |
| Quality | Questionnable | Questionnable | Standard | Standard |
| Main Concern | Radiolysis (H ₂) Vessel corrosion Purification Mo Waste Not proven | Radiolysis (H ₂) Vessel corrosion Purification Mo Waste Prototype | Cyclotron Design Pb-bi target Waste (Po210) Not proven | ? Not proven |

***New Reactor/System Concept & Prototype
for Fission Radioisotopes Production***



THE FUTURE

- Radioisotope demand increasing by 5 to 10 %/year
- Bottle neck in reactor availability in the Future
 - Project MAPLE 1 & 2 in Canada : stand-by
 - Definitive shutdown scheduled in Europe for :
 - STUDEVIK reactor by July 2005
 - FRJ-II reactor by May 2006
 - New reactors in Europe :
 - FRM-II : limited use for isotope production
 - JHR : possible use in network by 2014 for back-up
- Definitive need for 1 new reactor in the European network by 2010-2015 (Preferably fully dedicated for RI production and R&D)
- A network of multipurpose reactors designed for R&D and advanced research has to be kept available for possible RI production back-up.
- Possible need for a second new reactor dedicated to RI production and R&D by 2015-2020.



OPTIMUM REACTOR CHARACTERISTICS FOR PRODUCING SHORT HALF LIFE RADIOISOTOPES TO BE USED IN NUCLEAR MEDECINE

1. **Very high availability** : (360 d/y, minimum outage 3-4 days)
2. **Very high reliability** : minimum risk of SCRAM & High Safety Level
 - Simple and proven design, no fancy features (neutron beams, test loops, ...)
 - Standard Pool reactor and not a prototype
 - Reactor design and network providing for diversity more than redundancy
3. **Performance**
 - Production capacity : minimum 50 % of World Market (SOS requires availability of 200 %)
 - High neutron flux (1 to 5 10^{14} n/cm².s) and predictable production yield
 - Large number of RIG's – easy handling of targets at power
4. **Economics**
 - Low investment cost (Max 100 M€)
 - Low operating cost (operation, maintenance, fuel, waste,...) (Max. 10 M€/year)
 - Possible additional revenues from irradiation of material, R&D for new isotopes, ... (by use of simple RIG's)



MING REACTOR

R.I. Production dedicated : proven design (ref. MPR30, FRG-1, BER-II)
(plate fuel) (be reflector)

Power : 10 MW Flux : $1.4 \cdot 10^{14}$ n/cm².sec

Operation : 3 cycle x 120 EFPD

Irradiation capacity : min. 6000 Ci_{6d}/week

Irradiation facilities : RIGS in Be reflector only – no n beam tubes

Investment cost : 60 M €₁₉₉₇ underground design + 15 M€₁₉₉₇

Operating cost : 7 M €₁₉₉₇/year

Construction time : 3.5 – 5 years

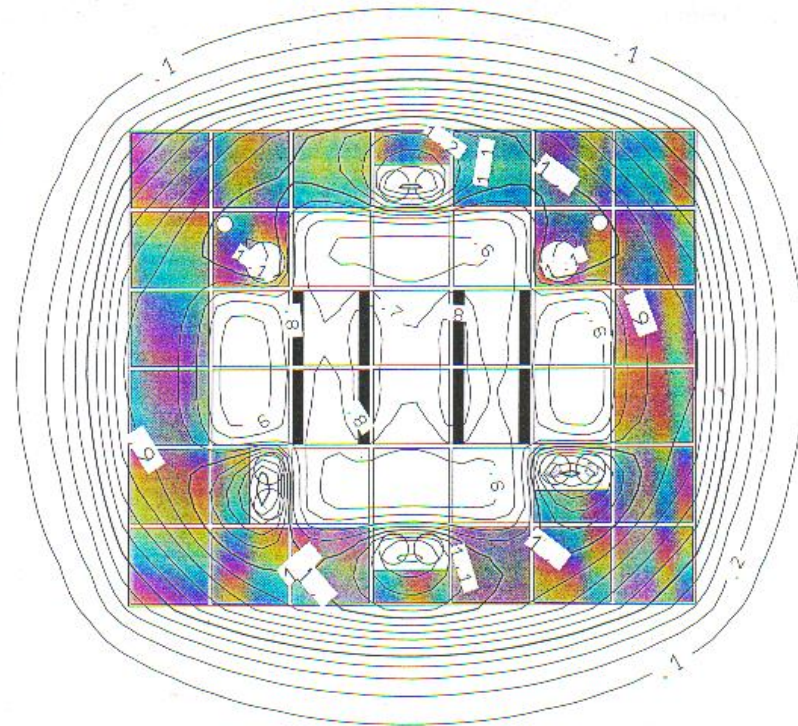
Economy of Mo Production : Irradiation : 30 % of production cost

Reactor Investment : pay back ± 6 year
 IRR = 6 % after 9 years
 IRR = 12 % after 12 years



SIEMENS

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[10¹⁴ n/cm²/s]

MING Reactor Core

Equilibrium Core

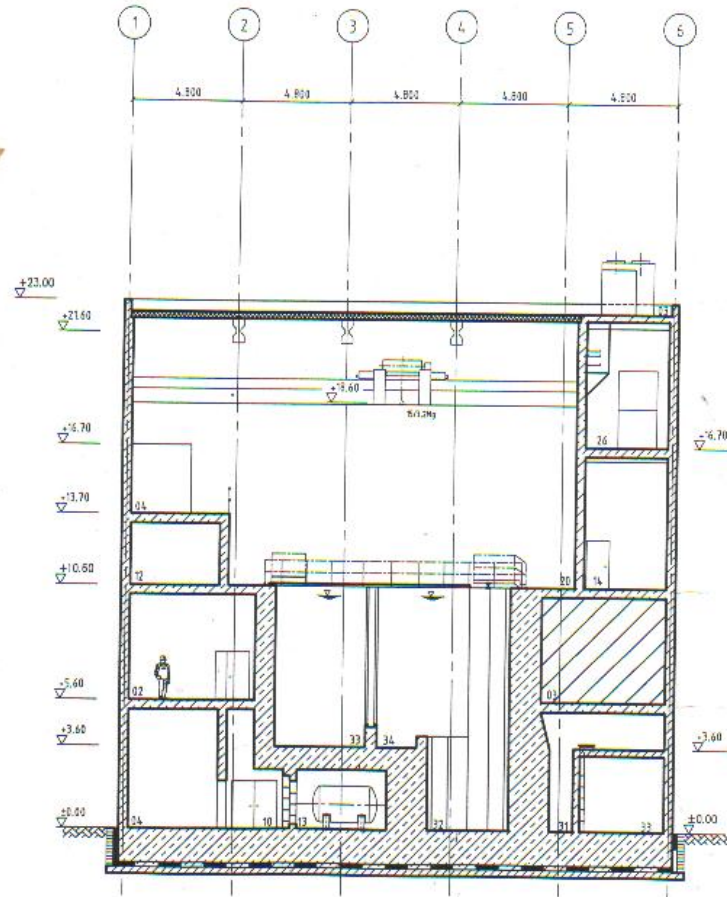
Neutron Flux Mapping for Core and Reflector

Thermal Neutron Flux ($E \leq 0.625$ eV), axially averaged



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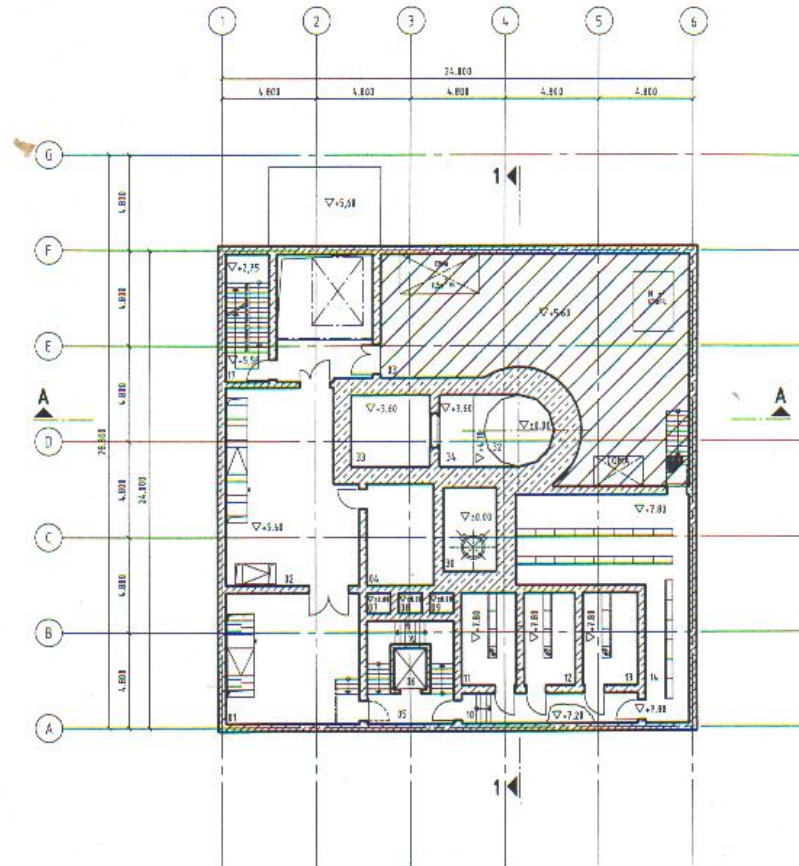


MING Building
Vertical Section A-A



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MING Building
Floor Level +5.60 m / +7.20 m

