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European Nuclear Society
Rue Belliard 65
1040 Brussels, Belgium
Phone + 32 2 505 30 54
Fax +32 2 502 39 02
E-mail ens@euronuclear.org
Internet www.euronuclear.org

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# Session V

## Utilisation

# ESTABLISHMENT OF A SINGLE-CRYSTAL FILTERED THERMAL NEUTRON BEAM FACILITY FOR NEUTRON CAPTURE THERAPY AT THE UNIVERSITY OF MISSOURI

John Brockman and Leslie P. Foyto
*University of Missouri-Columbia Research Reactor*
*1513 Research Park Drive, Columbia, Missouri, 65211 – USA*

David W. Nigg
*Idaho National Laboratory*
*P.O Box 1625, Idaho Falls, Idaho, 83415 – USA*

## ABSTRACT

The University of Missouri Research Reactor (MURR), the highest-powered (10 $MW_{th}$) University-operated research reactor in the U.S., is a pressurized, reflected, open-pool-type, light-water moderated and cooled, heterogeneous system that provides a broad range of analytical, radiographic, and irradiation services to the research community and the commercial sector.

The International Institute of Nano and Molecular Medicine (IINMM) at the University of Missouri (MU) has begun a new research effort to increase the catalogue of boron delivery agents for boron neutron capture therapy (BNCT). In support of this effort the MURR and Idaho National Laboratory (INL) have designed and constructed a new thermal neutron beam facility for small animal BNCT. The thermal neutron beam facility is designed around a 15.24 cm (6 in) diameter beam tube that extends radially from the beryllium reflector. The design uses single-crystal silicon and bismuth sections to produce a thermal neutron beam while minimizing the dose from reactor gamma rays. Initial parameter studies and design calculations were preformed using the computer codes MCNP5 and DORT.

The calculated and measured thermal neutron flux produced at the irradiation location is 9.8 x $10^8$ n/$cm^2$-s, with a measured cadmium ratio (Au foils) of 136.8, indicating a well-thermalized neutron spectrum with sufficient thermal neutron flux for a variety of small animal BNCT studies. The calculated combined epithermal and fast neutron kerma of the beam is approximately 1.0 x $10^{-11}$ cGy-$cm^2$, and the calculated incident gamma kerma is approximately 4.0 x $10^{-11}$ cGy-$cm^2$.

## I. Introduction

The International Institute for Nano and Molecular Medicine (IINMM) at the University of Missouri (MU), Idaho National Laboratory (INL), and the University of Missouri Research Reactor (MURR) are collaborating in a new research initiative to further the development of improved boron neutron capture therapy (BNCT) agents and treatment protocols for a broader array of tumor types.

Modern (post-1994) BNCT clinical trials differ from earlier trials largely because of the availability of significantly improved, near-optimal, neutron sources and the implementation of much more accurate computational and experimental dosimetry, including the required analytical chemistry. In contrast, there have been essentially no improvements in the boron containing targeting agents approved for human applications in the past 30 years. The currently available, approved BNCT compounds, while offering some attractive features, are still not optimal for the treatment of tumors of interest [1]. A key first step in the development of new BNCT agents and treatment protocols has involved the design and construction of a thermal neutron beam irradiation facility for cell and small-animal radiobiological research at the MURR. In this paper we present the beamline design and construction with the results of pertinent design calculations as well as initial neutronic performance measurements.

## II. General MURR Facility Description and Basic Reactor Design

The MURR is a multi-disciplinary research and education facility providing a broad range of analytical and irradiation services. Scientific programs include research in archaeometry, epidemiology, health physics, human and animal nutrition, nuclear medicine, radiation effects, radioisotope studies, radiotherapy, and nuclear engineering; and research techniques including neutron activation analysis, neutron scattering, and neutron interferometry.

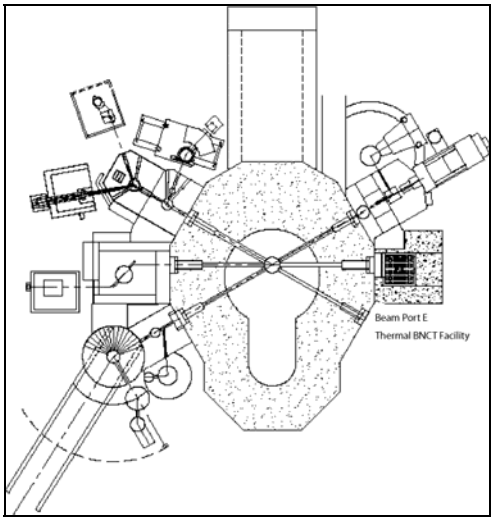


Fig 1. Biological shield and beamlines

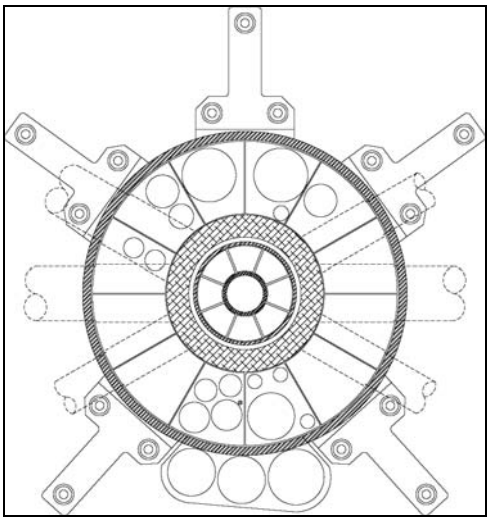

Fig 2. Reactor core assembly

The reactor (Fig. 1 and 2), which is licensed to operate at a maximum steady-state power level of 10 $MW_{th}$, is a pressurized, light-water moderated and cooled, reflected, heterogeneous, open pool-type design, which first achieved criticality on October 13, 1966. The reactor core is located eccentrically within a cylindrically-shaped, aluminum-lined pool, approximately 3.0 m (10 ft) in diameter and 9.1 m (30 ft) deep. The reactor core consists of three major regions: fuel, control blade, and reflector. The fuel region has a fixed geometry consisting of 8 fuel assemblies positioned vertically around an annulus in between two cylindrical aluminum pressure vessels. The control blade region consist of 5 control blades operating in an annular gap between the outer pressure vessel and the inner reflector annulus. The reflector consists of two concentric right circular annuluses surrounding the control blade region – the inner reflector annulus is a 6.9 cm (2.71 in) thick solid sleeve of beryllium metal, whereas the outer reflector annulus consists of vertical elements of canned graphite having a total thickness of 22.6 cm (8.89 in).

## III. Computational Methods and Modeling of the BNCT Facility

A small animal BNCT beam must provide a thermal neutron fluence of ~$10^{12}$ n/$cm^2$ at the tumor site in one hour. The beam must also contain minimal reactor gamma ray, epithermal and fast neutron contamination to minimize non-specific radiation damage. Key features of the new MURR BNCT facility which help meet these design criteria include the use of a single-crystal silicon neutron filter followed by a single-crystal bismuth filter in a manner similar to that reported by Kim et al. [2], but without cryogenic cooling of the crystals. Independent deterministic and stochastic models of the coupled reactor core and beamline were developed using the two-dimensional radiation transport code DORT [3] and the Monte Carlo transport code MCNP5 [4], respectively. The BUGLE-80 47-neutron, 20-gamma group cross section library [5] was employed for the DORT computations, in keeping with previous practice at the INL for analysis of a number of other NCT facilities worldwide. ENDF/B Version 6.8 cross section libraries were used with MCNP5, except for two specialized cross section sets for the single-crystal bismuth and silicon filters in the MCNP5 calculations that

were provided to MU and INL for this study by the Korean Atomic Energy Research Institute (KAERI) [6].

A comprehensive set of scoping calculations were conducted using DORT and, independently, with MCNP5, varying the thicknesses of the silicon and bismuth filter sections to find an optimum that maximized the thermal neutron flux while maintaining the fast neutron and gamma components of the beam within acceptable ranges. Both the DORT and the MCNP beamline optimization computations led to the conclusion that the silicon filtering section should be 50-55 cm (19.7-21.7 in) in thickness along the beamline, while the bismuth section should be 8-10 cm (3.2-3.9 in) in thickness.

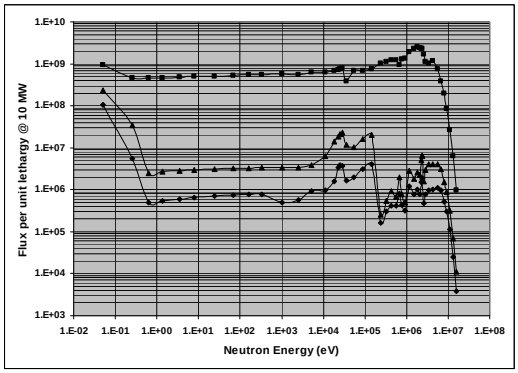


Fig 3. Computed neutron spectra

Neutron spectra at the irradiation location, computed using the DORT model, are shown in Figure 3 for the unfiltered beamline (■), for the beamline with 50 cm (19.7 in) of silicon only (▲), and for the fully-filtered configuration of 50 cm (19.7 in) of silicon and 8 cm (3.2 cm) of bismuth (♦). The spectral shapes computed by MCNP5 were consistent with the DORT results shown, within the MCNP5 statistical uncertainties. Modification of the spectrum to reduce the above-thermal component relative to the thermal component from one case to the next is apparent in the spectrum plots. The total calculated thermal neutron flux (0 – 0.414 eV) delivered to the irradiation location by the fully-filtered beam with reactor power at 10 MW was approximately 9.6 x $10^{8}$ n/cm$^{2}$-s with an estimated uncertainty of approximately 10%. The DORT calculations yielded a combined epithermal and fast-neutron kerma for the beam of approximately 1.2 x $10^{-11}$ cGy-cm$^{2}$, and an incident gamma kerma of approximately 4.0 x $10^{-11}$ cGy-cm$^{2}$.

The anticipated neutron and gamma ray radiation fields in the irradiation position were initially estimated using the MCNP5 and DORT scoping calculations. The gamma dose and fast neutron dose were calculated using an F4 tally coupled with the 1977 ANSI/ANS and ICRP-21 photon and neutron flux-to-dose conversion factors provided in the MCNP5 manual. The gamma and fast neutron doses were calculated to be 2.6 Gy/hr and 2.0 Gy/hr, respectively.

## IV. Design and Construction of the BNCT Facility

The BNCT facility has been constructed to meet the following three design criteria: (1) the thermal neutron beam must support the needs of small animal BNCT experiments, (2) the facility must provide adequate shielding for safe and routine operation, and (3) there must be systems in place that prevent the facility from being operated in a manner that is inconsistent with as low as is reasonably achievable (ALARA) radiation protection practices.

The BNCT facility consists of six main integrated components: the neutron and gamma ray filter system; the two beam shutters; the neutron and gamma ray detection system; the sample loading system; the gross shielding; and the control system. The peripheral gross shielding, silicon neutron filter, and neutron detectors are static. The beam shutters (vestibule and bismuth boxes), the sample loading system, and the shield lid are operated by the experimenter using a control panel stationed near the BNCT facility. The control panel interfaces to a programmable logic control (PLC) board that, during routine operation of the facility, restricts the user to the following three configurations: “beam on,” “beam safe,” and “sample load.” These six major components of the BNCT facility are described in the following paragraphs.

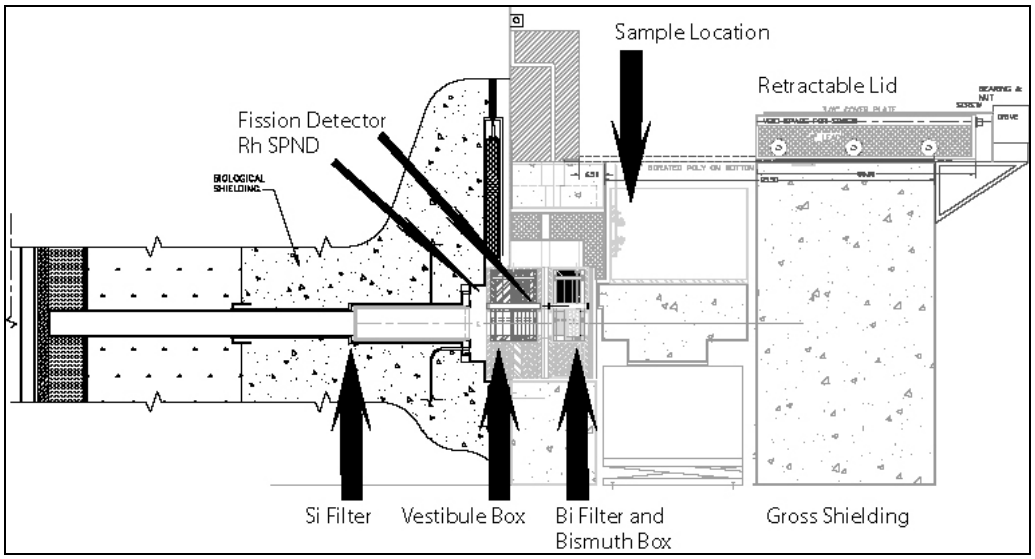


Fig 3. Detailed view of MURR BNCT facility, shown in the "sample load" configuration

(1) The neutron and gamma ray filter system**:** This system consists of a 50 cm (19.7") length of single-crystal silicon and an 8 cm (3.2") length of single-crystal bismuth. The silicon crystal is housed within the beamport collimator liner which is installed in the 20.32 cm (8") cutout section of Beamport 'E.' The bismuth crystal is housed in a rotating drum within the bismuth box outside of the biological shield, adjacent to the vestibule box.

(2) The beam shutters: The beamline shutters are positioned within the vestibule box and the bismuth box. The vestibule and bismuth boxes each consist of an automated rotating drum assembly that allows the researchers to turn the beam on and off. The vestibule box drum consists of, starting on the reactor side, a 2.54 cm (1.0 in) steel plate, a 0.635 cm (0.25 in) boral plate, a 11.43 cm (4.5 in) Pb plate, a 1.27 cm (0.5 in) steel sprocket, 7.62 cm (3.0 in) of polyethylene, a 2.54 cm (1.0 in) Pb plate and a 2.54 cm (1.0 in) steel plate. The drum rotates into an open position for "beam on" and a closed position for "sample load" and "beam safe." Rotation of each drum is controlled by a fiber optic limit switch.

(3) The neutron and gamma ray detection system: The detection system consists of four rhodium self-powered neutron detectors (Rh-SPND), two fission chambers and in some experiments a set of FarWest™ paired ion chambers. The Rh-SPND detectors are mounted on the reactor side of the vestibule box. The fission chambers are located inside the detector housing between the vestibule box and the bismuth crystal. The paired ion chambers, when used, are mounted in the irradiation chamber and are used to measure the neutron and photon dose at the sample position.

(4) The sample loading system**:** The sample loading system is designed to reproducibly place targets in the irradiation position and minimize the gamma and neutron radiation fields that researchers are exposed to in the "sample load" position. The irradiation chamber is 0.9 m (3 ft) wide and extends 0.9 m (3 ft) from the bismuth crystal housing. The loading system is designed around a TL LS4-24 hydraulic lift table manufactured by Southworth. The lift table is used to raise and lower a beam stop that incorporates approximately 910 kg (2000 lbs) of heavy concrete into the beam path during the "beam safe" and "sample load" positions. In the "beam on" position the lift table collapses to the floor, placing the sample position reproducibly into the beam.

(5) The gross shielding: The gross shielding design was largely based on prior facility experience and MCNP5 scoping calculations of neutron and gamma ray radiation fields. The large, periphery shield blocks are constructed of a steel frame, re-enforced with rebar and filled with a seven bag concrete mix provided by the MU campus. Whenever structurally possible the shield blocks that face the irradiation chamber were constructed of bare

concrete covered with sheets of 1.27 cm (0.5 in) thick borated flex panel (9% boron by weight) or 0.3175 cm (0.125 in) thick flex boron shielding (25% boron by weight). The borated flex panel and flex boron shield have thermal neutron attenuation factors of 400 and 273, respectively. The borated flex panel and flex shielding materials were used to reduce neutron scatter within the chamber, minimize neutron activation of the chamber walls, and reduce the production of high energy prompt gamma rays from hydrogen.

(6) The control system: The control system is designed to operate the beam in a safe and reproducible manner using mechanical and software interlocks. Under normal operation the BNCT facility has three available configurations which are outlined in Table 1.

| Facility Configuration | Vestibule Shutter | Bismuth Shutter | Sample Lift | Retractable Lid |
|---|---|---|---|---|
| "Beam Safe" | Closed | Closed | Up | Closed |
| "Beam On" | Open | Open | Down | Closed |
| "Sample Load" | Closed | Closed | Up | Open |

Table 1. Configuration in the "beam safe," "beam on," and "sample load" positions

## V. Gamma and Neutron Dose and Flux Measurements

Initial flux measurements were performed using foils, wires and dosimeters in the following four beamline configurations: (1) with an open, unfiltered, beam, (2) with only the bismuth filter, (3) with only the silicon filter, and (4) with both the silicon and bismuth filters. This allowed an independent evaluation of the performance of each of the two filter components separately, and in combination. The results are listed in the first four columns of Table 2. Final beamline measurements were just recently completed after final configuration of the BNCT facility. Those valves are listed in the final column of Table 2. The irradiation times varied from 5 minutes in the unfiltered beam configuration to a maximum of 10 minutes in the filtered beams.

| | **Unfiltered Beam** | **8 cm Bi** | **50 cm Si** | **8 cm Bi + 50 cm Si** | **Final Beam** |
|---|---|---|---|---|---|
| Saturation Activity, Bare Au Foil (Bq/atom) | $1.31 \times 10^{-12}$ (5%) | $3.82 \times 10^{-13}$ (5%) | $2.38 \times 10^{-13}$ (5%) | $8.67 \times 10^{-14}$ (5%) | $8.63 \times 10^{-14}$ (5%) |
| Saturation Activity, Cd covered Au Foil (decays/atom-s) | $4.11 \times 10^{-13}$ (5%) | $7.49 \times 10^{-14}$ (5%) | $3.64 \times 10^{-15}$ (5%) | $8.21 \times 10^{-16}$ (5%) | $6.31 \times 10^{-16}$ (5%) |
| Diff. in Saturation Activity, Bare-Cd (decays/atom-s) | $8.95 \times 10^{-13}$ (8%) | $3.07 \times 10^{-13}$ (5%) | $2.34 \times 10^{-13}$ (5%) | $8.59 \times 10^{-14}$ (5%) | $8.57 \times 10^{-14}$ (5%) |
| Measured Thermal Flux ($n/cm^2$-s) | $9.80 \times 10^{9}$ (11%) | $3.36 \times 10^{9}$ (8%) | $2.56 \times 10^{9}$ (8%) | $9.40 \times 10^{8}$ (8%) | $9.80 \times 10^{8}$ (8%) |
| Calculated Thermal Flux from DORT ($n/cm^2$-s) | $9.38 \times 10^{9}$ (10%) | $3.81 \times 10^{9}$ (10%) | $2.22 \times 10^{9}$ (10%) | $9.62 \times 10^{8}$ (10%) | $9.62 \times 10^{8}$ (10%) |
| Cd Ratio (Bare/Cd) | 3.18 (7%) | 5.10 (7%) | 65.3 (7%) | 105.5 (7%) | 136.8 (7%) |
| Wire saturation activity ratio (Au/Cu) | 36.4 | 28.4 | 22.4 | 22.4 | 22.1 |

Table 2. Performance results of the MURR BNCT facility

Gold foils with and without cadmium covers as well as with flux wires composed of natural copper alloyed with 1.55% gold by weight were used. The gold foils were nominally 0.0254 mm (0.001 in) in thickness and 12.7 mm (0.5 in) in diameter, with masses of approximately 60 mg. The flux wires were 1 mm (0.039 in) in diameter and approximately 10 mm (0.39 in) in length, each with a mass of approximately 70 mg. Some scoping measurements to

estimate the gamma dose rate at the irradiation location were also performed using Landauer TLD-100 dosimeters.

The incident gamma and neutron components were also measured using a set of FarWest$^{TM}$ paired ion chambers, in keeping with the recommendations of the International BNCT Dosimetry Exchange [7]. The gamma dose was measured at 2.26 cGy/min whereas the neutron dose was measured at 1.72 cGy/min.

Final gamma and neutron dose rate measurements surrounding the facility were also performed while the BNCT facility was in various configurations at 10 MW operation. In the "sample load" configuration, a field of 0.25 mSv/hr gamma and 0.25 mSv/hr neutron exists inside the irradiation chamber. In "beam on," there is a field of 0.45 mSv/hr neutron on the lid, a field of 0.50 mSv/hr gamma on the east shielding blocks, a field of 0.25 mSV/hr on the beam axis on the north shield block, and a field of 0.25 mSv/hr on the west side.

## VI. Conclusions and Future Work

Parameter studies, design calculations and initial performance measurements have been completed for the new BNCT facility. These results indicate that a more than adequate thermal neutron beamline facility has been designed and constructed for BNCT cell and small-animal radiobiology studies at the MURR. Near future work will include small animal phantom dosimetery measurements using flux wires and the FarWest$^{TM}$ paired ion chambers. Animal phantoms, containing flux wires, will be placed in the beam and the neutron dose will be calculated by taking the ratio of the flux wire activity measured on the paired ion chambers with the flux wire activity measured in the phantom. Kerma factors will be applied to calculate the primary biological components of the non-specific neutron dose (hydrogen knock on damage n(H,H')n', n($^{14}$N,p)$^{14}$C and hydrogen prompt gamma production) and the dose from the reaction with $^{10}$B.

## VII. References


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# USE OF FISSION RADIATION IN LIFE SCIENCES AND MATERIALS CHARACTERISATION

F.M. WAGNER, B. LOEPER-KABASAKAL, H. BREITKREUTZ, W. PETRY
*Forschungsneutronenquelle Heinz Maier-Leibnitz (FRM II), Technische Universität München*
*85747 Garching – Germany*

T. BÜCHERL
*Institut für Radiochemie, Technische Universität München*
*85747 Garching – Germany*

## ABSTRACT

At FRM II, a pair of uranium plates can be driven in front of beam tube SR10 where they become the source of an unmoderated fission neutron beam. Without converter plates, the thermal flux can be used. For the application of fast neutrons, the accompanying gamma radiation is normally reduced by a set of lead filters. The related standard neutron beam has essentially a Watt spectrum with mean energy 1.9 MeV. Clinical and biological applications use the high linear energy transfer of fission neutrons. Human tumours suitable for neutron treatment are situated near to the surface, e.g., head and neck tumours, lymph node or skin metastases from various cancer diseases, and chest wall metastases of breast cancer. Fast neutrons are also used for radiography and tomography of specimen consisting of metal and/or organic substances. Especially hydrogen generates a good contrast. Further applications deal with the radiation hardness of electronic components.

## 1. Introduction

Within the moderator tank of FRM II, a pair of highly enriched uranium plates (499 g $^{235}U$) can be moved in front of the beam tube SR10 (Fig. 1,2).

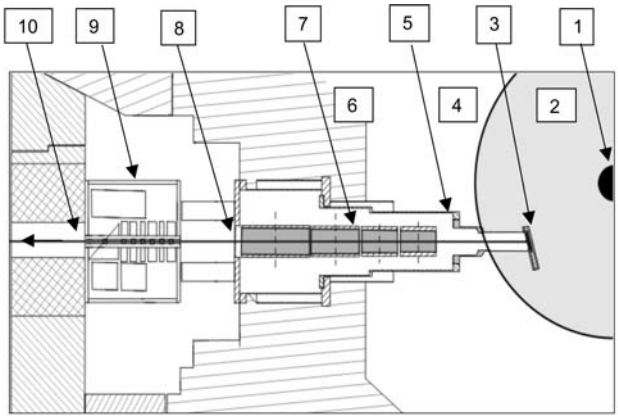


Fig 1: Horizontal section of the irradiation facility: 1 Compact reactor core; 2 $D_2O$ moderator tank; 3 Vertically movable converter plates in a shaft; 4 Light water zone; 5 He-filled beam tube SR10; 6 Biological shield; 7 Four revolving shutters; 8 Position for irradiations with thermal neutrons; 9 Filter bench; 10 Multi leaf collimator

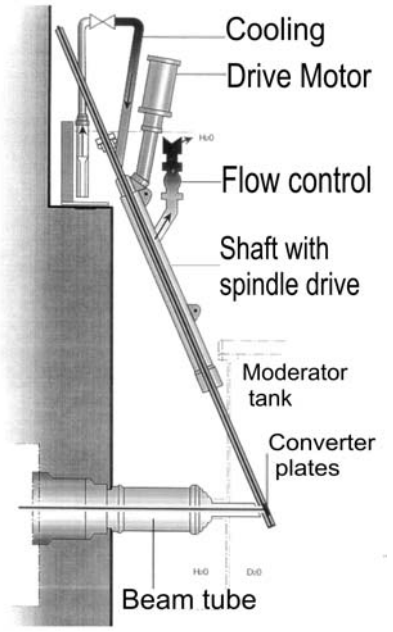


Fig 2: Vertical section of the reactor pool with converter facility

The high flux of thermal neutrons from the core induces fission processes and generates fast neutrons (conversion). These fission neutrons with mean energy 2 MeV can escape through the adjacent beam tube without moderation. At the exit of the beam tube, the beam quality is determined by three components: fission neutrons, gamma radiation, and thermalized neutrons. Their respective contributions to the beam can be varied.

## 2. Available beam qualities

### 2.1 Thermal neutron beam

When the converter plates are withdrawn from the moderator tank, the beam consists only of thermalized neutrons from the reactor core and the accompanying gamma radiation. The flux at point 8 in Fig. 2 is $3{,}9 \times 10^{9}$ $s^{-1}cm^{-2}$ with a beam area of 23x18 cm². The cadmium ratio for gold foils is greater than 400.

### 2.2 Fast neutron beam

In order to use fast neutrons, the converter plates are positioned in closest touch with the entrance of the beam tube (Fig. 2, point 3). With a converter power of 80 kW, the fast neutron flux at the reference point at 5.9 m distance from the source is $7.0 \times 10^{8}$ $s^{-1}cm^{-2}$. Thermal neutrons originating from the moderator tank are suppressed at the exit of the beam tube by a filter consisting of 1 cm $B_4C$ (50%) in epoxy. The operation of the uranium converter generates also an intense flux of high energy photons which is generally unwanted. Therefore, the beam is additionally filtered by 3.5 cm lead what reduces the gamma dose more effectively than the neutron dose. This filtering generates the standard beam used for medical applications with a fast flux of $3.2 \times 10^{8}$ $s^{-1}cm^{-2}$; further details about the facility are given in [1].

The standard neutron beam has a Watt spectrum with a minor addition of epithermal neutrons; the mean energy is 1.9 MeV [2, 3]. The fraction of gamma to neutron flux is conveniently expressed in terms of energy dose rates measured in a water phantom in 2 cm depth where equilibrium of secondary electrons is reached. With a collimator opening of 9x9 cm², the neutron and gamma energy dose rates on the beam axis are 0.54 and 0.20 Gy/min, respectively; and the 50% neutron isodose is at 5 cm depth. For non-medical applications, the fraction of photon flux to fast neutron flux can further be reduced. With, e.g., 6 cm lead in the beam, the remaining fast neutron flux is $1.9 \times 10^{8}$ $s^{-1}cm^{-2}$ at the reference point; for radiography and tomography (see section 3.3.2), 11 cm lead are inserted.

## 3. Applications

### 3.1 Trace element distribution (thermal beam)

In connection with worldwide efforts to develop suitable compounds for boron neutron capture therapy, many boron compounds are investigated as to their specific enrichment and sufficient retention period in selected tumours [4]. For this sake, whole body cryosections of small animals are fixed on etch track films and irradiated. In this way, e.g., the distribution of $^{10}B$ in the tumour model of a mouse can be visualized and evaluated quantitatively. By etching of the film a picture as that of Fig. 3 is obtained. Due to the low contamination by fast neutrons, the background due to recoil protons is very low; a minimum detectable fraction of boron (element) of 0.1 ppm can be reached when counting single tracks.

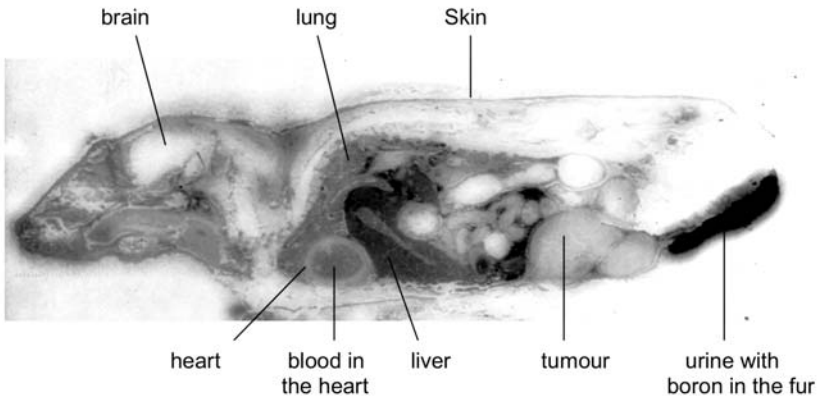


Fig. 3. Scan of an etch track film which was irradiated together with a whole-body section of a mouse. The darker the area the higher the concentration of boron. The tumour contains about 5 ppm $^{10}B$ (courtesy of D. Gabel, University of Bremen).

## 3.2 Medical and biological use (standard fast neutron beam)

Biological and clinical applications are principally motivated by the high linear energy transfer (LET) up to 100 keV/µm of fission neutrons. The high LET leads to biological radiation effects different from photons of the same energy, e.g., a much weaker dependence of the cell deactivation on the oxygenation. The quick energy loss, however, goes hand in hand with a low penetration depth, therefore fission neutron therapy is primarily suited for tumours situated near to the body surface.

There is already an expertise from 715 patients with tumours who were treated at the fission beam of the former research reactor FRM [5-7] until July 2000. The best suited tumours for neutron treatment are radiation resistant adenoid cystic carcinoma of the salivary glands, head and neck tumours, malign melanoma, lymph node metastases or skin metastases from various cancer diseases, and chest wall metastases of breast cancer. In the latter case, the steep depth dose curve is an advantage because the radiation dose to the lungs and the heart is generally not prohibitive even with vertical incidence. At FRM II since June 2007, 36 patients have undergone 209 irradiations. 40 % of the patients have been treated with (locally) curative intent, the others for palliation.

In parallel to the clinical applications, the dose-response relationship of dicentric chromosomes in human lymphocytes for fission neutrons has been investigated at different depths in a polyethylene phantom [8]. The dose-response curves for dicentrics suggest a significantly lower biological relative effectiveness RBE (compared to $^{60}Co$ γ-rays) with increasing depth (Fig. 4).

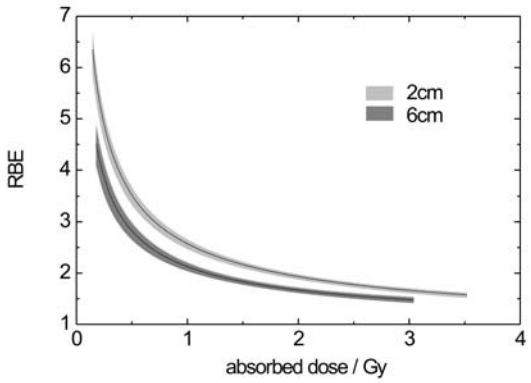


Fig 4: RBE of the MEDAPP beam vs. dose at depths of 2 and 6 cm, resp., in a PE-phantom.

## 3.3 Materials Characterisation

### 3.3.1 Radiation hardness of electronic components (fast neutron beam with additional filters)

CMOS sensors, also being referred to as Monolithic Active Pixel Sensors (MAPS), demonstrated their ability to serve as sensors for minimum ionising particles (MIPs). They represent a novel technology for charged particle tracking in high-energy particle and nuclear physics. Small pixel sizes of down to 10x10 $\mu m^2$ allow unprecedented spatial resolutions and a low material budget at the same time. The Technology Laboratory of the University Frankfurt, Institute of Nuclear Physics, participates in the R&D, design and construction of the vertex detector of the Compressed Baryonic Matter (CBM) experiment (FAIR at GSI Darmstadt, Germany). An essential performance criterion in such applications is the radiation tolerance of the pixel chip. The detectors are foreseen to operate in close vicinity to the primary interaction vertex and exposed to the maximum track density. Moreover, the experiments are optimized to measure rare probes such as open charm and, hence, cope with very high integral particle fluences.

At FRM II, irradiations were performed with equivalent fluences up to $10^{13}$ cm² (1 MeV equivalent neutron fluence for silicon according to ASTM standard E 722-04). They proved that the latest MAPS generation (MIMOSA-18) may stand more than $10^{13}$ $n_{eq}/cm^2$ representing a progress of one order of magnitude with respect to previous designs [9].

### 3.3.2 Neutron computerised tomography and radiography (NECTAR)

Radiography and tomography using collimated fission neutrons on objects from different fields of applications (e.g., technical objects like turbine blades, archaeological and art historical objects, wooden samples as Fig. 5 left) have become a standard application at the NECTAR facility located at the rear end of the SR10 beam [10].

Recently, preliminary measurements for investigation of real-time applications were performed successfully using a lead filtered beam with no additional collimator unit [11]. The abdication of the collimator results in a decreased spatial resolution (Fig. 5 right) by a factor of about two, but an increased fission neutron flux by a factor of about 8.

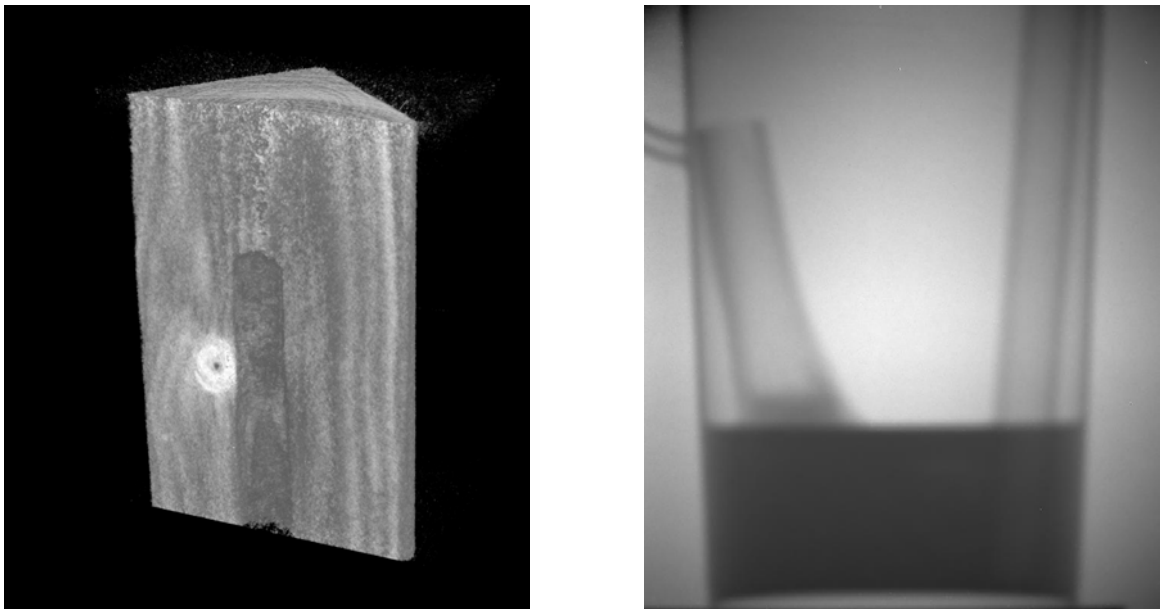

Fig 5: Left: Cut through a 3D-CT of a timber using a collimated neutron beam.
Right: Radiography of a test sample consisting of a plastic can containing a thighbone and a polyethylene rod, both covered by water in the lower part, using no collimator.

## Summary

The unique fission neutron facilities MEDAPP and NECTAR, both located at the beam tube SR10 at FRM II, offer a wide range of applications in different fields. These are permanently extended by intensive use and continuous improvements of the facilities.

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# RADIOISOTOPE PRODUCTION FOR NUCLEAR MEDICINE USING THE IEA-R1m REACTOR AT IPEN-CNEN/SP-BRAZIL

J. A. OSSO JR., K.N. SUZIKI , R.F. COSTA, G. BARRIO, T.P. BRAMBILLA, P. C. LOPES, S.E.T. BORBOREMA, C.R.B.R. DIAS, A.L.P.V. LIMA, H.T. GASIGLIA, J.L. CORREIA, L.A. VILLELA, N.C. SILVA, R.R.L. BEZERRA, R.F. LIMA, M.R.F.F. BARBOZA, J. MENGATTI

*Radiopharmacy Center, Instituto de Pesquisas Energéticas e Nucleares, IPEN-CNEN/SP*
*Av. Professor Lineu Prestes 2242 - 05508-000 - São Paulo SP – Brazil,*
*jaosso@ipen.br,jaossoj@yahoo.com.br*

## ABSTRACT

The Radiopharmacy Center of IPEN/Brazil has an established radioisotope production program to supply radiopharmaceuticals for use in Nuclear Medicine. IPEN has a Research Reactor that nowadays operates at 3.5MW for 64 hours continuously. This paper describes the story of the radioisotope production using this very reliable Reactor, reporting the radioisotopes produced for routine and also for research in the radiopharmacy field. Nowadays it is possible to produce all the demand of $^{153}$Sm, 50% of the demand of $^{131}$I and viability studies were performed towards the production of $^{166}$Ho, $^{177}$Lu and $^{188}$W. A $^{99m}$Tc gel generator was developed using the $^{98}$Mo(n,$\gamma$)$^{99}$Mo reaction. Several tracers used in radiopharmacy development research were produced, such as $^{122}$Sb, $^{124}$Sb, $^{85}$Sr and $^{88}$Y. A decision of building a Radioisotope Production Reactor in Brazil is about to be made in order to nationalize the production and avoid the distribution problems that affect the big world producers nowadays.

## 1. Introduction

Nuclear medicine is the field of Medicine that involves the use of small amounts of radioactive materials, the so-called radiopharmaceuticals to help diagnose and/or treat a variety of diseases. Nuclear medicine determines the cause of the medical problem based on the function of the organ, tissue or bone. This is how nuclear medicine differs from an x-ray, ultrasound or any other diagnostic test that determines the presence of disease based on structural appearance.

The radiopharmaceuticals are prepared through the labelling of a specific substrate with an adequate radioisotope. The substrate can be a biomolecule, a complex or a simple compound with a specific biodistribution in the body and the radioisotope is chosen by its physical decay properties. For diagnose purposes the radioisotope must decay emitting a single photon (SPECT technique) or positrons (PET technique). For therapy the radioisotope must emit a particle, most commonly a beta particle. The radioisotope is produced by nuclear reactions that occur in a Nuclear Reactor or in a particle accelerator such as the Cyclotron.

The Radiopharmacy Center of IPEN-CNEN/SP-Brazil has an established radioisotope production program to supply radiopharmaceuticals to the Nuclear Medicine community in Brazil [1]. These radiopharmaceuticals are prepared with radioisotopes produced in both a Nuclear Reactor and a Cyclotron accelerator. IPEN has a Research Reactor, so called IEA-R1m, that nowadays operates at 3.5MW for 64 hours continuously. This paper describes the story of the radioisotope production using this very reliable Reactor, reporting the radioisotopes produced for routine production and also for research in the radiopharmacy field.

## 2. Methods

### 2.1 Reactor produced radioisotopes at IPEN – brief story

The Reactor produced radioisotope program started at IPEN in 1959 with the production of $^{131}I$ and since then all the efforts were made towards the development of methods for the production of the desirable radioisotopes. Since 1981 IPEN produces and distributes $^{99m}Tc$ generators with imported fission-produced $^{99}Mo$ and in-house developed technology of chromatographic alumina based generators. In 1996 IPEN started a project aiming the nationalization of the production of $^{99}Mo$ through the $^{98}Mo(n,\gamma)^{99}Mo$ reaction and developing the MoZr gel generator technology. The technology was developed but the Reactor could not reach the power required for the production, 5 MW. However, this project made it possible to upgrade the Reactor power to 3.5 MW and nowadays it is possible to produce at least 50% of the Brazilian demand of $^{131}I$. Since 1995 IPEN produces all $^{153}Sm$ required in Brazil, and viability studies were performed towards the production of $^{166}Ho$, $^{177}Lu$ and the $^{188}W$-$^{188}Re$ generator. Several tracers were produced in order to help the development of radiopharmaceutical production methods and among them it can be reported the use of $^{122}Sb$, $^{124}Sb$, $^{88}Y$ and $^{85}Sr$.

### 2.2 Irradiations

All irradiations for the routine production and for the research were performed at the IEA-R1m reactor at IPEN-CNEN/SP, a pool type reactor using LEU as fuel material, with power varying from 2 to 5 MW. All the targets were irradiated inside aluminium holders either directly or inside quartz tubes. A Beryllium irradiator was specially fabricated for high power irradiations of $MoO_3$ and $TeO_2$ targets.

### 2.3 Routine Production

Nowadays two radioisotopes are routinely produced using IEA-R1m Reactor: $^{153}Sm$ and $^{131}I$.
$^{153}Sm$, a $\beta^-$ emitter with a half-life of 46 hours is produced through the reaction $^{152}Sm(n,\gamma)^{153}Sm$, irradiating a target of $Sm(NO_3)_2$ enriched in $^{152}Sm$. The target is encapsulated inside a quartz tube and irradiated for 24-48 hous, depending on the demand. Two main radiopharmaceuticlas are further prepared with $^{153}Sm$: $^{153}Sm$-EDTMP used bone pain palliation and $^{153}Sm$-hidroxiapatite, used for radiosinoviortesis.
$^{131}I$, a $\beta^-$ emitter with a half-life of 8 days is produced through the indirect reaction $^{130}Te(n,\gamma)^{131}Te \rightarrow ^{131}I$ irradiating pressed $TeO_2$ targets inside an aluminium holder. The targets are irradiated at least for 2 cycles of 64 hours inside the Be irradiator. The separation between the irradiated target and $^{131}I$ is performed by the dry distillation technique. This radioisotope can be used to prepare radiopharmaceuticals for both diagnose and therapy uses. The radiopharmaceuticals are: $^{131}I$-Iodide, $^{131}I$-MIBG, $^{131}I$-Lipiodol and $^{131}I$-Hippuran.

### 2.4 Viability Studies

Viability studies have been performed for the production of 3 important radioisotopes for Nuclear Medicine: $^{166}Ho$, $^{177}Lu$, $^{188}W$.
$^{166}Ho$ is a $\beta^-$ emitter with a half-life of 26.8 hours and is produced through the reaction $^{165}Ho$ $(n,\gamma)$ $^{166}Ho$. Targets of $Ho_2O_3$ were irradiated inside quartz tubes in selected positions of the Reactor. After the irradiation the samples were dissolved and the activity produced evaluated by $\gamma$ ray spectroscopy. The main use of $^{166}Ho$ is the labelling of ion exchange resins as potential agents for liver cancer therapy [2].
$^{177}Lu$ is a $\beta^-$ emitter with a half-life of 6.7 days and is produced through the reaction $^{176}Lu$ $(n,\gamma)$ $^{177}Lu$. Targets of $Lu_2O_3$ were irradiated inside quartz tubes in selected positions of the Reactor. After the irradiation the samples were dissolved and the activity produced evaluated by $\gamma$ ray spectroscopy. One $^{177}Lu$ based radiopharmaceutical is currently produced at IPEN,

$^{177}$Lu-octreotate, used for therapy of neuroendocrine tumors, and prepared with imported $^{177}$Lu [3].
$^{188}$W is the parent radioisotope of $^{188}$Re, a $\beta^-$ emitter with a half-life of 16.7 hours and is produced by the reaction $^{186}$W(2n,$\gamma$)$^{188}$W. Activation studies complied of the irradiation of W targets (metallic W and $WO_3$) placed inside an aluminium holder and irradiated for a definite length of time (2 hours up to 64 hours) with low neutron fluxes ($1x10^{13}$ n.cm$^{-2}$.s$^{-1}$) and inside a Be irradiator located at the reactor core with the highest possible flux ($4x10^{13}$ n.cm$^{-2}$.s$^{-1}$). After the irradiation, qualitative and quantitative analyses were performed in the targets by $\gamma$-spectroscopy, and $^{188}$Re generators were prepared with the $^{188}$W produced [4,5].

## 2.5 $^{99}$Mo-$^{99m}$Tc Gel Generator

$^{99m}$Tc is the most used radioisotope for diagnosis exams performed in Nuclear Medicine worldwide due to its favorable physical decay properties and its distribution in the generator form of $^{99}$Mo-$^{99m}$Tc [6,7]. In 1996 IPEN started a project aiming the nationalization of the production of $^{99}$Mo through the $^{98}$Mo(n,$\gamma$)$^{99}$Mo reaction and developing the MoZr gel generator technology [8,9].
Two different targets were tested: metallic Mo and $MoO_3$ in the form of powder or molten. The targets were irradiated irradiated for a definite length of time with low neutron fluxes and inside a Be irradiator located at the reactor core with the highest possible flux. After the irradiations, the activity of $^{99}$Mo was determined in a dose calibrator and by $\gamma$-ray spectroscopy employing a hiperpure Ge detector from Canberra. $^{99}$Mo was further employed for the studies aiming the preparation of a MoZr gel type generator.

## 2.6 Tracer studies

Several tracers were produced in order to help the development of radiopharmaceutical production methods, such as $^{122}$Sb, $^{124}$Sb, $^{88}$Y and $^{85}$Sr.
Two Sb tracers, $^{122}$Sb and $^{124}$Sb, were prepared to follow the biodistribution of a pharmaceutical used in the therapy of Leishmanioses, glucantime, and its liposomal formulations, also to evaluate the amount of Sb incorporated inside the liposomes. The tracers are produced through the reactions $^{121}$Sb(n,$\gamma$)$^{122}$Sb and $^{123}$Sb(n,$\gamma$)$^{124}$Sb. Samples of glucantime and liposomes in clean polypropylene tubes were placed together with antimony standards inside the aluminium container. Irradiations were carried out at a thermal neutron flux of $0,8x10^{12}$ n.cm$^{-2}$.s$^{-1}$ for 15 minutes [10].
An important research aiming the nationalization of the production of $^{90}$Y is being carried out using radioactive tracers produced in the reactor. $^{90}$Y is a radioisotope for use in therapy, nowadays imported by IPEN at a very high cost and it is produced by a generator system, $^{90}$Sr-$^{90}$Y. The gamma emitting tracers $^{88}$Y and $^{85}$Sr are produced through the reactions $^{89}$Y(n,$\gamma$)$^{88}$Y and $^{84}$Sr(n,$\gamma$)$^{85}$Sr, respectively, and are employed to optimize the techniques studied for the development of the generator [11].
The experimental work was developed at the laboratories of the Radiopharmacy Center at IPEN-CNEN/SP.

# 3. Results

## 3.1 Routine production

The local production of $^{153}$Sm meets the total demand for the radiopharmaceuticals distributed at IPEN. About 74 GBq (2 Ci) of $^{153}$Sm can be produced in one irradiation and the total activity delivered every week is about 37 GBq (1 Ci).
$^{131}$I has a big market in Brazil, and every week more than 1480 GBq (40 Ci) of $^{131}$I radiopharmaceuticals are distributed by IPEN. Nowadays, with the Reactor operating at 3.5 MW, half of this demand is achieved by the local production (740 GBq – 20 Ci).

## 3.2 Viability studies

The results of the viability studies showed that it is possible to produce $^{166}$Ho for clinical use, with the current operating conditions of the Reactor. A specific activity of at least 1480 GBq/ g Ho (40 Ci/ g Ho) can be achieved in these conditions.
The specific activities achieved for $^{177}$Lu and $^{188}$W were not enough for the local production, even if the reactor power is upgraded to 5 MW. These studies were important for the developing of the targets, dissolution of the targets and also for the development of the methodology of producing the $^{188}$W-$^{188}$Re generators.

## 3.3 $^{99}$Mo-$^{99m}$Tc Gel Generator

A successful technology was developed for the preparation of the gel and a hot processing cell was assembled for the routine preparation of MoZr gel and gel generators. The Quality Control group validated the generators, approving the required quality and generators up to 11.1 GBq (300 mCi) could be produced with reactor power of 5 MW. Unfortunately, the Reactor could not reach the power required for the start of the clinical trials with the generator, 5 MW. Nowadays, even if the Reactor operates at this power, the Nuclear Medicine market requires generators of higher activities. Studies are under way aiming the concentration of the $^{99m}$Tc activity eluted from the gel generator.

## 3.4 Tracer studies

Enough activities of $^{122}$Sb, $^{124}$Sb, $^{88}$Y and $^{85}$Sr could be produced for the studies where these tracers are involved. Their use speeds up the researches and in particular, the use f the activation process to evaluate Sb in liposome formulations was the only reliable analytical technique, when compared with the other techniques employed, such as ICP, UV-Visible and Atomic absorption.

## 4. Conclusions

This story full of successes and the results showed in this paper, together with the growing demand of radioisotopes for Nuclear Medicine in Brazil and the new technologies developed in this field are enough reasons to reach a decision of building a Radioisotope Production Reactor in Brazil, in order to nationalize the production and avoid the distribution problems that affects the big world producers nowadays. In particular, the main concern is the production of $^{99}$Mo, that represents a demand of nearly 16,650 GBq (450 Ci) of $^{99}$Mo per week in Brazil, at calibration time.

## 5. Acknowledgement

The authors wish to thanks CNEN, IPEN, CNPq, CAPES and FAPESP for the fellowships provided for this work.

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# DESIGN OF A FLOWING-NAK EXPERIMENTAL DEVICE FOR IN-CORE MATERIAL IRRADIATION IN THE JULES HOROWITZ REACTOR

Damien MOULIN, Christine BISCARRAT, Sébastien CHRISTIN, Guy LAFFONT, Aurélien MORCILLO, Eric PLUYETTE, Frédéric REY

*CEA, DEN, Department of Nuclear Technology,*
*F-13108 Saint-Paul-Lez-Durance, France.*

## ABSTRACT

One of the characteristics of the Jules Horowitz Reactor will be its very high fast neutron flux. To take benefits of these irradiation locations, a specific test device called CALIPSO is under development. The accurate temperature control of the samples is possible by the mean of a small in-pile loop of circulating NaK. Progresses on designing the CALIPSO device have been made concerning its operating range and its main components such as the electromagnetic pump, the electrical heater and the heat exchanger. The overall dimensions of the tubes have been defined according to the safety requirements. The present design of CALIPSO provides a large operating range regarding to nuclear heating values and NaK temperature while keeping homogeneous the temperature distribution around the samples.

## 1. Introduction

The Jules Horowitz Reactor (JHR) is under construction in the CEA's Cadarache centre in the south of France. It will be mainly dedicated to study material and fuel behaviour under irradiation for the benefits of existing nuclear plants as well as the development of the future reactors (1).

One of the characteristics of the JHR is its very high fast neutron flux providing irradiation possibilities that will be quite unique among the existing Material Testing Reactors (2).

To take benefits of these irradiation locations, a specific test device was designed: initially known as "M3" in its definition phase (3), it has been named CALIPSO since the beginning of its development phase. As summarized in its acronym it is an in-Core Advanced Loop for Irradiation in Potassium Sodium (NaK) coolant.

After a short presentation of the JHR in-core experimental locations, this paper will give the operating principle, then the design of the main components will be detailed in accordance to the operating constraints and the safety requirements.

## 2. In-core experimental locations

Operating at 100 MWth the JHR core is composed of 34 fuel elements. Each fuel element is composed of 3 zones of 8 curved plates leaving a central hole. The total fissile length is 60 cm-long and the core is inserted in a 74 cm-diameter pressurized vessel.

Several experimental locations are available in the centre of fuel elements and some others by replacing fuel elements (4).

Standard in-fuel element experimental locations are characterized by very high fast neutron flux (E>1 MeV) reaching $5.10^{14}$ $n.cm^{-2}.s^{-1}$ at full power in the core mid-plane. These are very efficient to perform experiments such as dose accumulation for cladding and structural materials (displacement per atom rate ~15 dpa/year). The counter part of such high fast neutron flux, is very high gamma and fast neutrons heating in materials, reaching about 20 W/g in steel. It is also important to highlight the small available space for the device part located in the fissile zone: maximal outer diameter ~ 33 mm.

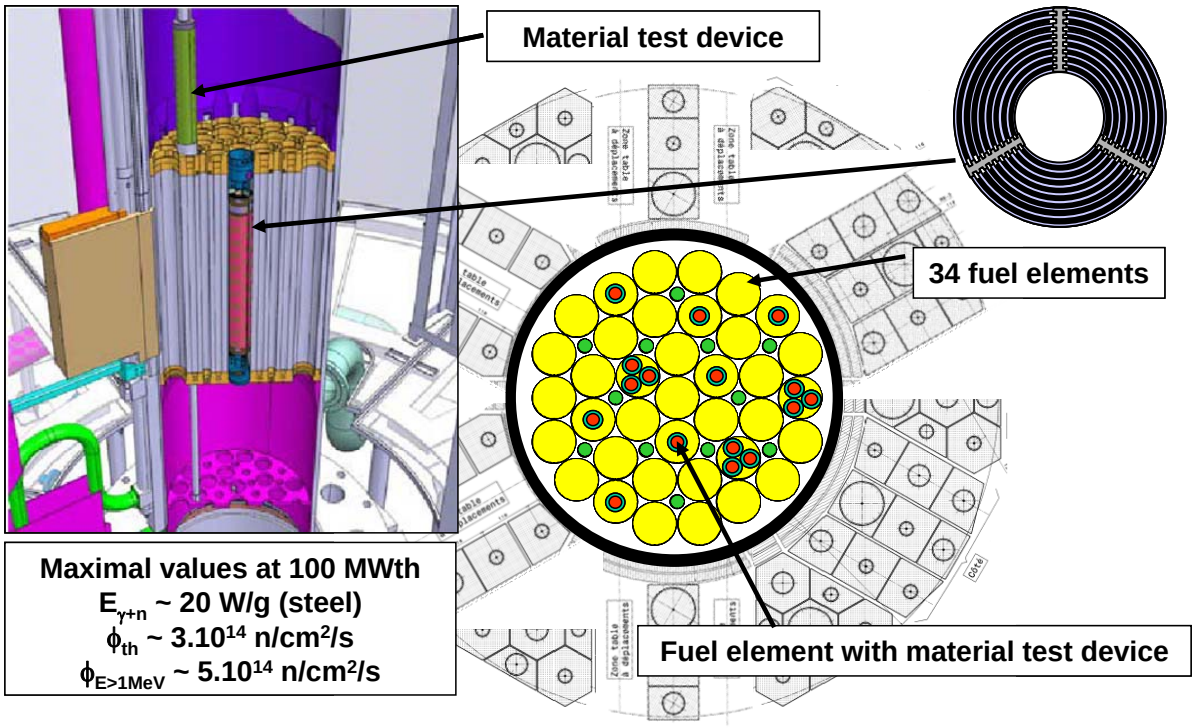


Figure 1 : View of the JHR core

## 3. Operating principle

An important requirement for experimental conditions in test devices with a lot of samples is to keep the temperature distribution homogeneous. This becomes a real challenge when dealing with high nuclear heating.

In the case of CALIPSO, the accurate temperature control of the samples is possible by the mean of a small in-pile loop of circulating NaK. Indeed, placed in the central hole of the fuel element, this device shall be autonomous for long-term irradiations and has to enclose in a confined space all the components needed to ensure a forced convection flow in the test section.

Therefore, located above the active zone, a very compact annular electromagnetic pump induces the NaK circulation. Electrical element situated just above the pump gives a part of energy deposit to the NaK. Then, the NaK flow gets the main energy deposit with nuclear heating in the active zone around the samples. Finally, the upward circulation into the heat exchanger, then the gap between the containement rig inner wall and the separator shell allows the NaK to cool down by heat exchange with the reactor primary circuit (Figure 2).

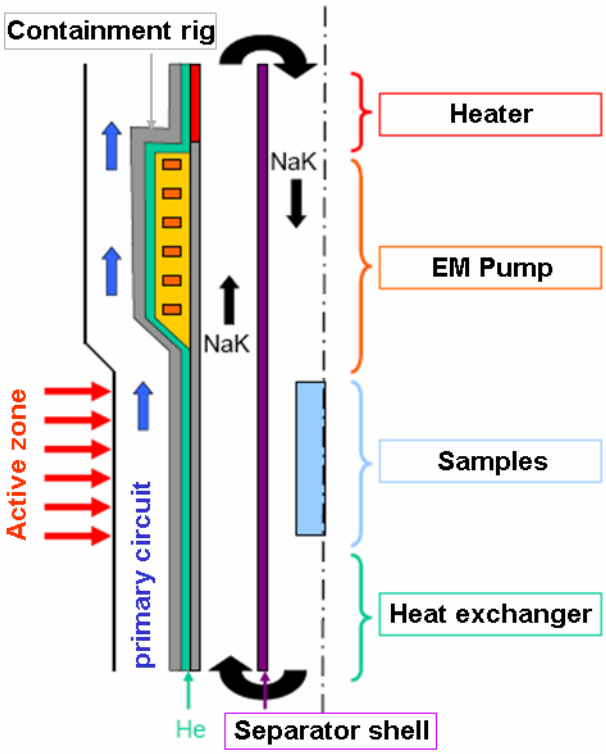


Figure 2 : Operating principle of CALIPSO

## 4. Containment rig and sample-holder

The CALIPSO test device is composed of two main parts: the containment rig and the sample-holder (Figure 3).

The containment rig is the outer shell of the test device that houses fluids. It is designed to resist to the thermal-mechanical stresses and comply with the safety requirement. It fits to the core mechanical structures and its outer shell is cooled by the reactor primary circuit.

The sample-holder is plugged into the containment rig through the circular opening situated in the upper part. It holds the material samples to be irradiated and the specific experimental instrumentation such as thermocouples, neutron dosimeters, pressure sensors, strain gauges, displacement transducers. The sample–holder designed so far for CALIPSO contains 15 tubular samples other the 60 cm of the active zone (5 levels of 3 samples placed at 120°), with thermocouples and neutron dosimeters. Different sample holders will be designed to reach specific experimental objectives such as dose accumulation, irradiation growth, tensile and creep test.

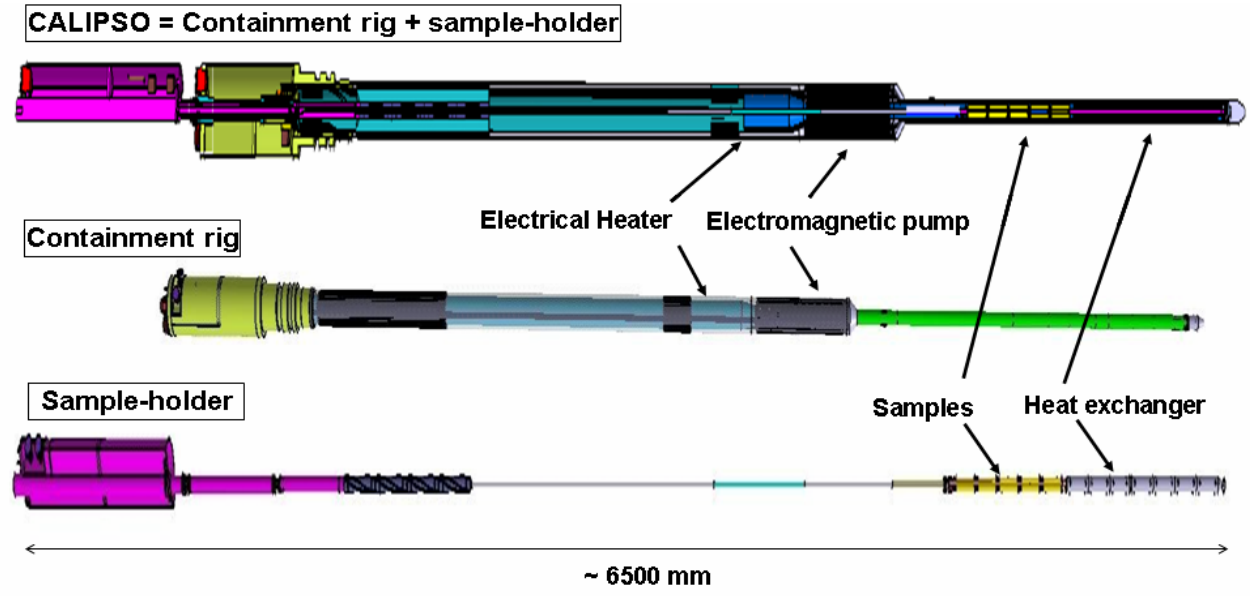


Figure 3 : Overview of CALIPSO and its main components

## 5. Electro-magnetic pump for NaK circulation

The annular electromagnetic pump is located above the active zone. It has been designed in order to fit in the available space of about 80 mm in diameter and for the following operating conditions: NaK flow rate 2 $m^3$/h, pressure drop 1.25 bar and NaK maximal temperature 450°C.

Induction coil and comb-shape ferromagnetic structures are mounted between the two tubes of the containment rig. In order to close the field lines, an annular core is placed in the centre of the pump. It is composed of ferromagnetic elements between two stainless steel tubes. The pumping channel results from the annular gap between the inner tube of the induction circuit and the outer tube of the magnetic core (Figure 4).

The three-phase inductor coil creates a magnetic field travelling along the annular duct gap, inducing electric current in the conducting liquid metal. The interaction between the resultant current and the magnetic field produces an electromagnetic body force pumping the NaK flow through the channel.

In the case of CALIPSO, the circulation in the pumping channel is bottom-up, and after being heated the return of the NaK is possible through the centre of the annular magnetic core.

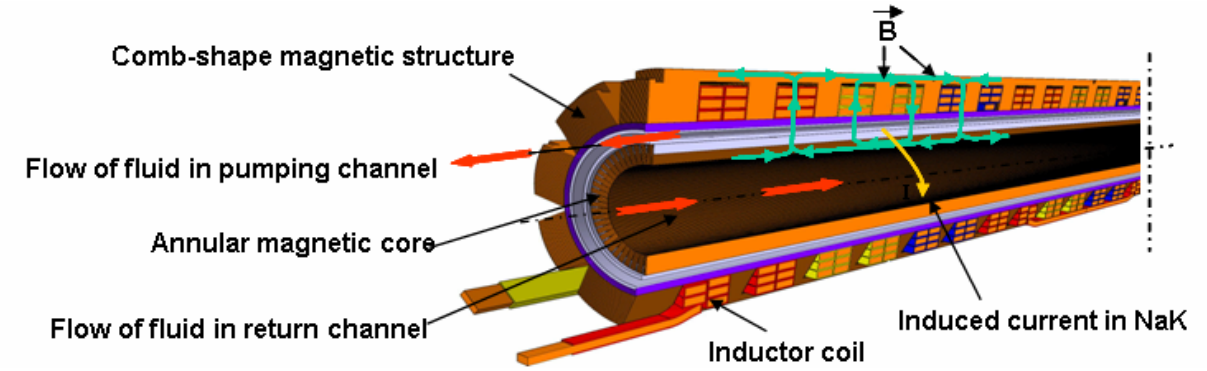


Figure 4 : Electromagnetic pump

## 6. Thermal balance and heat exchanger

The NaK temperature in the sample zone is the result of the thermal balance between the injected power and the heat losses. The main power deposits into the NaK come from nuclear heating into the samples, the test device structures and the NaK itself, but also from the electrical heater and the electromagnetic pump. The heat losses of the circulating NaK toward the water of the reactor primary circuit are controlled by three parameters:

- the NaK flowrate,
- the thickness of the Helium gas gap between the two tubes of the containment rig,
- and the length of exchange.

The temperature homogeneity in the sample zone depends on the heat exchange through the separator shell between the two reversed part of the NaK flow.

Sensibility studies were performed to determine the optimum geometry and operating conditions of the test device. The main parameters were tested taking into account thermal-mechanical dimensioning consideration. Therefore, to cope with the target of less than 7.5°C difference over the 600 mm of the fissile zone, and to cover a large range of nuclear heating (11 W/g - 23 W/g in steel) and NaK temperature (225°C - 450°C), the studies showed the necessity to have a variable length of exchange: from 100 mm to 800 mm.

A component called heat-exchanger located at the lower part of the sample-holder, below the active zone, is designed to be removed and replaced in hot cells (4). It allows having the NaK flow return at a variable altitude (Figure 5).

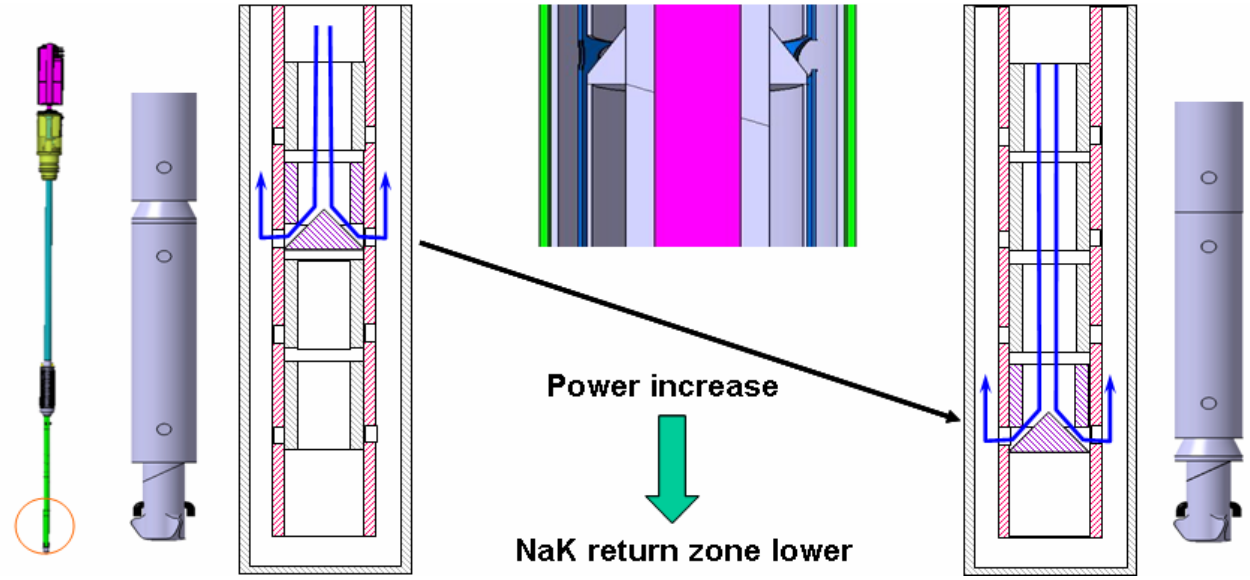


Figure 5 : Principle of the heat exchanger

## 7. Electrical heater

The external heating is provided through electrical elements inside two stainless steel cylinders and the gap between elements is filled with high thermal conductivity material (Figure 6). The maximal power will be roughly 20 kW over a total length of about 300 mm. This system allows heating the upward NaK before it returns and the downward NaK before it enters into the pump core. The injected power is an important parameter to control and adjust the operating conditions in the sample zone. Thermocouples will be placed into the NaK and inside the heating cylinder between electrical elements.

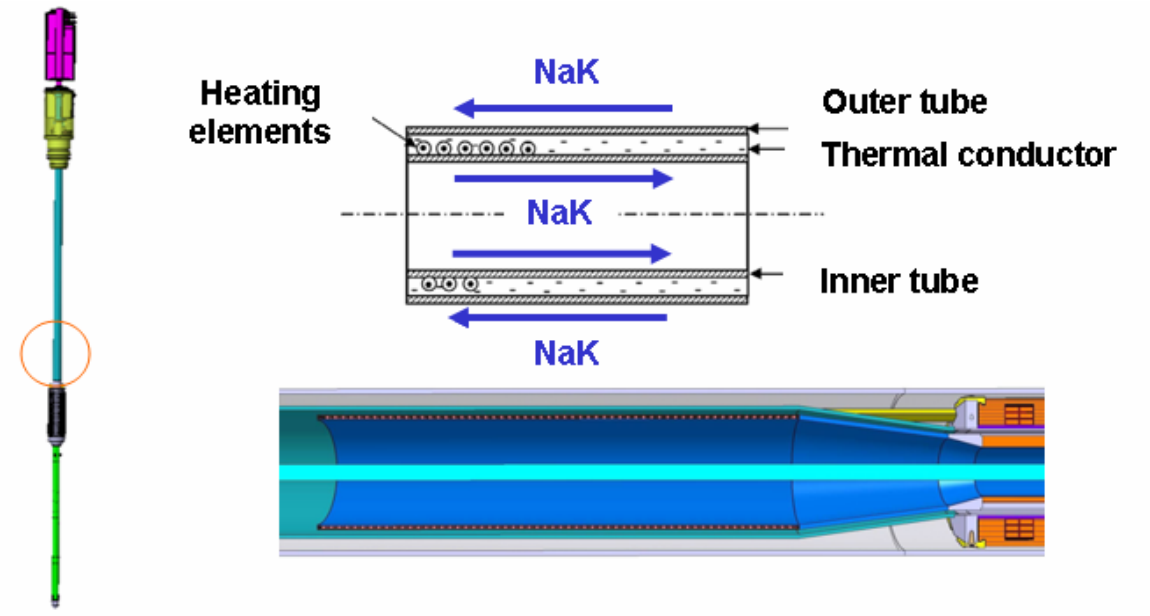


Figure 6 : Electrical heater

## 8. Safety requirements and pressurized equipment rules

The safety approach of the test device must be consistent with the safety rules applied to the reactor and to the nuclear pressurized equipment rules. In the case of CALIPSO, the major hazard source is the NaK circulation inside the experimental device. In order to prevent interaction between NaK and the water of the reactor primary circuit, which could damage the core fuel or other close experimental devices, the two test device envelopes will be classified as containment barrier and respect specific quality requirements following the RCC-MX mechanical reference code (5) (design, materiel supply, manufacturing and controls).

In the design phase, thermal-mechanicals calculations have been performed taking into account the irradiation effects on mechanical properties of stainless steel tubes. Geometry of the containment was adjusted to follow the reference code requirements in order to have maximal stress values less than admissible criterion.

## 9. Conclusion

Progresses on designing the CALIPSO device have been made concerning its operating range and its main components such as the electromagnetic pump, the electrical heater and the heat exchanger. The overall dimensions of the tubes have been defined according to the safety requirements and regulatory rules.

A contract towards industry is now in progress for the CALIPSO development covering the detail engineering design study and the manufacturing of a prototype. It will be qualify in an out-of-pile facility that will be built in 2010. The main objective will be to validate the dimensioning and the performances of the main components of CALIPSO and the global behaviour of the NaK loop.

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# EVITA: a semi-open loop in BR2 for RJH fuel qualification.

E. KOONEN, PH. GOUAT, PH. BENOIT, V. KUZMINOV
*SCK–CEN, Boeretang 200, B –2400 Mol - Belgique*

P. LEMOINE[(1)], M-C. ANSELMET[(2)],
*[(1)]CEA– Saclay, 91191 Gif sur Yvette – Cedex – France*
*[(2)]CEA– Cadarache, 13108 St Paul lez Durance – Cedex – France*

S. BRISSON, S. GUILLOT
*AREVA–TA, 1100, av JR Guilibert de la Lauzière, 13593 Aix-en-Provence– France*

## ABSTRACT

The qualification of the RJH fuel requires the demonstration of the satisfactory operation of an actual full-size fuel element in representative conditions. Because the flux characteristics of BR2 are closest to the future characteristics of RJH and because the fuels are of comparable design, test irradiations have already been made, including a mixed BR2-RJH fuel element, manufactured at BR2 standard dimensions.

However, for a full-size irradiation, there are considerable differences between the BR2 and the RJH regarding the full element diameter, the fuel cooling conditions and the pressure drop over the core. Fortunately, there are some channels in BR2, one of which is in the centre of the reactor, allowing the reach of the RJH required conditions with a limited equipping addition. Furthermore, the reactor is rated at 120 MW but operates around 60 MW, so there is ample cooling capacity.

So, EVITA is a semi-open loop, "grafted" on BR2 vessel, in which water is taken in the upper plenum and taken to a "booster" pump, installed in BR2 pool. The pump raises the available $\Delta P$ and re-injects the water in the central channel of BR2. The central channel is modified to accommodate the dimensions of the full-size RJH fuel element. The water re-mixes with BR2 primary circulation in the lower plenum of the reactor. This concept takes advantage of all safety systems of BR2 and does require minimal additional safety systems.

Given that there is no burnable poison in RJH fuel and that there is no way to move to a "warmer" or "colder" position to compensate for the fuel burnup, the biggest challenge with this irradiation will be to provide adequate - for RJH - irradiation conditions without penalising too much the other users and the productions of BR2.

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S. BRISSON, S. GUILLOT
*AREVA–TA, 1100, av JR Guilibert de la Lauzière, 13593 Aix-en-Provence– France*

## Introduction

For the REACTEUR JULES HOROWITZ (RJH) under construction at Cadarache, CEA and AREVA-TA designed a fuel element which has some similarities with the design of BR2 fuel, but needs to be qualified full-size due to its operating specificities [1]. BR2 appears to be the only reactor able to provide adequate irradiation conditions. The fuel has following characteristics:

- 8 plates fuel element, generating almost twice the power of a BR2 fuel element
- 96.2 mm O.D. fuel element (BR2 is 80 mm O.D.)
- $U_3Si_2$ 4.8 g/cc fuel enriched at 27% with 0,61mm meat thickness
- 516 W/cm² at the hot spot including fuel density uncertainty
- Up to 60 % burnup reached in 5 BR2 cycles
- Distance between plates: 1,95 mm (3,0 mm in BR2)
- 15 m/s average coolant velocity (10 m/s in BR2)

The requirement to irradiate a full-size fuel element led to the selection of the 200 mm O.D. H1 channel of BR2. This channel is the central position of the reactor and has to be modified to accommodate the RJH fuel.

The distance between plates is considerably smaller than at BR2. As the ΔP available on BR2 core is limited to 4 bars, a booster pump is required. Hence, **EVITA** stands for **E**nhanced **V**elocity **I**rradiation **T**est **A**pparatus.

In addition, such a small distance between plates does not allow natural convection to develop as it would in BR2. Hence, the "station black-out" accident, which is benign in BR2, has the potential to severely damage the RJH fuel.

## Fuel adaptation to BR2

In BR2, the normal practice is to keep the fuel element attached to its support rod and channel plug during the irradiation and this philosophy was kept for EVITA. Therefore, both end parts of the tested fuel were changed into the BR2 design, as shown on fig.1, in order to get similar handling procedures.

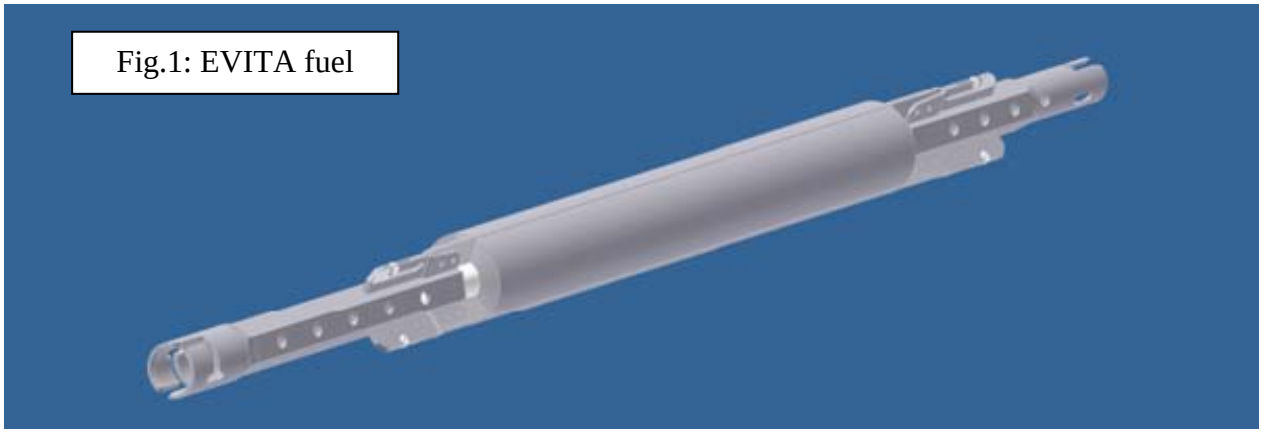


Fig.1: EVITA fuel

**General description**

The water is taken in the upper plenum of BR2 via two unused peripheral channels. Instead of allowing the flow downwards through the reflector, the suction channels forces it upwards to a pipe connected to the suction side of a booster pump, where its available ΔP is increased. The booster pump is installed submerged on BR2 working floor. The pumped water is injected in a modified central channel H1 of BR2 containing the fuel to be tested. The water re-mixes with BR2 water in the lower plenum. The fig.2 shows a realistic picture of the main piping in BR2 pool.

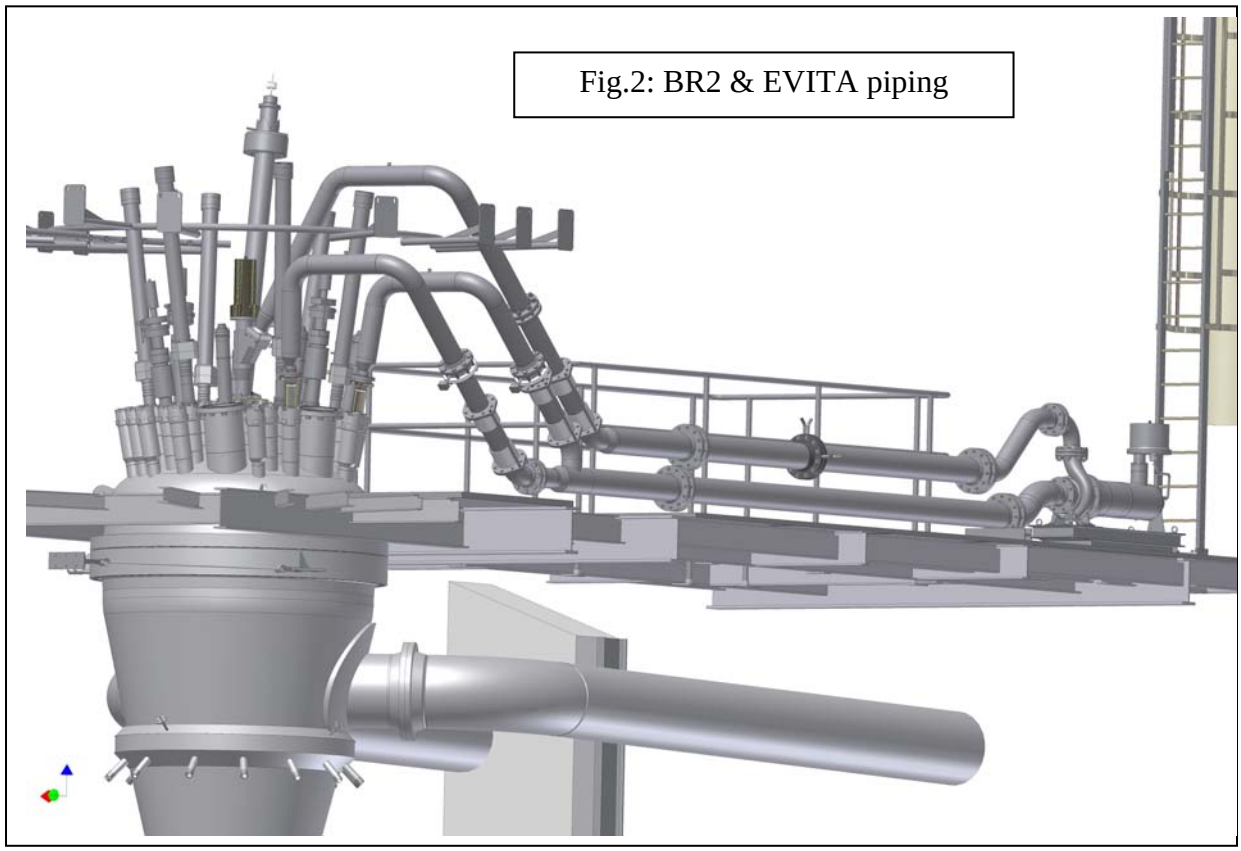


Fig.2: BR2 & EVITA piping

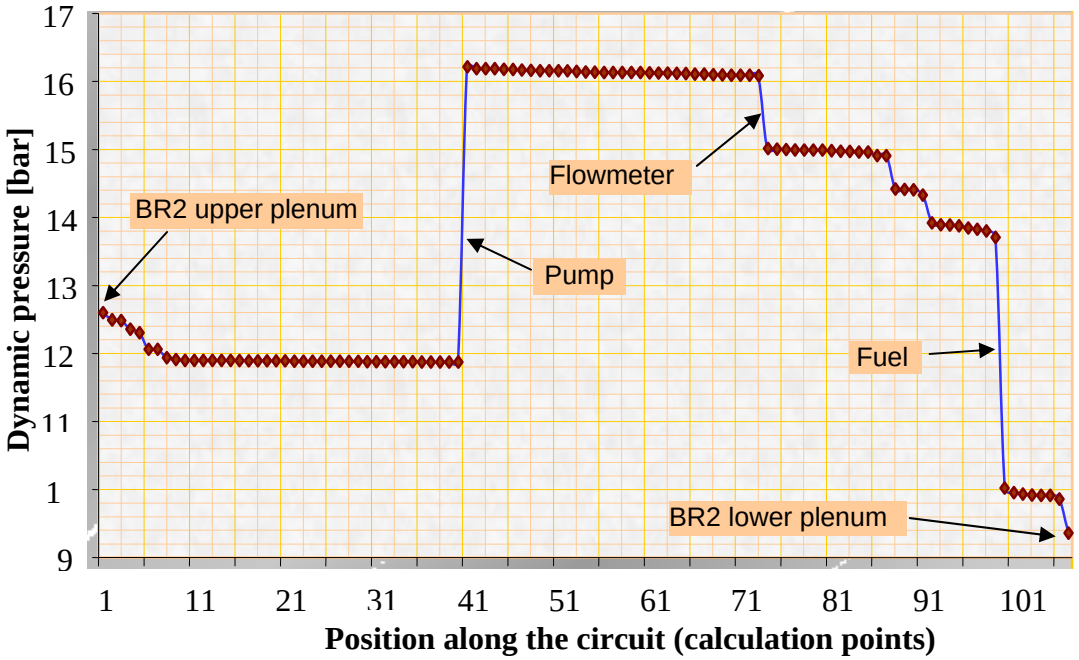
In the chosen configuration, there are always two pumps in series (BR2 primary pumps and EVITA booster pump) and no mobile flow restriction in the circuit. The main circuit of EVITA is completely enclosed in BR2 pool, thereby providing radiation protection for the personnel during the operation, insensitivity to leaks and protection of BR2 in case of LOCA. EVITA takes profit of all safety systems and protections of BR2.

The benefits of this configuration outweigh the cost increase for the submerged pump:

- No penetration of piping through the pool walls is used
- No room occupation around the pool (space availability is limited there)
- No need to install radiation shielding (against $N_7$ and possible fission products)
- No additional risk for BR2 in case of rupture of the piping
- Improved safety, when compared to non-submerged solution:
  - The ΔP provided by BR2 primary circulation remains available in case of failure of the booster pump. So reduced cooling remains available to remove the decay heat of the tested fuel.
  - EVITA being submerged in BR2 pool, a breach in EVITA piping will not lead to the fuel dry-out.
  - EVITA uses mostly BR2's own protection systems.

The pressure profile in EVITA is shown on fig.3 below. The diagram starts at 12,7 bar (static pressure of BR2 plus ΔP delivered by BR2 primary pumps, which is the pressure in the upper plenum of BR2) and ends at 9,3 bar, the pressure in the lower plenum of BR2.

Fig.3: Pressure profile in **EVITA**




.

If the EVITA booster pump fails, the ΔP delivered by BR2 primary pumps is still more than sufficient to remove the decay heat of the fuel element, and vice versa. If both pumps fail, which could happen in case of loss of external electrical power, a batteries powered UPS group will deliver current for one hour, at which time the residual heat can be removed by natural convection.

**Challenges**

When operating BR2, fresh fuel is initially loaded in a position at the periphery of the core and progressively moved towards the core center as the burnup accumulates. Furthermore, burnable poisons incorporated in the fuel help to mitigate the reactivity drop induced by the burnup.

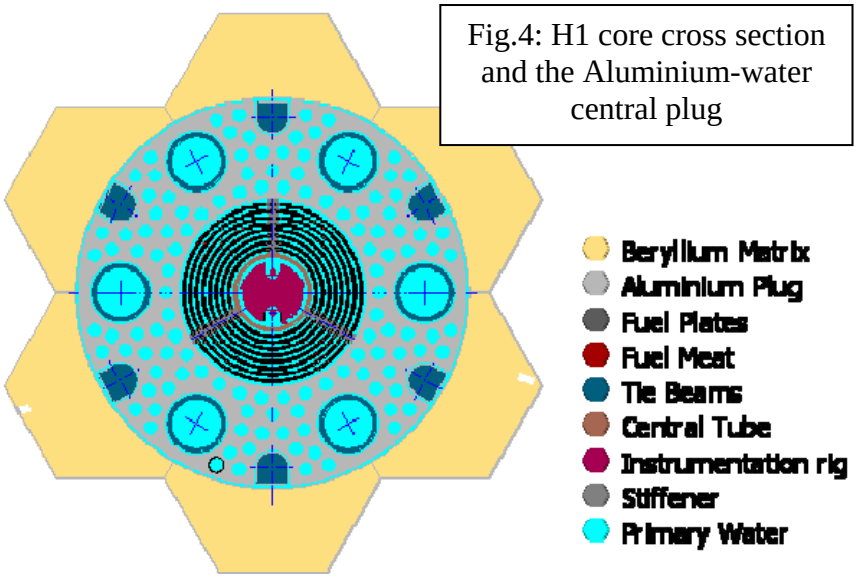



Fig.4: H1 core cross section and the Aluminium-water central plug

The RJH fuel does not incorporated burnable poisons and there is only one possible irradiation position. Hence, in this irradiation, the challenge is to operate BR2 with a fuel element in its center, operating at a required power level, while still being able to operate the reactor at a convenient power level for the other users at the periphery.

This is achieved by several means. First, there is the choice of materials surrounding the central fuel, a mixture aluminium-water which presents optimum absorption – moderation – transmission characteristics when the fuel is fresh and most reactive. Then, by playing on the number of Iridium production targets surrounding the central fuel, it is possible to compensate the decrease of reactivity of the central fuel caused by burnup accumulation and still operate BR2 at constant power. At the end of life, the aluminium-water mixture has to be replaced by beryllium, which was found better suited for depleted fuel.

The accuracy of the BR2 load determination before each cycle will be a critical element for this irradiation.

## Power generated in the RJH Fuel Element

The summary Table 1 contains the calculated power deposited into the cooling water between the fuel plates, and in both plugs. The total power in the fresh RJH fuel element was calculated for the model with Al plug in the central trap of the fuel element and for the power of BR2 equal to $Q_{BR2}$=62 MW.

| | | Irradiation |
|---|---|---|
| 1 | Fission and decay reactions | 4.77 MW |
| 2 | $\gamma^{prompt}$ in cooling water | 61 kW |
| 3 | $\gamma^{prompt}$ in Al cladding | 77 kW |
| 4 | $^{28}$Al decay in the cladding | 4 kW |
| 5 | Deposit of energy by $\gamma^{delayed}$ emitted by fission products to the cooling water and to Al cladding | 47 kW |
| 6 | **Total in fuel element, MW** | **5.0 MW** |
| | | |

Table 1. Thermal power in the RJH fuel element.

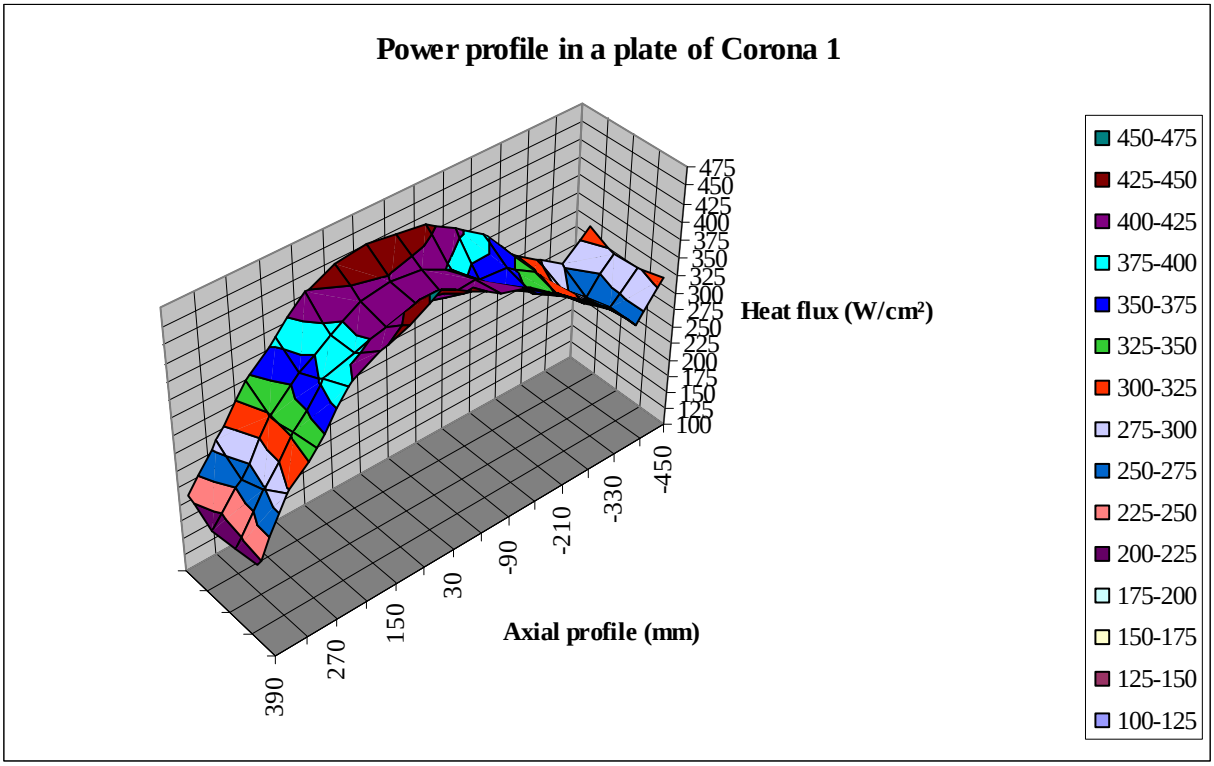



Fig.5: Power profile in the external plate

## Irradiation instrumentation

A number of parameters are measured during the irradiation, either directly, either after processing by the data acquisition system. The following are of particular interest for the follow-up of the fuel performance:

- The power generated, by thermal balance. The measure of the ΔT on the fuel assembly and of the flow rate delivered by the booster pump gives a value of the heat generated in the H1 Channel. The measurements must be corrected for the gamma heating and the heat losses to get the correct value of nuclear power generated in the fuel.
- The ΔP on the fuel assembly allows to detect on-line a possible excessive build-up of oxides on the fuel.
- The neutron flux and the gamma flux are directly measured by sensors attached to the aluminium rod filling the central hole of the fuel element. The signals delivered by the sensors are converted in usable values by the data acquisition system.

## Conclusions

Although the irradiation of a fuel element of similar design as the one of BR2 might seem something quite straightforward, the differences in geometry, cooling and irradiation conditions are such that a huge extension to BR2 has been made necessary.

Furthermore, the absence of burnable poisons for the compensation of the reactivity drop consecutive to the burnup accumulation, and the exclusive use of the central H1 central channel puts a formidable challenge on the operation of BR2. Indeed, the BR2 load, and even the general core configuration, has to be adapted, cycle by cycle, to compensate this effect and still provide acceptable irradiation conditions to other experimenters.

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Qualification Program for JHR Fuel ELEMENTS, This meeting

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# A MULTI-NATIONAL PRACTICAL TRAINING COURSE FOR NUCLEAR CANDIDATE COUNTRIES ORGANIZED BY EERRI

H. BÖCK, M. VILLA
*Vienna University of Technology, Atomic Institute of the Austrian Universities*
*Stadionallee 2, A-1020 Wien – Austria*

E. BRADLEY, A. SOARES, P. ADELFANG, I. GOLDMAN
*Division of Nuclear Fuel Cycle and Waste Technology/Research Reactor Group, IAEA*
*POB 100, A-1400 Wien – Austria*

S. TOZSER
*Atomic Energy Research Institute, KFKI*
*POB 49, H-1525 Budapest – Hungary*

A. ASZODI
*Institute of Nuclear Technology, Budapest University of Technology*
*POB 91, H-1521 Budapest – Hungary*

M. RAVNIK, L. SNOJ
*Jozef Stefan Institute*
*Jamova 39, SI-1000 Ljulbjana – Slovenia*

## ABSTRACT

An increasing number of Member States (MS) requests IAEA assistance to develop nuclear skills and resources in support of national nuclear power programmes under development. For countries with little or no existing nuclear infrastructure, human resources and skills must be developed to support planning, analysis, evaluation and other prerequisite activities for the decision making process. The Eastern European Research Reactor Initiative (EERRI) was approached by the IAEA to organize and implement a Group Fellowship Training Program on Research Reactors (GFTPRR) to satisfy the increasing demand for the aforementioned skill development. The GFTPRR will be offered to participants from MS who have expressed interest in this subject to the IAEA. The first training course is planned for spring 2009 with six participants organised by the Vienna University of Technology/Atominstitute, two Hungarian Nuclear Research Institutes and the Jozef Stefan Institute/Ljubljana, Slovenia. This paper presents the planning procedures, the detailed course content and logistics for the first model course.

## 1. Introduction

An increasing number of Member States (MS) are requesting IAEA assistance to develop nuclear skills and resources in support of national nuclear power programmes under development. Some of these MS are planning to construct a research reactor as a first step to develop nuclear competence and infrastructure. For countries with little or no existing nuclear infrastructure, human resources and skills must be developed to support planning, analysis, evaluation and other prerequisite activities involved in the related decision making process and subsequent projects. The Eastern European Research Reactor Initiative (EERRI) was approached by the IAEA to organize and implement a Group Fellowship Training Program on Research Reactors (GFTPRR) to satisfy the increasing demand for the aforementioned skill development. The GFTPRR will be offered to participants from MS who have expressed interest in this subject to the IAEA.

The program is being organized in collaboration with the Vienna University of Technology/Atominstitute (VUT/ATI). The first iteration will involve VUT/ATI, two Hungarian

Nuclear Research Institutes, and some staff members from the Jozef Stefan Institute's/Ljubljana (IJS), Slovenia. The duration of the training program is estimated between 4 to 6 weeks and will cover about 30 topics ranging from theoretical lectures to practical experiments at the TRIGA reactor Vienna and at the training reactor of the Budapest Technical University grouped into three main areas:

- Organisational Matters
- Research Reactor Operation and Maintenance
- Radiation Protection

The first training course is planned for spring 2009 with six participants. This paper presents the planning procedures, the detailed course content and logistics for the first model course.

## 2. Involved institutions

### 2.1 Vienna University of Technology/Atominstitute (VUT/ATI), Austria

The Atominstitute being the closest nuclear facility to the IAEA has long-term experience in organising national and international training courses. The course contents origins from the regular students curriculum at the Vienna University of Technology (VUT). The courses are part of the eligible course during the Masters Program in the Technical Physics Curriculum. There are three practical courses offered by the Atominstitute which are

- Practical Exercises in Reactor Physics and Kinetics
- Practical exercises in Reactor Instrumentation and Control
- Practical Exercises in radiation protection

Each of these exercises are composed of about 10 topics where the students have to work directly at the reactor in groups of maximum eight students according to the provide programme. Each exercise is introduced by theoretical part and followed by the practical experiment. As the TRIGA reactor is designed primarily for education and training the students also have the possibility to start-up the TRIGA reactor on their own towards the end of the course. Out of these totally 30 exercises any combination of exercises are possible according to the interest for the group. In addition a number of course are available covering all legal and technical aspects of research reactor planning an d operation. During the past 5 years the request for training courses have increased from about 10% to almost 50% which poses a limit to the course staff but also to the reactor availability.

### 2.2 KFKI Budapest, Hungary

The KFKI Atomic Energy Research Institute (AEKI) has been operating a research reactor, namely: Budapest Research Reactor (BRR) since 1959. Since its initial criticality, the BRR has been utilized as a neutron source for research and various industrial and health care applications, as well as education and training purposes in the nuclear field. The reactor contribution in the training course is aimed to the practical subjects. Thus, on the basis of the operational and utilization experiences the subjects of the training courses are:

Research Reactor operation and utilization matters
Water chemistry in general and in practice at BRR
Emergency procedures
QA issues at a research reactor and practical approaches of nuclear project planning and implementation

The listed subjects are divided in theoretical topics and on site training where the students can see how the operating matters are managed in the everyday practice. The students have

an opportunity to take an insight into the regulatory environment from the viewpoint of operators as well as from the specific research reactor management practice (the operation and utilization meters are separated) applied at BRR.

## 2.3 Budapest University of Technology, Hungary

The training reactor of the Budapest University of Technology and Economics is a pool type reactor located at the university campus. The facility was designed and built between 1961 and 1971, by Hungarian nuclear and technical experts. It first went critical on May 22, 1971. The maximum power was originally 10 kW. After upgrading, which involved modifications of the control system and insertion of one more fuel assembly into the core, the maximum licensed power was increased to 100 kW in 1980. The reactor still operates with the original LEU fuel assemblies. The reactor building houses reactor physical, radiation protection, and radiochemical laboratories, and a small hot cell too.

The main purpose of the reactor is to support education in nuclear engineering and physics for Hungarian graduate students, but international training courses (e.g. „Eugene Wigner Course for Reactor Physics Experiments") are organized and IAEA fellows are hosted too.
The reactor is used, among others, in the following fields: experiments in reactor physics and thermal-hydraulics, activation analysis for radiochemistry and archaeological research, analysis of environmental samples, determination of uranium content of rock samples, nuclear instrumentation development and testing, development and testing of tomographic methods for safeguards purposes, and investigation of radiation damage to instruments/equipment. The subjects in the training course will be the following:

- Thermal hydraulics
- Radiation protection and waste management
- Site requirements
- Public information

## 2.4 Jozef Stefan Institute, Ljubljana, Slovenia

Jozef Stefan Institute (JSI) has been operating a 250 kW TRIGA type research reactor since 1966. In 1991 it was reconstructed and equipped for pulse operation. The reactor has been used for neutron activation analysis, irradiation of various samples (semiconducting detectors, fusion reactor materials, etc.), neutron radiography, training (Slovenian nuclear power plant (NPP) reactor operators, students of University of Ljubljana, IAEA trainees from developing countries, etc.) and for testing and development of various computer codes. The reactor physics division at JSI has long-term experience in usage and development of various computer codes for reactor core calculations. Among others they have developed a program package for reactor calculation of TRIGA Mark II research reactor cores, TRIGLAV-W (http://www.rcp.ijs.si/triglav/), which will be explained and demonstrated during the course. Lately they have been working extensively also on validation and verification of modern Monte Carlo computer codes, such as MCNP, which will be presented at the course. Since the 1980's the group has been using the WIMS-D computer code together with other home-developed codes for performing core design of the NPP Krško in Slovenia. The WIMS-D will also be presented at the course. The computer codes mentioned above are one of the most commonly used codes for performing the reactor calculations world-wide. The trainees will become familiar with the WIMS-D, TRIGLAV-W and MCNP computer packages and will learn how to calculate various research reactor physics parameters and models that lie behind the calculations. They will also learn the basics of burn-up calculations and core optimization. The lectures will be followed by computer exercises and practical case studies.

# 3. Research Reactor Situation Worldwide

According to the latest IAEA information totally 768 research reactors have been built world wide, from these 170 have decommissioned, 248 are in various shut down states and 250 are operational and one is under construction. A number of countries exist who have not been involved in any nuclear activities but they realize that nuclear power could be an option for the future. Although in these countries highly trained technical staff is available in other fields such as oil or chemical industry there is however a lack of nuclear experience. The normal first step to nuclear is the planning of a research reactor as this was the usual way in the late 50ies and early 60ties for industrialized countries to gain nuclear experience and to step further into nuclear power. This is the reason why it was decided to offer a training course covering all aspects of the pathway to nuclear.

## 4. EERRI Training Course Background Information

The Research Reactor Group of the Nuclear Fuel Cycle and Waste Technology Division, organises a Group Fellowship Training programme to assist Member States that consider building a Research Reactor as a first step to develop nuclear competence and infrastructure in the Country. The training programme has been elaborated with the purpose to assist such Member States in the pursuit of these ambitious endeavours. It will help develop the necessary skills and background to carry out activities related to planning, evaluating, development, construction, commissioning, operation and maintenance of research reactors.

The programme has been organised within the framework of the “Eastern European Research Reactor Initiative (EERRI)”. In its inaugural edition in spring 2009, the programme will involve the participation of 4 institutions that operate research reactors, the Atominstitute of the Vienna University of Technology (Vienna-Austria), the Jozef Stefan Institute (Ljubljana - Slovenia), the KFKI Atomic Energy Research Institute and the Budapest University of Technology and Economics, the last two from Budapest, Hungary.

The programme is aimed at young, technical professionals who have no nuclear experience. Candidates should have technical degrees in engineering or science and may eventually be responsible for research reactor activities in countries that have no experience with such facilities. Participants may also include technical professionals seeking hands-on nuclear rector facility experience and familiarisation to support the development of a nuclear power programme. Due to limited infrastructure for practical/experimental activities, for each edition the programme at most 8 students may be accommodated. If the programme is successful it may be repeated in Fall 2009 depending on the outcome of this first programme and ongoing interest from IAEA Member States.

- Administrative and organisational topics such as regulatory requirements, site requirements nuclear project planning and implementation/control, decommissioning planning and implementation
- Reactor related topics such as reactor physics, I&C Systems, thermo-hydraulics, maintenance and inspection programs
- Radiation monitoring such as radiation protection, personnel and environmental monitoring
- Practical courses such as Practical courses on reactor physics and kinetics, on I&C systems, in radiation protection and dosimetry and on fuel management

The overall time of this course is scheduled for five weeks where the participants start in Vienna due to the location of the IAEA and the Atominstitute. The first three weeks will take place mainly at the Atominstitute with some presentations also at the IAEA. To reduce travel expenses the lecturers from the Institute Josef Stefan/Slovenia will come to the Atominstitute and present their contribution in Vienna. After these three weeks the participants will move to Budapest by train for one week of training at the KFKI and one week at the TU Budapest. The termination of this course is planned again in Vienna together with the IAEA. When planning the course other institutes (i.e. Rez/Czech Republic,

TU Prague, Swierk/Poland, Pitesti/Romania) indicated also their interest to contribute however to save travel costs the present schedule was selected for the initial course . It is however easily possible in future to offer this course also in cooperation with the above mentioned institutes. The proposed course schedule is shown below:

| Education and training subjects | TU Vienna | NRI Rez | TU Prague | BRR Budapest | TU Budapest | TRIGA Ljubljana | MARIA Swierk | TRIGA Pitesti |
|---|---|---|---|---|---|---|---|---|
| Regulatory requirements | | | Yes | | Yes | | | Yes |
| Code of Conduct | | Yes | | | | | Yes | Yes |
| RR Management | | Yes | Yes | Yes | | | Yes | Yes |
| Staffing requirements | | Yes | Yes | | | | | Yes |
| Site requirements | | | | | Yes | | | |
| Waste management | | | | | Yes | | | Yes |
| Public information | | | | Yes | Yes | | | Yes |
| Physical security | | | Yes | | | | | Yes |
| Emergency procedures | | Yes | Yes | Yes | Yes | | Yes | Yes |
| Nuclear project planning and implementation/control | | | | Yes | | | | |
| Decommissioning planning and implementation | | Yes | | | Yes | | Yes | Yes |
| RR Overview | Yes | | Yes | Yes | Yes | | Yes | Yes |
| RR Utilization | Yes | Yes | Yes | Yes | | | Yes | Yes |
| Introduction to atomic and nuclear physics | Yes | | Yes | | Yes | | | Yes |
| Reactor physics | Yes | | Yes | | Yes | Yes | Yes | Yes |
| I&C Systems | Yes | | Yes | | | | | Yes |
| Thermohydraulics | | | | | Yes | | Yes | Yes |
| Maintenance and inspection programmes | Yes | Yes | Yes | Yes | | | Yes | Yes |
| Fuel management | | Yes | Yes | Yes | | Yes | Yes | Yes |
| Fuel Cycle | | Yes | | Yes | Yes | Yes | Yes | Yes |
| Water chemistry | | Yes | | Yes | | | | |
| Radiation protection | Yes | Yes | Yes | Yes | Yes | | | Yes |
| Personnel monitoring | Yes | Yes | Yes | Yes | Yes | | | Yes |
| Environmental monitoring | Yes | Yes | | Yes | Yes | | Yes | Yes |
| Practical course on reactor physics and kinetics | Yes | | Yes | | Yes | | Yes | Yes |
| Practical course on I&C systems | Yes | | Yes | | | | | Yes |
| Practical course in radiation protection and dosimetry | Yes | | Yes | Yes | Yes | | Yes | Yes |
| Practical course on fuel management | | Yes | Yes | Yes | | Yes | Yes | Yes |

| Day | Morning 9-12h, 20 min Coffee | Afternoon 13-16h Break as required | Location |
|---|---|---|---|
| WEEK 19 | | | |
| 4. May | Administrative Procedures Visit to the TRIGA facility | Research Reactor Overview | ATI |
| 5. May | RR utilization | RR vs Nuclear Power Plants | ATI |
| 6. May | Regulatory requirements | RR Staffing | ATI |
| 7. May | Strategic planning for RR | Code of Conduct for RR | ATI |
| 8. May | Introduction to Radiation Protection and instrument demonstration | Test and discussion on week no 1 | ATI |
| WEEKEND | VIENNA | | |
| WEEK 20 | | | |
| 11. May | Reactor Physics 1 | Determination of the thermal neutron flux density in the TRIGA reactor | ATI |
| 12. May | Reactor Physics 2 | Power calibration and temperature coefficient | ATI |
| 13. May | Critical experiment | Calibration of control rods, etermination of reactivity worth and excess reactivity | ATI |
| 14. May | RR I&C systems | Demonstration of I&C Detectors | ATI |
| 15. May | Demonstration of fuel handling and fuel transfer | Test and discussion on week no 2 | ATI |
| WEEKEND | VIENNA | | |
| WEEK 21 | Preparation of Safety Analysis Report (SAR) | Safety assessment for RR | ATI or IAEA |

| | | | |
|---|---|---|---|
| **18. May** | **Training of operating personnel** | **Special applications of RR (BNCT, Silicon doping, isotope production etc)** | **ATI or IAEA** |
| **19. May** | **RR maintenance and in-service inspections** | **RR Decommissioning** | **ATI or IAEA** |
| **20. May** | **NPP-PWR overview** | **NPP-BWR overview** | **ATI or IAEA** |
| **21. May** | **Public Information** | **Physical security** | **IAEA** |
| **22. May** | **Demonstration of prompt criticality** | **Test and discussion on week no 3** | **ATI** |
| **WEEKEND** | **VIENNA** | | |
| **WEEK 22** | | | |
| **25. May** | **Introduction to reactor calculations** | **Introduction to computer codes WIMS (demonstration and computer exercises)** | **IJS at ATI or IAEA** |
| **26. May** | **RR reactor physics parameters and models** | **Introduction to computer codes - TRIGLAV (demonstration and computer exercises)** | **IJS at ATI or IAEA** |
| **27. May** | **Calculation of RR safety parameters** | **Introduction to computer codes MCNP (demonstration and computer exercises)** | **IJS at ATI or IAEA** |
| **28. May** | **Burn-up calculations and core optimization** | **Questions and problems from the participants (discussion and computer exercises)** | **IJS at ATI or IAEA** |
| **29. May** | **Questions and problems from the participants (discussion and computer exercises)** | **Questions and problems from the participants (discussion and computer exercises) Test and discussion on week 4** | **IJS at ATI or IAEA** |
| **WEEKEND** | **TRANSFER FROM VIENNA TO BUDAPEST** | | |
| **WEEK 23** | | | |
| **1. June** | **The BRR (VVR-10 MW) reactor as a tank type RR reactor** | **Reactor systems - site visit BRR's utilization** | **KFKI Budapest** |
| **2. June** | **RR management (operation and utilisation issues)** | **Emergency procedures** | **KFKI Budapest** |
| **3. June** | **Water chemistry in general and in practice at BRR** | **Personal monitoring Environmental monitoring** | **KFKI Budapest** |
| **4. June** | **Quality assurance in practice at a research reactor** | **Nuclear project planning and implementation** | **KFKI Budapest** |
| **5.June** | **Site visit** | **Site requirements, Public information Test and discussion on week 5** | **TU Budapest** |
| **WEEKEND** | **BUDAPEST** | | |
| **WEEK 24** | | | |
| **8.June** | **Thermal hydraulics** | **Thermal hydraulics** | **TU Budapest** |
| **9.June** | **Waste management** | **Radiation protection** | **TU Budapest** |
| **10.June** | **Practical course in radiation protection and dosimetry** | **Practical course in radiation protection and dosimetry Test and discussion on week 6** | **TU Budapest** |
| **11.June** | **TRANSFER FROM BUDAPEST TO VIENNA** | | |
| **12.June** | **FINAL COURSE DISCUSSION** | | **IAEA** |

## 5. Conclusions

It has been demonstrated that among the various research reactors in Central and Eastern Europe a very positive and beneficial co-operation is possible which can be offered to nuclear candidate countries. In addition part of these courses could also be used for training or retraining of junior and/or senior research reactor operators as the various course modules can be extracted from the above mentioned course can be and recombined in a customized manner.

# THE JAMAICAN SLOWPOKE-2 RESEARCH REACTOR: NEUTRON ACTIVATION ANALYSIS IN ENVIRONMENTAL AND HEALTH STUDIES

***Charles Grant, Gerald Lalor and John Preston***

*International Centre for Environmental and Nuclear Sciences*

*University of the West Indies*

*Mona Road, Kingston 7 – Jamaica*

## ABSTRACT

In its 24 years of existence the reactor has been utilized mainly for Neutron Activation Analysis (NAA) and has played an important role in the development of research programs in the areas of archaeology, biology, chemistry, forensics, geochemistry, and mining as well as for the production of short lived radioisotopes for experimental work in the physics department. However, over the last fifth teen years our main thrust has been environmental geochemistry, agriculture and health related studies, with interesting results that have implications for land use, farming practices, diabetic control and dietary intakes during pregnancy.

## 1. Introduction

The SLOWPOKE-2 research reactor at the International Centre for Environmental and Nuclear Sciences (ICENS) is a light water moderated open swimming pool type reactor with a beryllium reflector. The first criticality was achieved in March 1984. The reactor in Jamaica is one of only seven ever built and is the only one outside of Canada. Presently there are still five operating, three of which utilise HEU [1] fuel and two LEU [2]. Although Jamaica presently operates with a HEU core, we are committed to convert to LEU and fully support the Global Threat Reduction Initiative (GTRI) and Reduced Enrichment for Research and Test Reactors (RERTR) programs.

The reactor normally operates at a power of 20 kW for approximately 5 hours per day, 5 days per week. It has now operated for a total $9.78 \times 10^4$ -kilowatt hours out of a calculated lifetime of $3.395 \times 10^5$ kWh or 28.8% of total lifetime. Although only rated at 20 kW, SLOWPOKE-2 has the highest flux to power ratio of research reactors in this class. The remarkable flux stability of the SLOWPOKE-2 has provided ICENS with a robust, reliable and economical analytical platform.

Although NAA is now a mature technique, improvements in gamma spectroscopy detectors and fast counting systems have lead to incremental reductions in detection limits allowing NAA to remain competitive with newer techniques such as ICPMS. The ability to quantify elements in almost any matrix without dissolution and the dynamic analytical quantification range make NAA an ideal tool for environmental studies where sample types vary from geological samples with percentage level concentrations to biological samples with elemental concentration levels in the parts per billion range.

This presentation reports on the performance of the SLOWPOKE-2 reactor and its utilization for the analysis of a wide variety of environmental samples using NAA. The detection limits for the major sample types are presented along with data obtained, for soils, plants, animal and human tissues and there possible implications.

## 2. Methodology

### 2.1 Irradiation Facilities

Brief technical specifications for both the LEU and HEU cores of SLOWPOKE-2 are shown in Table 1, and highlights the expected increase in performance of the LEU core [2].

| Parameters | HEU | LEU |
|---|---|---|
| Maximum operating power | 20 kW | 20 kW |
| Maximum operating time at 3 mk | 6 hrs | 12 hrs |
| Maximum operating time at 4 mk | 16 hrs | 24 hrs |
| Core Life-Time | 20 Years | 40 Years |

Tab 1: Performance Comparison of HEU & LEU Cores

### 2.2 Neutron Activation Analysis

Unlike most reactors quantification by NAA with SLOWPOKE using the in-core irradiation sites does not require co-irradiation of flux monitors or standards. The uniformity, stability and reproducibility of the neutron spectrum within the core, and with time, facilitates the use of activation constants [3]
The irradiation, decay and counting schemes utilized in the acquisition of all data presented here are shown in Table 2.

| Schemes | Sample mass(g) | Flux x $10^{11}$ $ncm^{-2}s^{-1}$ | Irradiation time | First Decay | First count | Second Decay | Second count |
|---|---|---|---|---|---|---|---|
| Geological Short | 0.20 | 2.5 | 2 | 10 min | 5 min | 60 min | 10 min |
| Geological Long | 0.25 | 10.0 | 60 | 4-5 d | 2 hr | 21 d | 5 hr |
| Biological short | 0.7 | 5.0 | 3 | 3 min | 5 min | 40 min | 10 min |
| Biological long | 1.0 | 10. | 240 | 2-3(5*) d | 3 hr | 14 d | 6 hr |

*Human and animal tissue

Tab2: Optimized irradiation and measurement conditions

## 3. Results

All batches of samples irradiated during this work were irradiated along with Standard reference materials (SRM) as part of the quality control, selected SRM's wherever possible, were matrix match to sample types. SRM's were purchased either from NIST or IAEA and were generally within ± 20% or better of certified values [4, 5, 6].

| Element | Geological | | Plants | | | | Animal | | | | Human | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Soil (N = 900) | | Root Crops (N = 600) | | Leafy Vegetables (N = 40) | | Bovine Liver Kidney (N = 200) | | Fish (N = 20) | | Placenta (N = 100) | | Blood (N = 80) | |
| | Range | D.L | Range | D.L | Range | D.L | Range | D.L | Range | D.L | Range | D.L | Range | D.L |
| Al | 1.9 – 24 * | 0.05 | <0.5 - 459 | 0.5 | 353 – 3718 | 6 | 1.5 - 272 | 1 | - | - | - | 13 | - | - |
| As | 3 - 380 | 0.2 | - | 0.01 | 0.02 – 0.97 | 0.02 | 0.1 – 1.4 | 0.07 | - | - | - | 0.1 | - | - |
| Au (ppb) | <5 - 64 | 5 | 0.5 – 2.0 | 0.01 | 0.4 – 40 | 0.1 | - | 0.4 | - | - | - | 1 | - | - |
| Br | 4 - 148 | 0.4 | 0.28 - 19 | 0.03 | 0.1 – 450 | 0.06 | 3 - 356 | 0.15 | - | - | 15.4 - 80 | 0.2 | - | - |
| Ca | 0.2 – 28* | 0.3 | <40 - 3980 | 40 | 1.3 - 3.75 | 44 | <12 - 2360 | 12 | - | - | 1126 - 32480 | 450 | - | - |
| Cd | 0.3 - 1000 | 2 | 0.06 - 4 | 0.05 | <0.1 – 151 | 0.1 | <0.4 - 449 | 0.4 | - | - | <0.4 | 0.4 | - | - |
| Co | 1 - 105 | 0.3 | - | 0.04 | 0.14 - 2.51 | 0.01 | 0.08 – 0.9 | 0.01 | 0.06 – 0.16 | 0.02 | 0.06 - 0.2 | 0.015 | - | - |
| Cr | 31 - 1100 | 7 | <0.1 - 3 | 0.1 | 0.16 – 12 | 0.1 | 0.3 - 4 | 0.1 | 0.1 – 2.9 | 0.15 | 0.2 - 96 | 0.2 | <0.01 – 0.14 | 0.01 |
| Cs | | 0.4 | <0.004 - 0.2 | 0.004 | <0.01 - 0.6 | 0.01 | 0.003 - 1.8 | 0.001 | 0.01 – 0.35 | 0.001 | 0.002 - 0.075 | 0.002 | - | - |
| Eu | 0.4 - 12 | 0.1 | - | 0.002 | 0.01 | 0.01 | - | 0.001 | - | 0.001 | <0.002 | 0.002 | - | - |
| Fe | 1.3 – 22* | 0.04 | 7 - 59 | 4 | 14 - 300 | 10 | 116 - 1250 | 6 | 16 - 98 | 5 | 494 - 3166 | 7 | 520 - 1080 | 15 |
| Hg | - | 1 | - | 0.05 | - | 1 | <0.02 - 0.18 | 0.02 | 0.1 - 24 | 0.02 | 0.02 – 0.3 | 0.02 | - | - |
| K (%) | 0.01 - 3 | 0.002 | 0.47 - 2.3 | 0.01 | 0.1 – 15 | 0.01 | 0.7 - 1.3 | 0.04 | - | - | 0.8 - 1.3 | 0.05 | - | - |
| La | 4 - 302 | 0.3 | - | 0.01 | 0.1 – 7 | 0.03 | 0.009 - 0.4 | 0.005 | - | - | <0.07 | 0.007 | - | - |
| Mg | <0.5 – 3.7* | 0.1 | 200 - 900 | 150 | 4470 – 9997 | 110 | 745 - 2410 | 450 | - | - | - | 750 | - | - |
| Mn | 10 – 7500 | 3 | 1 - 15 | 0.3 | 76 – 678 | 0.4 | 2.9 - 4480 | 1.5 | 0.1 - 4 | - | <0.3 - 8 | 0.3 | - | - |
| Na | 0.02- 2.8* | 0.0004 | 6.9 - 630 | 1 | 13 – 4242 | 1 | 6160 - 11233 | 2 | - | - | 0.9 - 1.4 | 4 | - | - |
| Rb | <50 - | 50 | 0.9 - 312 | 0.2 | 0.5 – 81 | 0.5 | 3 - 98 | 0.3 | 1 - 16 | - | 11 - 22 | 1.4 | - | - |
| Sc | 5 – 64 | 0.04 | - | 0.01 | 0.001 - 0.9 | 0.01 | - | 0.02 | - | 0.02 | <0.001 | 0.001 | - | - |
| Se | - | 7 | <0.04 - 0.13 | 0.04 | <0.05 - 0.7 | 0.05 | 0.2 - 10 | 0.1 | 1 - 10 | 0.1 | 0.9 - 1.4 | 0.2 | 0.16 – 0.3 | 0.1 |
| Sm | 0.85 – 49 | 0.01 | - | 0.01 | 0 - 0.7 | 0.01 | - | 0.01 | - | 0.1 | <0.002 | 0.002 | - | - |
| Sr | 80 – 700 | 50 | <2 - 7 | 2 | <6 – 616 | 6 | <4 - 11 | <4 | 7 - 80 | 3 | <2.8 | 2.8 | - | - |
| Th | 1.9 – 39 | 1 | - | 0.004 | 0.06 - 0.9 | 0.01 | - | 0.05 | - | 0.02 | <0.009 | 0.009 | - | - |
| Ti | 0.13 – 2.0* | 0.1 | - | 10 | 76 – 199 | 30 | - | 0.1 | - | 0.1 | - | 0.1 | - | - |
| U | 1.6 – 25 | 0.3 | - | 0.02 | <0.02 - 0.5 | 0.02 | <0.05 - 0.6 | 0.05 | - | 0.05 | <0.05 | 0.05 | - | - |
| V | 29 – 793 | 15 | - | 0.5 | 0.8 – 6 | 0.1 | - | 0.5 | - | 0.5 | - | 0.5 | - | - |
| Zn | 75 - 936 | 45 | 5 - 23 | 0.4 | 9 – 222 | 1.5 | 10 - 254 | 0.7 | 11 - 290 | 0.5 | 49 - 87 | 1.8 | 10 - 22 | 1.9 |

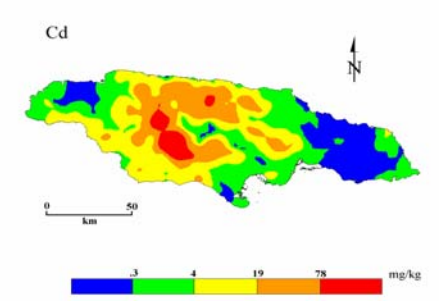
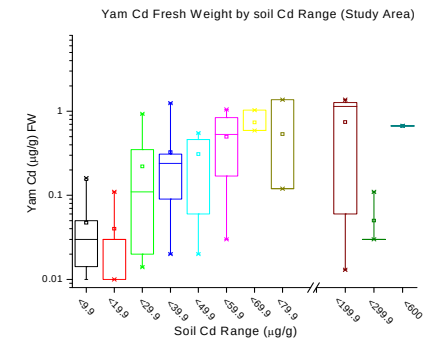
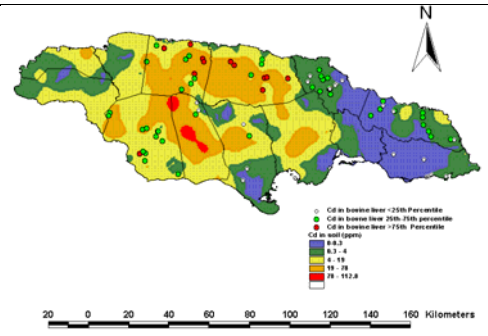
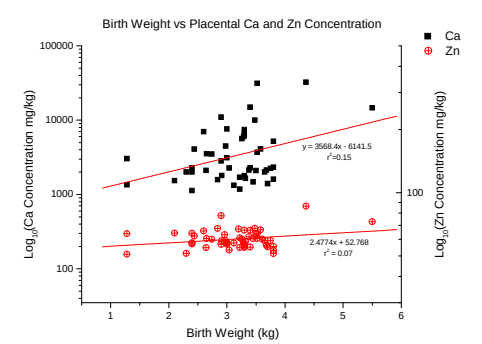
*%

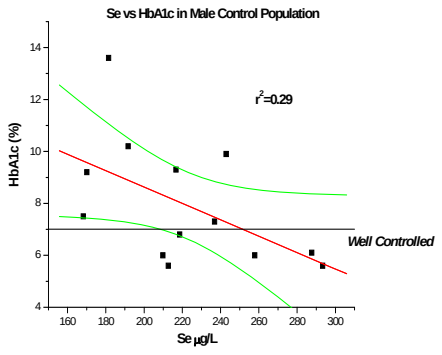
Tab 3: Concentration Ranges and detection limits for selected elements in various matrices

### 3.1 Detection limits

Although it is often stated that NAA is matrix independent orders of magnitude differences in detection limits for particular elements are obtained dependent upon matrix. Many medium and short-lived elements in Jamaican soils for instance are severely affected by elements that have either high cross-section and or high concentrations that produce gamma rays of energies very close to or identical to the gamma rays of the elements we wish to quantify [7].

## 4. Discussion

| Figures | Discussion Points |
|---|---|
|  | Soil<br><br>During an island wide soil survey it was found that some Jamaican soils, especially the bauxitic soils that overlie White Limestone geological group, are much enriched in several heavy metals compared with world levels [8]. The soil cadmium levels in particular are up to a thousand times higher than the world averages. These high levels in soils have encouraged efforts to establish relationships between plant animal and human health. |
|  | Yam<br><br>The exact locations of over 600 paired soil and food samples were recorded by use of global positioning systems (GPS). This allowed precise plotting of maps showing Cd in soils and foods stratified by soil Cd concentrations. There were significant soil/plant Cd correlations ($R^2$ >0.5); typically with soil Cd levels greater than 10 mg $kg^{-1}$ the yams will exceed the 0.1 mg $kg^{-1}$ limit set by the EU. These data confirm the significant uptake of Cd by some foods and show that low-Cd products can be produced by judicious land use selection; a technically simple solution to meeting food standards. [9] |
|  | Bovine Liver and Kidney<br><br>Paired liver and kidney samples from 100 free-range cattle in different parts of Jamaica were analyzed for the essential and non-essential trace elements. The map shows that the Cd levels found in cattle kidney closely followed that of the soil on which they reared. The intake of Cd from bovine liver and kidney was estimated to be 5.2µg/day based on an Island wide survey or 7% of the Provisional tolerable daily intake. [10] |
|  | Low Birth Weight and Calcium<br><br>Approximately 100 placenta samples were dried and ground into a fine powder. The samples were analyzed by NAA for Br, Ca, Cl, Fe, Hg, K, Na, Rb, Se and Zn, a further 20 elements were below the LOD. Approximately 12% of the population had low birth weight (LBW), <2.5kg, with a further 4% with very low birth weight (VLBW), 1.5kg. Higher birth weights were well correlated with the Placenta Ca content and to a lesser extent Zinc. (To be published) |

| | |
|---|---|
|  | Diabetic control and Selenium<br><br>Approximately 80 red blood cell samples (40 Diabetic and 40 Control) were analyzed by NAA for Cr, Fe, Rb, Se and Zn. To determine the effect of trace elemental concentrations on the diabetic control the haemoglobin A1c (HbA1c) was also measured and correlated with elemental data. The test for HbA1c is a key indicator of diabetes control. It measures the amount of sugar attached to haemoglobin, the primary protein in red blood cells and indicates a patient's average blood sugar level over the previous two to three months. Further work on the relationship between Se and Cr and diabetic control will continue. (To be published) |

## 5. Conclusion

Although the neutron flux of SLOWPOKE is relatively low, the reduced scale of operation makes it possible to safely house the gamma spectroscopy systems close to the irradiation facility, thereby reducing transfer times from irradiation site to counting systems, as a result small reactors are ideally suited for short-lived analysis and can obtain detection limits in biological and geological matrices that are similar or better to those obtained in larger reactors. The data presented here illustrates how NAA can be used to provide valuable data applicable across a wide spectrum of programmes, particularly in the earth and environmental sciences. It is this type of application that may well play an important role in the future of small reactor facilities.

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# PARTIAL DISMANTLING OF RESEARCH REACTOR-SOFIA PRIOR ITS REFURBISHMENT INTO LOW POWER REACTOR

T.G.APOSTOLOV, E.I.ANASTASOVA, V.D.ANASTASSOV, A.D.MLADENOV
*Institute for Nuclear Research and Nuclear Energy/Bulgarian Academy of Sciences*
*72, Tzarigradsko chausse blvd., 1784 Sofia - Bulgaria*

## ABSTRACT

The strategy for the Research Reactor IRT-Sofia, after decision of the Government for its refurbishment, is a partial dismantling of the old systems and equipment. Removal of the reactor core and replacement of old equipment will not pose any significant problems for dismantling. For most efficient use of resources there is a need for implementation of the engineering project, “Plan for partial dismantling of equipment of the IRT-Sofia as a part of its refurbishment and conversion into low power RR”, which has been already prepared.

*Key words: partial dismantling, decontamination, reactor equipment, radioactive wastes*

**The IRT-Sofia research reactor was designed and constructed in the period from 1958 to 1961. First criticality was reached in September 1961 and the research reactor (RR) was in operation for 28 years, until July 1989. The reactor was started up 4189 times, and it was in operation 24623 hours altogether, at different power levels (by 2 MW) requested by the users at regular weekly meetings. It was shut down in 1999 after the Council of Ministers of the Republic of Bulgaria decision.**

**The IRT-Sofia is a pool type reactor, cooled and moderated with light water. The core contains up to 48 fuel and graphite assemblies. There are 14, 15 or 16 fuel rods per assembly. The fuel rods are of the EK-10 type (10% enrichment) and C-36 (36% enrichment). The reflector includes 13 graphite blocks.**

**The principal areas of reactor usage ranged from basic and applied research, to technological and commercial applications. Moreover, it was an important place for university and post graduate education and training.**

**To carry out the resolution # 552 of the Council of Ministers of the Republic of Bulgaria from 2001 for refurbishment and partial dismantling of the Research reactor IRT-Sofia during its conversion into a low-power reactor, a Plan for the partial dismantling of the RR has been developed, according to IAEA Safety Guides [1] and [2]. We are going to carry out this plan in the near feature.**

The partial dismantling activities are part of the overall process of IRT-Sofia Research Reactor refurbishment. The final state of the IRT-Sofia after the partial dismantling will be the initial state of mounting the IRT-Sofia new systems and equipment. The criteria that should be taken into account are the following:

1. Ensuring full technological possibility for mounting of the new systems and equipment in the reactor pool, the shaft for SNF repository and the primary circulation loop.
2. Removal of the dismantled equipment from the mounting sites (Reactor hall and the rooms inside it).
3. Ensuring surface contamination levels and effective dose rate (EFD) below the admissible values according to the Norms of Radiation Protection (NRP) -2004.

The detailed plan for partial dismantling is intended to describe in detail the succession of procedures related to the dismantling of the equipment of the Research reactor IRT-Sofia that is not going to be used for the purposes of its refurbishment into a low-power reactor,

and the operations for reduction of the radioactive waste volume, decontamination, sorting, packaging, temporary storage and transportation for delivery to state company “Radioactive waste”. The main purpose of the partial dismantling planning is to ensure the safety of personnel and population as well as protection of environment.

Taking into consideraton the actual radiation conditions inside the reactor pool, determined by the reactor operational staff, the Plan for partial dismantling describes:

- Dismantling and waste processing activities (characterization, conditioning, decontamination, transport, storage and final disposal);
- Facilities and equipment required to support the dismantling activities according to safety guide [3];
- The decontamination process for internal or external surfaces of components, systems, and instruments. Decontamination will be carried out before and/or after dismantling;
- Special transportation and shield containers for the transportation of the dismantled structure units and elements to the final storage facilities for radioactive waste (RAW) which would ensure safety for the staff, population, and the environment, according to safety guide [4];
- The way how the plant and operations will be monitored during the partial dismantling. Appropriate radiation and dosimetry controls are to be supplied during the course of the above mentioned operations;
- The technological process of dismantling developed taking into account the actual radiation situation in the aluminum tank. These procedures also will be carried out according to appropriate radiation and dosimetry control measures;
- Management organization structure that is appropriate for partial dismantling. The structure used during the operation of the RR is not necessarily optimal for partial dismantling according to safety guide [5];
- Safety assessment;
- Environmental impact assessment;
- Quality assurance program.

**Prior the commencement** of the partial dismantling of the IRT-Sofia activities it was necessary to transport the Spent Nuclear Fuel (SNF) to Russia under the international project RRRFR, financed of US-DOE.

**The following documents have been prepared and revised by IAEA experts:**

- General Plan for partial dismantling of IRT-Sofia equipment, revised by IAEA Expert Mission #1 In February 2004 and IAEA Expert Mission #2 in March 2005. It was submitted in BNRA in October 2005.
- Detailed Plan for partial dismantling of IRT-Sofia equipment, including radiation waste and personnel doses, revised by IAEA Expert Mission #3 in December 2007. It was completed in December 2008.
- Safety Analysis Report for the Partial Dismantling, revised by IAEA Expert Mission #4 in June 2008. The Report was completed in December 2008.

**Dismantling activities:**

The disassembly of reactor systems, removal of the reactor core and replacement of old equipment will not pose any significant problems. Many of these activities are within the scope of what would be termed refurbishment, a common procedure during power upgrading of pool-type reactors. These tasks will be within the capability of the operator’s organization – the Institute for Nuclear Research and Nuclear Energy (INRNE), supported by the contractors when it comes to the provision of tools and skills required for size reduction, handling, and transportation of the wastes. The Plan for partial dismantling identifies the roles, chain of command and responsibilities within the dismantling team, interacts with supporting organizations involved at the INRNE site and subcontractors.

**The refurbishment of the IRT-Sofia** will comprise the following reactor systems: ·

- Core – liable to full scale replacement. The new core loading shall be consistent with the type of the new, converted (low-enriched) fuel;
- All Reactor internal pool equippment;
- Primary cooling system – replacement of the pool liner, piping, pumps, heat exchangers;
- Secondary cooling system – partial replacement of piping and fittings and the cooling open-air pools;
- Horizontal experimental channels – their number shall be reduced from the existing 11 to 7 and a new channel is to be set for boron neutron capture therapy (BNCT);
- Spent fuel storage – replacement of the aluminum reservoir of 12 $m^3$ capacity;
- Electric power supply (EPS) – full scale replacement of cables and equipment;
- Control and Protection System (CPS) – full scale replacement of cables and equipment;
- Radiation Monitoring and Dosimetry System (RMDS) – full scale replacement of measuring lines and equipment; ·
- Heating and Climatic Systems – new systems to be built and installed; ·
- Ventilation Systems – partial reconstruction.

**Inventory and characteristics of radioactive wastes generated by the partial dismantling of IRT**

The radiological characterization was carried out by a combination of neutron activation calculaions and direct measurements. The characterization is essential for developing a Radiation Protection Plan which meets the ALARA principles. The characterization is also a basis for defining the radioactive wastes (RAW) quantities and categories.

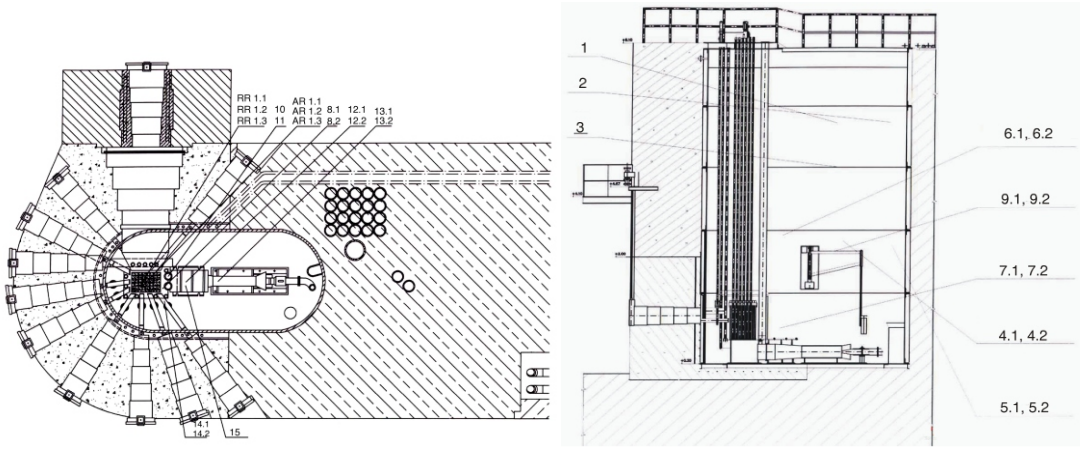


**Figure 1. Places of smears and samples taken**

**Fig ure 2. Smears and samples taking**

**There is the List of RAW streams in IRT-Sofia further down:**

| | | |
|---|---|---|
| S2DED01 | Solid RAW, reactor pool, partial dismantling, RAW storage, metal 2-I and 2-II category | 1680 kg |
| S2DED02 | Solid RAW, reactor pool, partial dismantling, RAW storage, non-metals 2-I and 2-II category | 70 kg |
| S3DED01 | Solid RAW, thermal column, partial dismantling, RAW storage, metal 2-I and 2-II category | 5040 kg |
| S3DED02 | Solid RAW, thermal column, partial dismantling, RAW storage, non-metal 2-I and 2-II category | 10000 kg |
| S4DED01 | Solid RAW, I CL, partial dismantling, RAW storage, metal 1-II and 2-I category | 5800 kg |
| L1DTL01 | Liquid RAW, Reactor hall, partial dismantling, tanks, coolant from reactor pool | 60000 l |
| L1DTL02 | Liquid RAW, Reactor hall, partial dismantling, tanks, decontamination solutions | 6000 l |
| L1DED03 | Liquid RAW, Reactor hall, partial dismantling, RAW storage, ion-exchange resins | 320 l |

**Fig. 3 Expected amounts of RAW**

**Radiation protection of personnel and environment**

The observance of the ALARA principle by the personnel throughout the IRT –Sofia normal operation is based on two basic approaches:

- Detailed preparation of the activities, which will be implemented in the areas with higher radiation contamination;
- Monitoring and control of individual and collective doses during the implementation of the activities and applying corrective activities in case the established levels are exceeded.
- The implementation of these principles is included in the Program for radiation protection during the IRT-Sofia partial dismantling. The program includes:
- The establishment of a safety intervention team that will act during the time of the dismantling;
- A radiation protection organization with obligations, responsibilities and relations with other organizations;
- Restriction of the dose exposure in the building /facility/ and on the reactor operating floor;

- Necessary individual dosimetry control for the personnel ;
- Monitoring for radioactive contamination; control over the radiation situation in the premises and radiation control of the air and the existing ventilation filters and supplementary (local) ventilation systems;
- Monitoring of emissions; control over radioactive wastes and drainage waters, including in possible mobile facilities for low and middle active liquid RAW treatment;
- Control over collection, sorting and categorization, storage and transportation of liquid and solid RAW.
- The limit for the personnel effective dose is 100 mSv during 5 years, and the maximum effective dose for each year must not exceed 50 mSv according to [6];
- For the purposes of the radiation protection and in accordance with the ALARA principle, dose administrative limits are introduced for the personnel in IRT-Sofia.

**During the implementation of the partial dismantling the following working zones are defined in general:**

| working zone | admissible time for the stay in the zone, h | expected collective dose |
|---|---|---|
| On the premises of the 1-st circuit loop | 72 | 4E-04 manSv |
| At the reactor site (the reactot pool is full) | 70 | 6E-05 manSv |
| In the reactor pool (drained) | 100 | 6E-03 manSv |
| In the dismantling of the thermal column externally | 165 | 4E-06 manSv |
| On the site for secondary treatment and decontamination of the dismantled equipment | 82 | 10E-05 manSv |
| Site for carrying out measurements of the obtained materials | 50 | 1E-06 manSv |

**Fig. 4 Working zones**

**In accordance with the ALARA principle the minimization of the dose exposure of the personnel is realized through:**

- Ventilation of workplaces - IRT-Sofia reactor is supplied with a standard ventilation system. For the needs of the partial dismantling it is accepted that its functions are going to provide some radiation protection of the workers. In the activities related to expected large amount of powder release (breaking of concrete, graphite blocks, cutting, etc.) a supplementary local ventilation system is also used, supplied with filters for dust removal;
- Automation and mechanization of processes - the dismantling activities in the reactor pool are going to be carried out mechanically and remotely;
- Radiation control systems - during the activities connected with the IRT-Sofia partial dismantling the radiation control system will continue to provide the necessary individual dosimetry control and control of radiation situation in the premises and radiation control of the air and the filters of working and supplementary (local) ventilation systems envisaged in the plan for partial dismantling; control of radioactive wastes and drainage waters; control over collecting, sorting and categorization, storage and transportation of liquid and solid RAW;
- Using of individual protective means (clothing, breathing masks, etc.).

**On the basis of the prepared documents Detailed Plan for partial dismantling of IRT-Sofia equipment and Safety Analysis Report for the Partial Dismantling and with the assistance of US DOE and IAEA we proceed towards the implementation of the partial dismantling activities shortly.**

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# SILICON DOPING AT FRM II

H. GERSTENBERG, X. LI, I. NEUHAUS
*TU München, Forschungsneutronenquelle Heinz Maier-Leibnitz (FRM II)*
*Lichtenbergstrasse 1, D-85747 Garching - Germany*

## ABSTRACT

FRM II, the 20 MW heavy water moderated research reactor of the Technische Universität München, is equipped with a Si doping facility suitable for the irradiation of ingots up to a maximum diameter of 200 mm and a maximum length of 500 mm. Using the results of test experiments and additional MCNP calculations which had already been performed during the nuclear commissioning of FRM II a Ni absorber surrounding the irradiation position had been shaped suitably to achieve the necessary high homogeneity of the doping profile. The installation of the irradiation channel within the heavy water moderator tank providing a very well thermalized neutron spectrum makes the facility particularly interesting for the doping aiming high target resistivities up to > 1000 Ωcm. Since 2007 the facility is working semiautomatically. The high demand from the industry initiated the implementation of a 2-shift operation mode which allowed the irradiation of more about 10 t of Si in 2008.

## 1. Introduction

The Forschungsneutronenquelle Heinz Maier-Leibnitz (FRM II) is a 20 MW heavy water moderated research reactor being operated since 2005 by the Technische Universität München on its campus in Garching. Due to its very compact core design – only a single cylindrical fuel element containing approximately 8 kg of highly enriched uranium – the reactor is clearly optimized for basic research by means of neutron beam tube experiments. Nonetheless it was evident already since the very beginning of the reactor project that FRM II has also to be made available to medical, industrial and commercial applications. For this purpose the reactor was equipped in addition to the scientific installations with a facility for cancer treatment by neutron irradiation [1] and several irradiation channels for technical applications [2]. Among those the Si doping facility turned out to be the most important and successful one from the commercial point of view.

## 2. Basic layout of the Si doping facility

The neutron transmutation doping (NTD) of the semiconductor Si in a research reactor is based on the nuclear reaction

$$^{30}Si(n,\gamma)^{31}Si \xrightarrow{\beta^-} {}^{31}P$$

It is important to note that no further neutron capture reactions take place in Si and that the half life of $^{31}$Si is only 2.62 h.

The main advantage of NTD as compared to other Si doping techniques is the high achievable accuracy and homogeneity of the doping profile. For commercial purposes the resistivity – the most common measure for the doping effect – has to meet the target resistivity to within ±5 %, and the inhomogeneity of the resistivity along the axis of the ingot has to be less than again ±5 %. NTD Si fulfilling these requirements is used by all major

suppliers for semiconductors as starting material for electronic components processing high electrical power.

From the facts mentioned above it is obvious that the most important requirement for a successful Si doping facility in a research reactor is the provision of an irradiation channel exhibiting a sufficiently homogeneous neutron flux density distribution. In addition at FRM II it was one of the main design criteria to allow the irradiation of Si ingots up to a maximum diameter of 200 mm and a maximum length of 500 mm. Taking into account these aspects the vertical thimble JBE12 BB001 was chosen for housing the doping facility. It is located within the heavy water moderator tank in a distance (center to center) of 1 m from the fuel element; the thimble itself, however, is light water filled to allow easy access from the top.

Already during the nuclear commissioning of FRM II the neutron flux density profile within the thimble JBE12 BB01 was monitored. In order to match the irradiation conditions within the future Si doping facility to the highest extent possible two single crystalline Si ingots (Ø = 150 mm, combined length l = 500 mm) were equipped with a total of 30 Al:Au(2‰) flux monitors and irradiated in a simplified test rig to be handled using the crane in the reactor building. In order to increase the radial homogeneity of the doping profile already in this test experiment the Si ingots were rotated around their central axis at a frequency of 5 turns/min. The result of the test irradiation is shown in figure 1 together with the distribution of the flux monitors within the Si ingots. It turned out that the neutron flux density in the central region of the ingots under irradiation was approximately 12 % higher as compared to the top and bottom faces. The radial inhomogeneity on the other hand was < 3 % and consequently acceptable. The idea for the necessary correction of the neutron flux density along the axis of the Si ingots was to introduce a suitably shaped Ni layer (so called liner) into the Al tube surrounding the irradiation position. Due to the higher neutron absorption cross section of Ni as compared to Al the neutron flux density is reduced in the covered area and the doping profile is homogenized correspondingly. The correct shape of the Ni liner was determined from MCNP calculations using the results of the test experiment as the input [3].

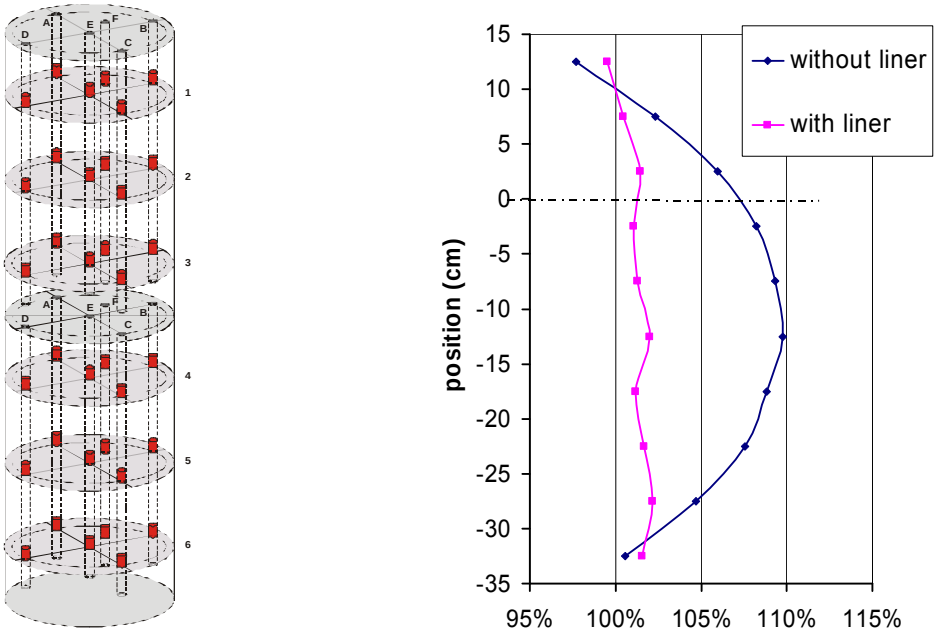


Figure 1: Schematic drawing of two Si ingots equipped with Al:Au(2‰) neutron flux density monitors and effect of the Ni liner on the neutron flux density within the Si ingots.

The resulting neutron flux density within the Si turned out to be 1.7E13 $cm^{-2}s^{-1}$ with an acceptable axial variation (see again figure 1) of < 5 %. Due to the distance of 1 m between fuel element and irradiation position the ratio of thermal and fast neutron flux density was determined to be as high as $\Phi_{th}/\Phi_f \approx 1700$. An important and wishful consequence of this high value is that during irradiation almost no extended, electrically charged defect clusters are generated. For this reason even weakly doped NTD Si with target resistivities up to $\rho > 1000$ Ωcm are available from FRM II.

In addition to the smoothening of the doping profile by means of the Ni liner and the rotation of the Si ingots during irradiation the vertical shift of the neutron flux density distribution during the 60 days reactor cycle has to be corrected for. For this purpose the entire irradiation rig mounted within the thimble JBE12 BB01 can be lifted by up to 150 mm. Due to the comparatively large distance between fuel element and irradiation position, however, this effect is only weak and an adjustment of only 25 mm during the entire cycle is sufficient to guarantee acceptable irradiation results. The effect from a change in shape of the neutron flux density profile with increasing burn-up of the fuel element is even smaller and can be neglected completely.

Finally the originally foreseen permanent forced cooling of the irradiation position turned out to be not necessary. Calculations showed that in case of purely passive cooling by pool water the layout temperature of the thimble (80°C) is not violated, although the temperature within the Si ingots increases up to 140 °C during irradiation.

## 3. Operation of the semiautomatic irradiation facility

After determination and optimization of all of the irradiation parameters during the nuclear commissioning and one year of operation using the simplified rig mentioned above the final semiautomatic facility was installed and taken into operation in 2007. The device was built in cooperation with the Hans Wälischmiller GmbH, Markdorf. The most important component of the facility is a lifting unit which is mounted on the movable bridge spanning over the reactor pool of the FRM II. The entire load chain of the doping facility is in accordance with the German KTA regulations.

For launching an irradiation run typically 2 or 3 ingots of equal diameter and target resistivity are put together into a so-called loading basket to form an irradiation batch and lowered into the handling position of the reactor pool approximately 4 m below the surface level. The same area - which offers space for a total of 12 Si batches - is used later for the radioactive cooling of the freshly irradiated ingots. In the next step the Si ingots are inserted into the irradiation basket by means of a vacuum lifter. The further irradiation process is controlled automatically. After the input of the irradiation parameters into the computer for operation and monitoring of the doping facility the lifting unit takes the irradiation basket and moves it along a predefined path into the irradiation channel (see figure 2). After the Si has reached the final irradiation position it is rotated automatically around the ingot's central axis at a low frequency of approximately 5-7 turns/min. After completion of the irradiation the Si is lifted again automatically by about 4 m into a decay position within the thimble. The entire setup stays in this position for at least 1 h to allow the short lived activities from the structural materials – mainly AlMg3 from the irradiation basket and its handling device – to decay to a value which allows working on the movable bridge at an acceptable dose rate of 30 µSv/h in maximum. Subsequently the freshly irradiated Si is moved back to the handling area in the storage pool using the same path as before. After disconnecting the irradiation basket from the lifting unit the latter is available for the next irradiation run.

During irradiation the reactor power and the frequency of rotation is monitored permanently by the computer program contrloing the doping facility. In case one of these parameters drops accidentally below a limiting value defined in the software the irradiation is interrupted and the irradiation basket is lifted into the decay position within the thimble.

Figure 2: The Si doping facility is approaching the irradiation channel

In order to allow the decay of the induced $^{31}$Si activity the irradiated Si ingots are left for 2 days in the decay area of the FRM II's storage pool. Subsequently they are taken out using again the vacuum equipment for moving them from the irradiation basket into the loading basket. Upon taking them out of the reactor pool the ingots are rinsed with fresh deionized water and finally cleaned in an ultrasonic bath.

Before shipment back to the customer the Si is released according to the German radioprotection regulations. The limiting values to be met are a surface contamination of < 0.5 Bq/cm² and a specific activity of < 0.09 Bq/g. It is noteworthy that in general the mentioned limits are kept without problems, a fact which impressively demonstrates the purity of the starting material.

Already in 2007, the first year of operation of the semiautomatic Si doping facility, a total 3.8 t of Si was processed in 180 separate irradiations. Since the industrial demand turned out to be continuously high these numbers were increased in 2008 to 9.8 t and 526 single irradiations. In order to satisfy this demand a two shift system for Si doping was implemented at FRM II in June 2008.

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# IAEA PUBLICATION: TECDOC-1601 – HOMOGENEOUS AQUEOUS SOLUTION NUCLEAR REACTORS FOR THE PRODUCTION OF $^{99}$MO AND OTHER SHORT LIVED RADIOISTOTOPES

E. BRADLEY, P. ADELFANG
*Division of Nuclear Fuel Cycle and Waste Technology, International Atomic Energy Agency*
*Wagramer Strasse 5, A-1400 Vienna – Austria*

N. RAMAMOORTHY
*Division of Physical and Chemical Sciences, International Atomic Energy Agency*
*Wagramer Strasse 5, A-1400 Vienna – Austria*

## ABSTRACT

The concept of a compact homogeneous aqueous reactor fuelled by a uranium salt solution with off-line separation of radioisotopes of interest from aliquots of irradiated fuel solution has been cited in a few presentations in the series of International Conference on Isotopes (ICI) held in Vancouver (2000), Cape Town (2003) and Brussels (2005) and recently some corporate interest has also been noticeable. Calculations and some experimental research have shown that the use of aqueous homogeneous reactors (AHRs) could be an efficient technology for fission radioisotope production, having some prospective advantages compared with traditional technology based on the use of solid uranium targets irradiated in research reactors. A review of AHR status and prospects by a team of experts engaged in the field of homogeneous reactors and radioisotope producers yielded an objective evaluation of the technological challenges and other relevant implications. The meeting to develop the IAEA-TECDOC facilitated the exchange of information on the 'state of the art' of the technology related to homogeneous aqueous solution nuclear reactors, especially in connection with the production of radioisotopes.

This paper presents a summary of the recently issued IAEA-TECDOC and related IAEA activities since the publication.

## 1. Introduction

The use of aqueous homogeneous reactors (AHRs), also called solution reactors, for the production of fission-based medical isotopes is potentially advantageous because of their relatively lower cost; small critical mass; inherent passive safety; and simplified fuel handling, processing and purification characteristics. These advantages stem partly from the fluid nature of the fuel and partly from the homogeneous mixture of the fuel and moderator in that an AHR combines the attributes of liquid-fuel heterogeneous reactors with those of water-moderated heterogeneous reactors. If practical methods for handling a radioactive aqueous fuel system are implemented, the inherent simplicity of this type of reactor should result in considerable economic gains in the production of fission-based medical isotopes. In June 2007, the IAEA convened a meeting of 10 technical experts from 7 institutions in 5 countries to review all the relevant issues and make recommendations for future work. The details of this meeting have been published in an IAEA TECDOC. This paper presents a summary of that report and IAEA activities since the publication.

## 2. Advantages of homogeneous aqueous reactors for the production of fission-based medical isotopes

### 2.1 Reactor design flexibility and inherent nuclear safety characteristics

The flexibility of solution reactor design parameters is an important feature of the AHR concept that allows customized design configurations to satisfy safety requirements and meet or exceed isotope-production targets. The greater flexibility afforded by solution reactors with respect to core operating power range is an important advantage with respect to $^{99}$Mo production demand. Solution reactors for isotope production could range from 50 to 300 kW.

The choice of fuel base and solution composition is contingent on core design, operating and product isotope processing strategy. Traditionally, uranyl-sulfate fuel was preferred over uranyl-nitrate because of its greater radiation stability. However, the distribution coefficient for $^{99}$Mo extraction is higher from irradiated uranyl-nitrate solutions than from irradiated uranyl-sulfate solutions; consequently a nitrate fuel base is clearly more advantageous from a processing yield point of view. The fuel concentration is selected to minimize core volume/fissile mass, optimize processing efficiency, or both. Solution reactors are typically operated at 80°C and slightly below atmospheric pressure. The low operating fuel-solution temperature, power density, and pressure provides thermodynamic stability, minimizes potential safety risks and yet allow for sufficient flexibility to optimize $^{99}$Mo production demands.

The inherent nuclear-safety characteristics of solution reactors are associated with the large negative density coefficient of reactivity in such systems. The reactivity effect resulting from the operation of solution reactors at power may be thought of as the superposition of two effects, namely: (1) an overall uniform volumetric expansion of the fuel solution due to the increase in fuel temperature and the formation of gas bubbles due to radiolysis; and (2) a corresponding density redistribution within the expanding volume in which the introduction of an equivalent void volume displaces fuel from regions of higher reactivity worth to regions of lower reactivity worth. The resulting density reduction is manifested in a large negative coefficient of reactivity which provides a self-limiting mechanism to terminate a reactivity excursion and provides inherent nuclear safety. Relevant experiments in the French CRAC and SILENE facilities have demonstrated these phenomena.

## 2.2 Efficient neutron utilization, elimination of targets, less post-processing uranium generated per curie of $^{99}$Mo produced, and overall simpler waste management

A unique feature of using the solution reactor for fission-based medical-isotope production compared to conventional production is that the reactor fuel and target are one, consequently a solution reactor can produce the same amount of $^{99}$Mo at 1/100th the power consumption and waste generation. Thus the potential advantages of utilizing solution reactor technology are lower reactor power, less waste heat, and a reduction by a factor of about 100 in the generation of spent fuel when compared with $^{99}$Mo production by target irradiation in heterogeneous reactors.

When one considers waste management in terms of both spent-reactor-fuel and spent-target disposition, waste management for the solution reactor is far simpler. A solution reactor has no need for targets and, therefore all processes related to the fabrication, irradiation, disassembly and dissolution of targets are eliminated. Because these target-related processes result in the generation of both chemical and radioactive wastes, $^{99}$Mo production in solution reactors can significantly reduce waste generation. Since the recovery and purification of $^{99}$Mo from conventional targets after dissolution will be quite similar (if not identical) to that of a solution reactor, the solid and liquid wastes produced will be similar, except for uranium disposition. Uranium from the solution reactor is recycled and only disposed at the end of the fuel solution's viability (up to twenty years).

## 2.3 Efficient processing of other isotopes using off-gas extraction

In addition to $^{99}$Mo, other radioisotopes used by the medical community can be processed more efficiently from a solution reactor. Radiolytic boiling enhances the off-gassing of volatile fission products from the fuel solution into the upper gas plenum of the reactor. A number of valuable radioisotopes such as, $^{133}$Xe and $^{131}$I, can be recovered from the off-gas. There is a large demand for $^{131}$I, as it continues to be widely used for therapy of thyroid disorders. Further, higher specific activity achievable in the off-gas recovery makes it much more effective for radiolabelling, compared to traditional uranium target irradiation technology. $^{89}$Sr and $^{90}$Y are two more products of interest for similar recovery due to their proven therapeutic utility and increasing demands, in particular for $^{90}$Y. While the conventional source of $^{90}$Y is from a radioisotope generator housing the long-lived $^{90}$Sr separated from the waste stream of

reprocessing plants, the AHR approach could be a potential new source for direct recovery from irradiated uranium salt solution..

## 2.4 Less capital cost and potential lower operating costs

The core cooling, gas management, and control systems and auxiliary equipment will be relatively small and simple compared to current research reactor target systems due to the lower power of solution reactors. Isotope separation, purification and packaging systems should be very similar to current target system facilities. The relatively smaller, less complex solution reactor will be less costly to design and construct than traditional research type reactors.

Operating costs may be reduced through many of the improvement mechanisms mentioned above. Specifically a target-free process eliminates all related costs, including the costs of target waste handling and disposition. Any resources involved in the transport of the irradiated target to a processing facility will be saved as will product losses due to any intermediate cooling periods. Reactor control and operation is expected to be simpler potentially resulting in reduced staffing requirements.

# 3. Design Challenges

Although AHR technology is well characterized in the research environment, the capability of a solution reactor to perform a medical-isotope production mission in a long-term continuous steady-state mode of operation in the 100 to 300 kW range is not guaranteed. Specifically, many technical challenges must be addressed in transitioning the technology to a commercial industrial environment.

## 3.1 Isotope separation technology

Solution reactor operation for medical isotope production could be challenged by the chemical stability of the fuel solution induced by a high radiation environment without introducing new undesired complex chemical structures in the product isotope and/or chemical reactions with the solution being processed. Furthermore, the potential problems caused by the build-up of adsorption and fission products, their effect on reactor operation, and the subsequent recovery system is another challenge which must be addressed. In addition, the effects of build-up of corrosion products, materials leached from the recovery system, and chemical additions must also be analysed and optimized. If the fission product build-up and/or corrosion product effects are important, a means to clean up the fuel solution in concert with waste-management and economic considerations must be devised.

Another important effect that has not been fully characterized is the effect of molybdenum redox chemistry of high radiation fields that will accompany fuel cooled for less time than current practices. Because recovery is based on maintaining Mo in the (VI) oxidation state, its reduction to lower oxidation states would diminish both its sorption in the loading phase and it's stripping from the column in alkaline solution, where the lower oxidation-state Mo species precipitate in the column as hydrous metal oxides. Limited studies have shown that four hours after irradiation, effects are seen by lowering of $^{99}$Mo distribution ratios, especially in sulfate media. Much more experimental work is required to understand and design for this effect.

## 3.2 Design optimisation

Several design parameters must be optimised during any specific design process. Two fuel solutions are currently being considered for solution reactors dedicated to radioisotope production, namely, uranyl-sulfate and uranyl-nitrate. As described above, sulfates facilate easier reactor operation while nitrates tend to optimise $^{99}$Mo recovery. Also the selected uranium concentration in the fuel solution is a compromise between reactor optimization and $^{99}$Mo separation efficiency. A lower uranium salt concentration in the fuel solution results in a larger Kd for Mo(VI) and therefore a more effective and efficient recovery of $^{99}$Mo. As a result, the size of the recovery column can be smaller making washing of impurities more effective and obtaining a more concentrated product solution of the raw molybdenum from the column. However, a higher concentration of uranium in the solution will minimize the reactor fuel solution volume leading to a more compact reactor.

## 3.3 Increasing power beyond current operating experience

Historically, solution reactors have been used either in a research capacity to: (1) study nuclear kinetics phenomena associated with nuclear excursions; (2) as a neutron generator to study the effects of irradiation on materials; or (3) to generate radioisotopes. As a result, most reactor operations were transient in nature, or limited with respect to steady-state operation. Physically, the radiolysis gas and vapour that form at high power densities create bubbles that migrate to the surface of the solution. The resulting perturbations at the liquid surface may cause reactivity variations, as well as waves and sloshing effects making it difficult for the automatic rod control system to maintain steady state power conditions. These phenomena are closely related to power density and need to be examined carefully to avoid potential power instabilities or uncontrolled power transients. The design of the core tank may also need to be reconsidered. These instabilities, while detrimental to predictable production operations, pose a relatively small potential hazard provided the reactor vessel design can accommodate pressure transients due to liquid perturbations. The use of Low Enriched Uranium (LEU) fuel requires a greater volume of fuel and thus results in an increase in core solution height which potentially diminishes the reactivity variations induced by perturbation of the solution surface. Furthermore, a non-cylindrical core tank design would probably attenuate the instability phenomena, thus further strengthening safety.

## 3.4 Licensing solution reactors

Since no operating license applications involving solution reactor facilities for isotope production have been submitted, world-wide nuclear regulatory bodies have not developed specific, relevant regulations. Hazard analyses for solution reactors have indicated significantly lower hazard to workers, surrounding populations and the environment than those reactors currently addressed by regulatory bodies. New regulations appropriately addressing specific hazards associated with solution reactors for commercial isotope production will be necessary. Until these regulations are formulated and issued, it may be feasible to address these facilities in a manner similar to current research reactor standards with appropriate modifications as needed.

# 4. Status of solution reactors for fission-based medical isotope production

Medical Isotope Production Reactors are under development in China, Russia and the United States. Two fundamental technologies have been patented in the US, Europe and Russia. These reactors use LEU solutions of a) uranyl-nitrate salt and b) uranyl-sulfate salt as the fuel. The ARGUS reactor, a 20 kW(th), High Enriched Uranium (HEU) solution reactor has been operated as an experimental development activity by Kurchatov Institute in Russia. Irradiated solution from this unit was processed to separate and purify $^{99}$Mo to European and US pharmacopoeia standards. It should be noted that meeting minimum pharmacopoeia purity requirements alone may not be sufficient for specific formulations used in the eventual medical imaging procedure.

In January 2009 Babcock & Wilcox Technical Services Group, Inc. (B&W TSG) signed an agreement with Covidien to develop technology for the manufacture of $^{99}$Mo. The program has the potential to supply more than 50 percent of U.S. demand for Mo-99. Under the agreement, B&W TSG and Mallinckrodt Inc., a subsidiary of Covidien, will collaborate on the development of solution-based reactor technology for medical isotope production [1].

# 5. Recent IAEA activities

During 2008 a CRP titled “Feasibility Evaluation of the Use of Low Enriched Uranium Fuelled Homogeneous Aqueous Solution Nuclear Reactors for the Production of Short Lived Fission Product Isotopes” was initiated and approved internally within the IAEA. As suggested by the title, the aim of this CRP is to evaluate the use of LEU fuel in solution reactors via the analysis of normal and accident conditions related to the operation of solution reactors.

At the end of the CRP, it is expected that all participants will have acquired an appropriate understanding of the feasibility of using LEU in AHRs and the potential of these reactors to produce short lived fission product isotopes. Specifically the CRP has the following objectives:

1. Study the technical feasibility of using LEU in AHRs;
2. Evaluate the impact of the use of LEU fuel on the design and operation of projected ARHs;
3. Carry out benchmarking and comparison exercises using available tools for modelling AHRs;
4. Evaluate feasibility of production of short lived fission product isotopes using aqueous LEU fuel solution in AHRs;
5. Study relevant isotope separation technologies applicable to LEU fuelled AHRs; and
6. Study relevant waste management challenges associated with LEU fuelled AHRs.

Participants are being sought from interested member states. Currently the IAEA has received proposals for contracts from China and Pakistan and expressions of interest for agreements from the USA, France and Japan. The first research coordination meeting is planned for 2009.

A few paragraphs of text on the problems faced in $^{99}$Mo supplies during 2008 and the need for enhanced international efforts were included in the draft 2009 Nuclear Technology Report (NTR) of the IAEA submitted to the Board of Governors Meeting held in the first week of March 2009. While responding, the Member States strongly urged the IAEA Secretariat to undertake all necessary measures to support the reliable availability of adequate supplies of $^{99}$Mo to all users across the world. In a number of other events held in 2008-2009, similar calls are there seeking greater international cooperation in planning strategies to meet the short, medium and long term demands for $^{99}$Mo. The AHR is likely to remain an option for the long-term in this connection.

## 6. Conclusion

The current technology level is well established within the performed research tests. The next step is to confirm that this new technology can be used in a day-to-day reliable production environment. Continuing, active participation by both pharmaceutical and commercial nuclear reactor industries will be necessary in order to successfully develop viable commercial applications of this technology. While the advantages are numerous, commercial markets must be involved in the establishment of an evolving technology in place of an existing well developed alternative.

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# A COATING TO PROTECT SPENT ALUMINIUM-CLAD RESEARCH REACTOR FUEL ASSEMBLIES DURING EXTENDED WET STORAGE

S.M.C.FERNANDES, O.V.CORREA, J.A.DE SOUZA, L.V.RAMANATHAN,
*Materials Science and Technology Center, Instituto de Pesquisas Energéticas e Nucleares- IPEN*
*Av. Prof. Lineu Prestes 2242, Cidade Universitaria*
*05508-000 São Paulo. Brazil.*

**ABSTRACT**

Pitting corrosion of aluminium (Al) alloy clad research reactor (RR) fuel in wet storage facilities can be reduced to a large extent by maintaining water parameters within specified limits. However, factors like bimetallic contact, settled solids and synergistic effects of many storage basin water parameters provoke cladding corrosion. Increase in corrosion resistance of spent Al-clad RR fuels can be achieved through the use of conversion coatings. This paper presents: (a) details about the formation of cerium dioxide as a conversion coating on Al alloys used as RR fuel cladding; (b) the corrosion resistance of cerium dioxide coated Al alloy specimens exposed to NaCl solutions. Marked improvements in corrosion resistance of cerium dioxide coated Al specimens were observed. This paper also presents details of a Latin American Project to develop conversion coatings for long term safe wet storage of spent Al-clad RR spent fuel assemblies.

## 1. Introduction

According to the IAEA´s Research Reactor Spent Fuel Data Base, there are about 62,000 spent fuel assemblies (SFA) stored in facilities around the world. [1] Most of the research reactor (RR) fuels are clad with relatively pure aluminium or an aluminium alloy. The main form of degradation of aluminium alloy clad RR fuel is that caused by corrosion. Pitting is the main form of corrosion and could lead to breach of the cladding and release of fissile material to the environment and contamination of the storage facilities as well as other fuels in the storage basin. The corrosion of spent RR fuel cladding can be reduced to a large extent by maintaining the storage pool or basin water parameters within specified limits. Work carried out within the context of two IAEA coordinated research projects on the “Corrosion of Al-clad spent RR fuel in water”, and within the corrosion activities of the IAEA supported Regional Project for Latin America on “Management of RR spent nuclear fuel” revealed that in spite of such stringent water parameters, factors such as bimetallic contact, settled solids and synergism between the effects of many of the basin water parameters result in corrosion of Al and its alloys. [2, 3]

Among the many forms of controlling corrosion of metals in general, the use of inhibitors or the application of conversion coatings are widely known and these are extensively used in a variety of industries. These methods of controlling corrosion were considered very briefly in the early 50’s to protect fuel cladding prior to use in the reactor and subsequently discarded. During the last 60 years, significant progress has been made and a wide range of new inhibitors and conversion coatings are available to protect Al surfaces. The use of inhibitors or conversion coatings to protect spent fuel surfaces has never been considered. Many facilities around the world store spent fuels in water of less than desirable quality and many instances of cladding failure have been reported. It is imperative that some form of corrosion

protection be given to stored spent RR fuel, primarily from the safety standpoint. A corrosion protected spent RR fuel could be stored for extended periods in waters of less than optimum quality. This in turn would reduce cost of water quality maintenance. During the last two decades, rare earth compounds have been investigated to develop corrosion protection systems for aluminium alloys as an alternative to chromates, which need to be replaced because of their toxic nature. [4-6]. Based on the formation of cerium hydroxide films on Al alloys immersed in solutions containing cerium compounds as inhibitors, other treatments have been proposed to develop rare earth based conversion coating on Al alloys. [7-12]

This paper presents the results of an exploratory investigation carried out to prepare cerium based conversion coatings on Al alloys used as RR fuel cladding material, namely AA 1100 and AA 6061. The effects of chemical pretreatments and processing parameters on cerium dioxide coating characteristics were determined. The corrosion resistance of cerium dioxide coated Al alloy specimens in NaCl solutions of varying concentrations was also determined. This paper also presents details of a Latin American Regional Project to develop coatings for long term safe wet storage of RR spent fuel assemblies.

## 2. Methods and materials

Specimens 2 x 2 x 0.2 cm were cut from AA 1100 and AA 6061 (Table 1) sheets prepared using standard procedures used for making RR fuel plates. These specimens were oxidized at 300° C in air for 4 h to form a surface oxide layer, simulating thus the surface conditions of a spent fuel plate. Table 2 shows the chemical composition of the solutions used in this study and also the treatment conditions to form cerium dioxide conversion coatings on the specimen surfaces.

Table 1. Chemical composition of aluminium alloys (wt%)

| Alloy | Cu | Mg | Mn | Si | Fe | Ti | Zn | Cr |
|---|---|---|---|---|---|---|---|---|
| AA 1100 | 0.16 | <0.1 | 0.05 | 0.16 | 0.48 | 0.005 | 0.03 | 0.005 |
| AA 6061 | 0.25 | 0.94 | 0.12 | 0.65 | 0.24 | 0.04 | 0.03 | 0.04 |

Table 2. Composition of solution and treatment conditions

| Treatment conditions | Solution | | |
|---|---|---|---|
| | 1 | 2 | 3 |
| Composition | 0.5 M $H_2SO_4$<br>1.28 M $HNO_3$<br>0.05 M $Ce(SO_4)_2$<br>0.04 M HF<br>0.1 M $(NH_4)_2SO_4$<br>1 litre | 10g/l $CeCl_3.6H_2O$<br>3ml/l100vol% $H_2O_2$<br>1 litre<br>pH 1.6 | 0.035 M $CeCl_3$<br>0.12 M $H_2O_2$<br>1mM Cu (glycinate)<br>1 mM $Ti(O_2)^{2+}$<br>1 litre<br>pH 2.0 |
| Temperature (° C) | 35 | 43 | 45 |
| Duration (minutes) | 10 | 4 | 5 |

The treatment consisted of simple immersion of the specimens in the solutions. Solution-1 was used to pickle and remove the surface oxide. Solutions 2 and 3 were used to form the coating. After treatment, the specimens were rinsed and their surfaces examined. Untreated and treated specimens were exposed to 0.001 M and 0.1 M NaCl solutions for 1 and 4 hours to determine their corrosion resistance. The surfaces of specimens: (a) untreated; (b) after treatment in the Ce containing solutions; (c) after exposure to NaCl solutions were examined in a scanning electron microscope (SEM) coupled to an energy dispersive spectrometer (EDS). The electrochemical behavior of untreated and treated specimens of the two alloys was determined from anodic potentiodynamic polarization measurements carried out with an electrochemical system using a standard 3-electrode arrangement in 0.1 M NaCl.

## 3. Results and discussion

The surfaces of AA 1100 and AA 6061 specimens immersed in Solution-2 revealed non-uniformly distributed $CeO_2$ on the former and preferred formation of $CeO_2$ at intermetallic precipitates (IMP) on AA 6061, as shown in Fig. 1a and in the EDS spectrum in Fig.1b. The AA 1100 and 6061 specimens immersed in Solution-3

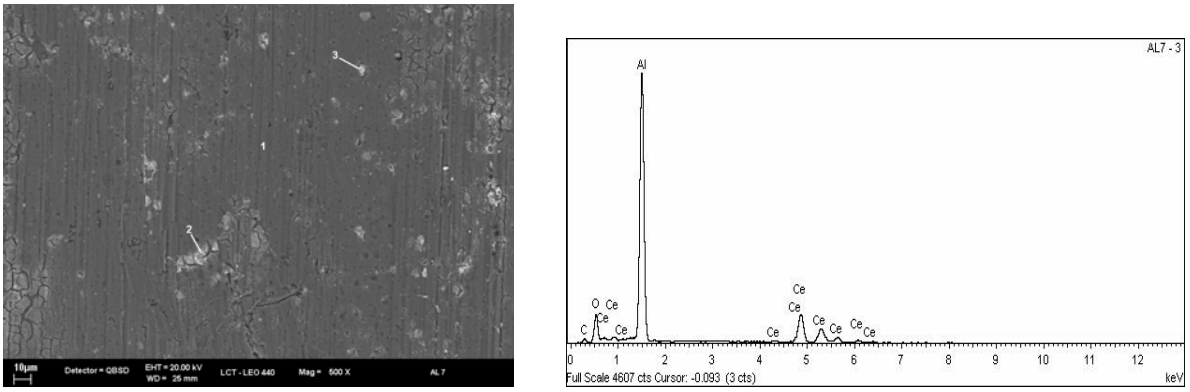


Figure 1. (a) Scanning electron micrograph of AA 6061 surface treated in Solution-2. (b) EDS spectrum of surface at region 3 in Fig. 1a.

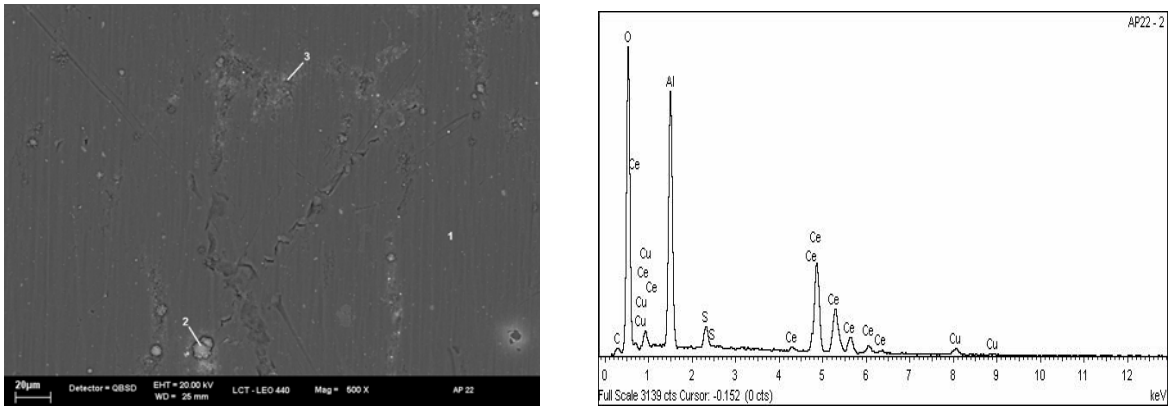


Figure 2. (a) Scanning electron micrograph of AA 6061 surface treated in Solution-3. (b) EDS spectrum of surface at region 2 in Fig. 2a.

revealed $CeO_2$ on the deposited Cu islands. $CeO_2$ was also observed on IMP in AA 6061. Other regions of the surface also revealed $CeO_2$, but to a lesser extent as shown in Fig. 2. The EDS at region 2 in Fig.2a is shown in Fig.2b, revealing Cu and

Ce peaks. The Cu and IMP free regions revealed significantly less $CeO_2$, due probably to the Al oxide on the surface. Specimens that were treated first in Solution-1, to remove surface oxide followed by treatments in Solutions 2 or 3 revealed $CeO_2$ even at regions without Cu or IMP.

The micrographs of $CeO_2$ coated AA 1100 and AA 6061 specimens did not reveal any marked changes upon exposure to NaCl of either concentration and for the two different durations. The results of the electrochemical measurements are summarized in table 3. The cerium dioxide coated specimens had higher corrosion potentials and lower corrosion currents. Specimens coated with $CeO_2$ from Solution-3 were even more corrosion resistant compared to those coated in solution-2. Increase in immersion time in either solution had no significant effect on the potential or the corrosion current.

Overall, these data indicate that cerium dioxide conversion coatings on AA 1100 and AA 6061 clad spent RR fuel assemblies would significantly improve the pitting corrosion resistance of the assemblies in spent fuel basins with significantly lower quality water than that in use presently.

Table 3. Corrosion current ($I_{corr}$) and potential ($E_{corr}$) of the alloys in 0.1M NaCl

| Alloy | Treatment | $I_{corr}$ ( $mA.cm^{-2}$) | $E_{corr}$ ( mV vs SCE) |
|---|---|---|---|
| AA 1100 | None | $5 \times 10^{-6}$ | - 850 |
| | Solution - 2 | $2 \times 10^{-6}$ | - 730 |
| | Solution - 3 | $5 \times 10^{-7}$ | - 660 |
| A 6061 | None | $4 \times 10^{-5}$ | - 770 |
| | Solution - 2 | $4 \times 10^{-6}$ | - 790 |
| | Solution - 3 | $1 \times 10^{-6}$ | - 630 |

## 4. The ARCAL project

A Latin American project titled “Development of novel cost effective conversion coatings for long term safe wet storage of spent aluminium-clad research reactor fuels” has been approved for execution during 2009-2012 and awaiting funding. The participating countries are Argentina, Brazil, Chile and Peru. The different stages of the project include: (a) development of transition metal (TM) and/or lanthanide conversion coatings on Al alloys; (b) evaluation of corrosion resistance of conversion coated mock fuel plates in Latin American spent fuel basins; (c) design and construction of a rig and remote handling equipment to conversion coat spent fuel assemblies; (e) Definition of procedures to prepare conversion coatings on spent fuel assemblies.

## 5. Conclusions

1. Immersion of AA 1100 and AA 6061 specimens in $CeCl_3$ containing solutions resulted in the formation of $CeO_2$ on the surface.
2. The $CeO_2$ on the alloy surface increased the corrosion resistance of the alloys in NaCl solutions.
3. Immersion of the alloys in Cu containing $CeCl_3$ solutions resulted in formation of increased amounts of $CeO_2$, mainly at the Cu islands.
4. The corrosion potentials of $CeO_2$ coated specimens in 0.1M NaCl solution were higher and the corrosion currents lower, indicating higher corrosion resistance, compared to uncoated specimens.

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# European Nuclear Society

Rue Belliard 65
1040 Brussels
Belgium
Telephone +32 2 505 30 54
Fax + 32 2 502 39 02

rrfm2009@euronuclear.org

www.euronuclear.org