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# **TRIGA** Session

# UNITED STATES DOMESTIC RESEARCH REACTOR INFRASTRUCTURE TRIGA REACTOR FUEL SUPPORT

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# Abstract

The purpose of this technical paper is to provide status of the United State domestic Research Reactor Infrastructure (RRI) Program at the Idaho National Laboratory. This paper states the purpose of the program, lists the universities operating TRIGA reactors that are supported by the program, identifies anticipated fresh fuel needs for the reactor facilities, discusses spent fuel activities associated with the program, and addresses successes and planned activities for the program.

# Purpose of Program

The Research Reactor Infrastructure (RRI) Program at the Idaho National Laboratory (INL) supports the United States domestic university research reactor community by providing nuclear reactor fuel to university research and training reactor facilities. The fresh nuclear reactor fuel is provided to U.S. universities at no, or low, cost to the university. The title of the fuel remains with the U.S. government and when the fuel is no longer needed at the university reactor facility, the program provides assistance and reimbursement for safe shipment of irradiated fuel from the universities to DOE used fuel receipt facilities located at the Idaho National Laboratory and Savannah River Site. University-led research and development (R&D) is important in nuclear energy, and the support of research reactors is important for universities if they are to develop and improve the technical competencies necessary to participate in and contribute to advanced Research and Development (R&D) programs.

The program also provides project management, technical support, quality engineering, quality inspection and nuclear material support to the reactor facilities.

Currently the Research Reactor Infrastructure program supports 25 U.S. universities which operate a total of 26 reactors at their reactor facilities. These facilities consist of:

- Twelve TRIGA facilities
- Nine plate fuel facilities
- Two AGN facilities
- One Pulstar fuel facility
- One Critical facility

Table 1 contains information for the TRIGA reactor facilities that are supported by the Research Reactor Infrastructure program.

University Facility	Rated Power	Reactor Type
University of California - Davis	2 Megawatt	TRIGA Mark II
Oregon State University	1 Megawatt	TRIGA Mark II
University of Texas at Austin	1 Megawatt	TRIGA Mark II
Penn State University	1 Megawatt	TRIGA Mark III
Texas A&M University	1 Megawatt	TRIGA Conversion
University of Wisconsin	1 Megawatt	TRIGA Conversion

#### Table 1. TRIGA Facility Details

Washington State University	1 Megawatt	TRIGA Conversion
Kansas State University	250 Kilowatt	TRIGA Mark II
Reed College	250 Kilowatt	TRIGA Mark I
University of California at Irvine	250 Kilowatt	TRIGA Mark I
University of Maryland	250 Kilowatt	TRIGA Conversion
University of Utah	100 Kilowatt	TRIGA Mark I

# **Fresh Fuel**

Several of the TRIGA reactor facilities have recurring or future needs for new fuel. Other universities operate life time cores and have no anticipated needs for fuel delivery in future years unless an unanticipated event occurs (such as leaking fuel elements). These are handled on a case-by-case basis as they occur. The universities that have recurring or future fuel needs include the University of California – Davis, University of Texas at Austin, Penn State University, Texas A&M, Washington State University, Kansas State University, the University of Maryland, and the University of Utah. An analysis was performed in March 2010 to identify what anticipated fuel needs exist for the supported reactor facilities. These needs were classified as short term needs (next five years) and long term needs (beyond five years). Results of the analysis are contained in Table 2.

Table 2. TRIGA Fuel Needs

University Facility	Short Term Fresh Fuel Needs	Long term Fresh Fuel Needs
University of California - Davis	16 Standard Elements     per year	16 Standard Elements     per year
University of Texas at Austin	<ul> <li>3 Standard Fuel Elements</li> <li>3 Instrumented Fuel Elements</li> <li>3 FFCR Fuel Elements</li> </ul>	<ul> <li>100 Standard Fuel Elements</li> </ul>
Penn State University	<ul> <li>4 Standard Fuel Elements</li> <li>1 Fueled Follower Control Rod Fuel Elements</li> </ul>	
Texas A&M University	1 Instrumented Fuel     Element	
Washington State University		68 Standard Fuel Elements
Kansas State University		<ul> <li>20 Standard Fuel Elements</li> <li>5 Instrumented Fuel Elements</li> <li>4 Instrumented Fuel Elements</li> </ul>
University of Maryland	<ul> <li>11 Standard Fuel Elements</li> <li>1 Instrumented Fuel Elements</li> </ul>	<ul> <li>36 Standard Fuel Elements</li> <li>1 Instrumented Fuel Elements</li> </ul>
University of Utah	10 Standard Fuel Elements	

# Spent Fuel

Reactor facilities must ship used fuel when either (1) they can no longer store additional fuel due to licensing and storage limitations, or (2) they must ship used fuel to continue decommissioning activities. The need for used fuel shipment is determined by the university, and this need is submitted to the RRI Program. The DOE receiving facility determines availability for the receipt of the used fuel, and the RRI Program uses this information to develop future funding requests and milestone commitments. In support of these requests, the RRI Program will determine if a government-owned used fuel cask, or a leased cask is required to complete the shipment. Upon funding receipt, the used fuel shipments are planned, scheduled, and executed. If funding is not provided in accordance with the anticipated shipment schedule, then the shipment will be delayed until adequate funding is received.

# Spent Fuel Shipment Casks

Two casks are licensed to ship irradiated TRIGA fuel elements. These include the BEA Research Reactor (BRR) cask (Package ID Number USA/9341/B(U)F-96) and the NAC Legal-Weight Truck (LWT) cask (Package ID Number USA/9225/B(U)F-96).

BEA Research Reactor (BRR) Cask

The Department of Energy has recently procured a new spent fuel shipping cask for shipment of fuel from the university facilities to the DOE Receipt and Storage Facilities. The BRR cask was designed and fabricated by AREVA Federal Services LLC. The cask is currently licensed for the shipment of four fuel types. These include TRIGA, MURR, MIT and ATR fuels (see Figure 1).

Within the cask, the fuel is contained in basket structures specifically designed for each fuel type, and that provide for optimum heat rejection and criticality control. The packaging consists of the payload basket, a lead-shielded cask body, an upper shield plug, a closure lid, and an upper and a lower impact limiter. The cask is of conventional design and utilizes ASTM Type 304 stainless steel as its primary structural material.

The BRR cask may be used in a pool or hot cell environment. The cask body is provided with a drain port, and is intended for use with a vacuum drying system to ensure that water is not present during transport. The cask is designed to be transported singly, with its longitudinal axis vertical, by highway truck or by rail in Figure 1. BRR Spent Fuel Cask exclusive use. When loaded and prepared for transport,



the BRR package is 119.5 inches long, 78 inches in diameter (over the impact limiters), and weighs 32,000 lbs.

A total of nineteen TRIGA fuel elements can be shipped per BRR cask load.

NAC Legal-Weight Truck (LWT) Cask

The NAC LWT cask is owned by NAC International and is available for lease for fuel shipments from university facilities (see Figure 2). The NAC LWT is a steel-encased, leadshielded shipping cask. The overall dimensions of the package, with impact limiters, are 232 inches long by 65 inches in diameter. The cask body is approximately 200 inches in length and 44 inches in diameter. The cask cavity is 178 inches long and 13.4 inches in diameter. The volume of the cavity is approximately 14.5 cubic feet.

The cask body consists of a 0.75-inch-thick stainless steel inner shell, a 5.75-inch-thick lead gamma shield, a 1.2-inch-thick stainless steel outer shell, and a neutron shield tank. The inner and outer shells are welded to a 4-inch-thick stainless steel bottom end forging. The cask bottom consists of a 3-inch-thick, 20.75-inch-diameter lead disk enclosed by a 3.5-inch-thick stainless steel plate and bottom end forging. The cask lid is 11.3-inch-thick stainless steel stepped design, secured to a 14.25-inch-thick ring forging with twelve 1-inch diameter bolts. The cask seal is a metallic O-ring. A second Teflon O-ring and a test port are provided to leak test the seal. Other penetrations in the cask cavity include the fill and drain ports, which are sealed with port covers and O-rings.

The weight of the NAC LWT is 52,000 pounds and the maximum weight of the contents and basket is 4,000 pounds.

The NAC LWT is able to ship up to 120 TRIGA fuel elements per shipment load.

#### **Challenges of Shipping Spent Fuel**

For many of the university reactor facilities, the shipment of spent fuel is a non-routine activity. There are several challenges that have been faced by facilities that have recently shipped fuel. Some of these challenges include:

- The university had not shipped fuel since 1968 and as such, they had no present procedures for shipping spent fuel.
- Floor loading rate was unknown.
- Many interferences had to be removed to allow direct access to the reactor tank.
- Floor space in the reactor cell was very limited.
- Pavement ended inside the reactor facility fence. Some of the surface was not finished. The truck approach was narrow, curving and downhill. A truck large enough to transport the cask could not pull into the lot and then back out (Nearly impossible / refused by drivers).



Figure 2. NAC LWT Cask

- A large capacity (100 ton), long boom crane had to be used due to loading dock obstructions.
- Reactor pool surface area was small (the cask could not be lowered into the pool) and entrance into the facility was limited.
- Facility entrance door was 7 feet tall by 5 feet wide.
- Reactor facility crane limit was 1 ton.

# RRI Program Successes

The RRI program has experienced many successes associated with the TRIGA reactor facilities over the past couple of years. These successes include:

- Four university TRIGA reactor facilities have been converted from HEU to LEU fuel cores. Conversion activities were managed by the U.S. RERTR program.
- All HEU spent fuel has been removed from all university TRIGA reactor facilities and is being stored in the DOE receipt facilities
- Thirty eight TRIGA fuel elements have been procured for four of the reactor facilities.
- All reactor fuel has been removed from the University of Arizona as part of their decommissioning activities.
- The aluminum clad core at Reed College has been replaced with the stainless steel clad core from the University of Arizona.
- The new BRR spent fuel cask has been licensed and fabricated that includes TRIGA fuel as a payload type.

• The program assisted four reactor facilities with core modeling to address information requests received from the Nuclear Regulatory Commission during reactor relicensing reviews.

# Planned Activities

The RRI program has the following activities planned:

- Identify a strategy to respond to future fuel needs in the event that the TRIGA International fuel fabrication facility closes.
- Design and fabricate a new dry transfer system for the BRR cask.

# REAL-TIME MONITORING OF NEUTRON CAPTURE CROSS SECTION IN THE IPR-R1 TRIGA RESEARCH REACTOR AS A FUEL TEMPERATURE FUNCTION

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# ABSTRACT

Nuclear reactor operators have to monitor the behaviour of different nuclear and design parameters that vary in time to ensure the operating safety of the reactor. In recent years several operating parameters for the IPR-R1 TRIGA research reactor were monitored and indicated in real-time by the data acquisition system developed for the reactor, with all the data being stored in a hard disk in the data acquisition computer, to build in this way a database. The goal of this work is to insert in the set of parameters already collected the neutron capture cross sections for the fuel, from the power and temperature numbers obtained in real-time. The experimental data was obtained by using a fuel element instrumented with temperature sensors, located in the core of the IPR-R1 TRIGA research reactor at the CDTN - Centre for Development of Nuclear. This information is useful for the continuous monitoring of the reaction rate in neutron capture. For that, a new analytical formulation is used for the Doppler broadening function proposed by Palma and Martinez which is free from special functions in its functional form and with easy computing implementation. The results obtained were satisfactory from the standpoint of accuracy in comparison with the numerical reference method and indicate that it is possible to carry out real-time monitoring of the neutron capture cross section in the fuel.

#### 1. Introduction

The thermal nuclei movement is well represented in the neutron-nuclei interaction through the Doppler broadening function. Considering a medium thermal equilibrium at temperature T where the target nuclei are in movement and their velocities given by the Maxwell-Boltzmann distribution and the expressions for effective absorption cross sections are obtained from the Briet-Wigner formalism [1],

$$\bar{\sigma}_{\gamma}(E,T) = \sigma_0 \frac{\Gamma_{\gamma}}{\Gamma} \left(\frac{E_0}{E}\right)^{1/2} \psi(x,\xi), \qquad (1)$$

where the Doppler broadening function are described, as in the Bethe and Plackzec approximation,

$$\psi(x,\xi) = \frac{\xi}{2\sqrt{\pi}} \int_{-\infty}^{+\infty} \frac{dy}{1+y^2} \exp\left[-\frac{\xi^2}{4}(x-y)^2\right],$$
(2)

being:

$$x = \frac{2(E - E_0)}{\Gamma}$$
(3)

$$\xi = \frac{\Gamma}{\left(4E_0 kT / A\right)^{1/2}},\tag{4}$$

and all the other parameters are well established in the literature [1].

In this work the absorption cross sections are calculated using the new formulation proposed by Palma e Martinez [2]:

$$\psi(x,\xi) = \frac{\xi\sqrt{\pi}}{2} \exp\left[-\frac{1}{4}\xi^2 \left(x^2 - 1\right)\right] \cos\left(\frac{\xi^2 x}{2}\right) \left\{1 + \operatorname{Re}\left[\eta(x,\xi)\right] + \tan\left(\frac{\xi^2 x}{2}\right) \operatorname{Im}\left[\eta(x,\xi)\right]\right\}, \quad (5)$$

where

$$\operatorname{Re}\left[\eta\left(x,\xi\right)\right] \cong -\operatorname{erf}\left(\frac{\xi}{2}\right) + \exp\left(-\frac{\xi^{2}}{4}\right) \left\{\frac{1}{\pi\xi} \left[\cos\left(\frac{\xi^{2}x}{2}\right) - 1\right] + \frac{2}{\pi} \sum_{n=1}^{n\max} \frac{\exp\left(-n^{2}/4\right)}{n^{2} + \xi^{2}} f_{n}\left(x,\xi\right)\right\}$$
(6)

$$\operatorname{Im}\left[\eta\left(x,\xi\right)\right] \cong \exp\left(-\frac{\xi^{2}}{4}\right) \left\{ \frac{1}{\pi\xi} \sin\left(\frac{\xi^{2}x}{2}\right) + \frac{2}{\pi} \sum_{n=1}^{n\max} \frac{\exp\left(-n^{2}/4\right)}{n^{2} + \xi^{2}} g_{n}\left(x,\xi\right) \right\}$$
(7)

$$f_n(x,\xi) = -\xi + \xi \cosh\left(\frac{n\xi x}{2}\right) \cos\left(\frac{\xi^2 x}{2}\right) - n \sinh\left(\frac{n\xi x}{2}\right) \sin\left(\frac{\xi^2 x}{2}\right) \tag{8}$$

$$g_n(x,\xi) = \xi \cosh\left(\frac{n\xi x}{2}\right) \sin\left(\frac{\xi^2 x}{2}\right) + n \sinh\left(\frac{n\xi x}{2}\right) \cos\left(\frac{\xi^2 x}{2}\right),\tag{9}$$

using temperatures measured with the onboard electronics of the TRIGA IPR-R1 Reactor at the Nuclear Technology Development Centre. The goal of this study is to evaluate whether the time savings in computing time to calculate the Doppler broadening function with the formulation expressed by equation (5) allows the continuous monitoring of the neutron absorption rate as it is already done with other important control variables in the TRIGA IPR-R1 Research Reactor.

#### 2. The IPR-R1 Reactor

The IPR-R1 TRIGA (Instituto de Pesquisas Radiativas - Reactor 1, Training Research Isotope General Atomic) reactor, located at the Nuclear Technology Development Center - CDTN (Belo Horizonte/Brazil), is a typical TRIGA Mark I light-water and open pool type reactor. The fuel elements in the reactor core are cooled by water natural circulation. The heat removal capability of this process is great enough for safety reasons at the current maximum 250 kW power level configuration. However, a heat removal system is provided for

removing heat from the reactor pool water. Figure 1 shows two photographs of the pool and the core with the reactor in operation.



Figure 1. The IPR-R1 TRIGA nuclear reactor

The IPR-1 TRIGA is used for training, research and neutron activation analysis. The reactor core has 58 aluminum-clad fuel elements and 5 stainless steel-clad fuel elements. One of these steel-clad fuel elements is instrumented with three thermocouples along its centerline (Figure 2). This instrumented fuel element was inserted by Mesquita (2005) in the reactor core in order to evaluate the thermal hydraulic performance of the IPR-R1 reactor. The fuel rod has about 3.5 cm diameter, the active length is about 37 cm closed by graphite slugs at the top and bottom ends which act as axial reflector. Figure 3 shows the diagram of the instrumented fuel element.



Figure 2. Instrumented fuel element before and after it was been positioned in the IPR-R1 reactor core.



Figure 3. Diagram of the instrumented fuel element.

The on-line monitoring of fuel temperatures became possible after the data acquisition and processing system implementation and the installation of the instrumented fuel rod in the reactor core (Figure 4). Several neutronic and thermo-hydraulics parameters are now registered. The data acquisition system used in the IPR-R1 reactor consolidates information about the reactor status and provides an on-line or off-line data analysis (Mesquita, 2005).



Figure 4. TRIGA IPR-R1 data acquisition system.

# 3. Results

This section presents the results obtained in the calculation of the absorption cross section, equation (1), for different reactor operation regimes where the temperatures are measured based on the methodology described in Section 2. The graphic in Figure 5 shows the behavior of the  $\xi$  parameter, calculated from equation (3) as the temperature within the nuclear fuel rises.



Figure 5: Behaviour recorded for the  $\xi$  parameter during the operation of the TRIGA – MARK I reactor for the first three <sup>238</sup>U resonances.

Table 1 shows the results for the calculation of the capture cross section for the energy  $E_0 = 6.67 eV$  resonance of <sup>238</sup>U during the operation of the TRIGA – MARK I reactor based on the method proposed for the determination of the Doppler broadening function, equation (5), at different temperatures. The numerical reference method (Gaussian quadrature method) was used in the calculation of the Doppler broadening function directly on equation (2). Table 2 shows the computer processing times for the calculation of the cross sections as a result of the quantity of terms in the expansions of equations (6) and (7).

Time(s)	Temperature (K)	ξ	Х	Reference Method	Proposed Method (n=40)
			0	7178.977	7178.977
[0,561]	298.55	0.4563	10	368.664	368.664
			50	8.732	8.733
			0	6544.393	6544.393
708	377.60	0.4057	10	458.324	458.324
			50	8.760	8.760

Table 1: Calculation of the capturing cross section for the  $E_0 = 6.67$ eV resonance of <sup>238</sup>U during the operation of the TRIGA – MARK I reactor

Table 2: Computer processing time to calculate the capturing cross section for  $E_0 = 6.67 eV$  resonance of <sup>238</sup>U, for T = 298.55K with function  $\psi(x, \xi)$  being calculated from equation

Number of terms	Average computacional time (s)
40	0.015
150	0.094
250	0.156
350	0.265
500	0.406

(5)	
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#### 5. Conclusions

A new analytical formulation was used in this paper based on series for the Doppler broadening function, in order to predict the absorption cross sections in instrumented fuels of the TRIGA – MARK I type. This formulation allows for fast and accurate calculations of the absorption cross section that enable the online monitory, with temperature data from the instrumented fuel. A new window is being developed in the data acquisition system used in the IPR-R1 reactor to calculate these cross sections and, along with information related to the neutrons flux in the region of the fuel and its burnup, to acquire information on the reaction rate for absorption in a real-time and continuous manner.

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# OPERATIONAL EXPERIENCE WITH THE TRIGA MARK II REACTOR OF THE UNIVERSITY OF PAVIA

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#### ABSTRACT

The Laboratory of Applied Nuclear Energy (LENA) is an Interdepartmental Research Centre of the University of Pavia which operates a 250 kW TRIGA Mark II Research Nuclear Reactor, a Cyclotron for the production of radioisotopes and other irradiation facilities. The reactor is in operation since 1965 and many home-made upgrading were realized in the past years in order to assure a continuous operation of the reactor for the future. The annual reactor operational time at nominal power is in the range of 300 - 400 hours depending upon the time schedule of some experiments and research activities. The reactor is mainly used for NAA activities, BNCT research, samples irradiation and training. In specific, few tens of hours of reactor operation per year are dedicated to training courses for University students and for professionals. Besides, the LENA Centre hosts every year more than one thousand high school students in visit. Lately, LENA was certified ISO 9001:2008 for the "operation and maintenance of the reactor" and for the "design and delivery of the irradiation service". Nowadays the reactor shows a good technical state and, at the moment, there are no political or economical reason to consider the reactor shut-down.

#### 1. Introduction.

The Laboratory of Applied Nuclear Energy (LENA) is an Interdepartmental Research Centre of the University of Pavia which operates a 250 kW TRIGA Mark II Research Nuclear Reactor, a Cyclotron for the production of radioisotopes and other sources Irradiation facilities. The reactor, operating since 1965, is at the disposal of researchers from Pavia University and of other users, both public and private, for research activities, training & education and other services. The annual reactor operational time at nominal power is in the range of 300 - 400 hours depending upon the time schedule of some experiments and research activities. The reactor is mainly used for NAA activities, BNCT research, samples irradiation and training. In specific, few tens of hours of reactor operation per year are dedicated to training courses both for University students and for professionals. Besides, the LENA Centre hosts every year a large number of high school students in visit. This particular activity field, aiming to a wide Nuclear culture diffusion, brought to LENA more than 1200 student in 2009 and about 1500 in 2010, with educational/training courses held for more than 400 hours (2009-2010). In October 2010, LENA was certified ISO 9001:2008 for the "Operation and maintenance of the reactor" and for the "Design and delivery of the irradiation service". During the past three years some home-made upgrading were also realized in order to assure a continuous proper operation of the reactor for the future.

#### 2. Status of Main Reactor Systems.

#### 2.1 Instrumentation & Control System

After many years of operation, the Instrumentation and Control (I&C) System of the TRIGA reactor of the University of Pavia was almost completely refurbished in 2007 with home-made new channels (linear & log recorder, percent power channel, fuel and water temperature channel, automatic power demand control, ion chambers power supply), showing, since then, a very good operational behaviour and reliability, without any relevant component failure or need for reworks.

Thanks to the utilization of new and up-to-date electronic components and measurement instruments, it was possible to couple the reactor I&C System to a digital acquisition board for the operational data collection. Data from I&C system are converted into 4-20mA signals and sent to an analogue input PLC board for their acquisition and manipulation. Then data are sent, through TCP/IP, to a remote PC for storage and display

(see Fig.1). The control program, built on LabVIEW platform, allows the acquisition, storage and display of the following parameters:

- Fuel temperature channel #1
- Fuel temperature channel #2
- Water temperature
- Linear power channel
- Log power channel
- Percent Power channel
- Period channel

Data collected are continuously analyzed, resulting very useful for research activities, processes performance monitoring, predictive maintenance, education & training. In particular, the automatic power control system was subject to further parameter adjustments for a more precise power control, using the collected data for the simulation of the reactor dynamic response.



Fig.1- Snapshot of Reactor data acquisition system and data displayed on a chart graph

In order to obtain a more accurate and stable fixture of the ion chambers of the reactor power channels, a micrometric positioning system was home-made designed and realized. Such a system, based on an endless screw transmission mechanism (see Fig.2), fixed above the water surface, allows a precise position adjustment. The result is a better position stability (i.e. avoiding cambers movements due to nuts loosening) along with efficiency improvement in reactor thermal power calibration operations.



Fig. 2 – Ion chamber micrometric-positioning system detail

#### 2.2 Reactor Water-Cooling Circuit

After many years of utilization, in August 2010, the filling-water demineralization system was completely replaced. The new system is a mixed-bed, laboratory-grade demineralizer. In this type of demineralizer, cation and anion resin beds are mixed together, resulting therefore equivalent to a two-step demineralizers in series. In a mixed-bed demineralizer, more impurities are replaced by hydrogen and hydroxyl ions, and the water that is produced is extremely pure. The unit is assembled on a stainless steel frame, with a stainless steel column containing the mixed bed demineralization resins. Along with all the necessary piping installations, an online electronic conductivity indicator was also installed for a better system maintenance. The main features of the demineralizer system are summarized in Tab. 1.

The new system allows a safer and quicker resins replacement avoiding the personnel to deal with resins regenerations and acid/basis solution handling.



Maximum flow rate	Max Pressure	Max. water purity	Electrical consumptio
			n
200 l/h	3 Bar	0.2 μS	50W
	1		

Tab. 1 – New demineralizer specifications

#### Fig<del>.</del> 3 – Water demineralizer system

#### 2.3 Radiation Area Monitoring System

The area radiation monitoring system has been completely renewed in 2007, after almost 30 years of operation. The new system is basically made with a commercial micro-computer and an home-made software developed on Lab-View platform. Since its start-up, the new system showed a very good performance and operational behaviour, without significant system failure during the past three years. Just few small maintenance operations, due to the normal use, have been carried out.

In 2009, after many years of operation, the reactor off-gas radiation monitoring system was completely replaced with a new measurement system based on a NaI scintillation detector. The  $\gamma$ -ray spectrum of reactor gaseous effluents is collected continuously using Gamma-Vision software and data are accessible from remote (i.e. form reactor control room or form reactor emergency room).

Lately, the environmental air particulate monitoring system was completely upgraded and redesigned for a better efficiency and reliability. The network consists of 5 air-sampling stations that suck air on a paper filter for particulate deposition. Four stations are located at 100m from the facility fence, in correspondence of the four cardinal directions; the fifth station sucks air from reactor off-gas on a paired paper/active-carbon filters. Filters are collected and analyzed daily by means of a  $\gamma$ -spectroscopy using a low-background HPGe detector. The system is used for environmental air particulate monitoring on both normal and emergency situation.



Figure 4 - Environmental air particulate monitoring station and air vent (mounted 5m above ground)

#### 2.4 Fuel Elements

At the moment, the core is made of 49 Aluminium clad and 34 SST clad TRIGA fuel elements. All the aluminium elements occupy the outer rings of the core (Ring E and F) while the SST elements occupy the inner rings (Ring B, C and D).

Only 7 fuel elements (all Al clad) have been permanently removed from the reactor tank and are stored in a fuel storage dry pit. Other 7 irradiated fuel elements (6 Al clad and 1 instrumented SST clad) are stored in racks inside the reactor tank. Three fresh fuel elements are in storage.

The fuel elements are controlled periodically for their elongation and/or bowing and until now no elements had to be removed permanently for excess elongation or bowing. At the moment no fuel elements present an elongation greater than 5 mm and only 4 fuel elements present an elongation greater than 4 mm.

#### 2.5 Implementation of Quality Management System

From the end of 2008, in order to continuously improve the safety and quality of reactor management and the accomplishment of all compulsory requirements, LENA decided to implement a Quality Management System (complying with the International Standard ISO 9001:2008) for reactor operation and maintenance activities.

In order to accomplish the implementation of the MS, during the initial processes analysis the path to be followed to build up the system was assessed and defined. The main steps for the system implementation have been:

- Project Team identification;
- budget and schedule development;
- processes analysis;
- Quality Policy definition;
- inventory of existing documentation;
- system implementation (development of documents and their application);
- system assessment (Internal audit).

At the end of the implementation process, the reactor documentation system increased by 45% (percentage of new documents issued / total number of documents), while 10% of existing documents was reviewed.

Furthermore, LENA reactor adopted appropriate methods to monitor and, where applicable, measure MS processes, in order to demonstrate their ability to effectively provide the planned results and to ensure adequate performance in the areas of the MS. One of these methods is the Internal Audit tool. During 2010, the Internal Audit process demonstrated the ability to identify opportunities for improvement: in fact Internal Audits identified 55% of Non Conformance (NC) detected during planned activities of surveillance and test; the Audit process resulted in 5 Corrective Actions (CA) and 5 Preventive Actions (PA).

All Corrective Actions and Preventive Actions were completed in scheduled time.

In addition, a documented procedure was established to define the controls along with the related responsibilities and authorities for dealing with Non Conformance.

During 2010, 95% of detected Non Conformance were handled and closed in the scheduled time; on total number of NC, 63% were classified as *Safety Related* (i.e. NC concerned with aspects that may affect reactor safety operation or health physics); among Safety Related NC, 42% were detected during planned activities of surveillance and test.

At last, the implementation process took about 2 years and eventually led to a third party Audit and to the system certification issued by an independent Certification Body, in October 2010 [1]. As far as financial aspects are concerned, the implementation costs can be groped as follows: 75% staff efforts (54% Project Team, 21% other staff involved, in work-hours), 5% external staff training, 15% (new equipment acquisitions). That means that the implementation process is more a matter of time and staff commitment than organization's financial capabilities.

#### 2.6 Maintenance Program

All components and systems undergo to ordinary maintenance according to the Technical Prescriptions, to Quality Assurance (QA) requirements along with all the pertinent "Good Practice Procedures". The execution of the scheduled controls and of the Preventive (planned) and Corrective (repair) Maintenance of the systems and components is registered in dedicated checklists, in maintenance authorization documents, in maintenance and reactor Log-books and in QA books.

As mentioned above, since 2009, the new QA program implemented at LENA led, through the joint application of maintenance program and QA procedures, to a better and safer reactor operation. In particular,

in order to give evidence of the results of the activities carried out, a series of performance parameters (*indicators*) have been defined and are periodically updated and assessed in order to get even better efficiency and effectiveness from the maintenance service. The following Tab. 2 shows a selection of maintenance performance indicators utilized at LENA. The difference between expected and calculated values can be perceived as an objective for the continual improvement or for maintaining the high standard already reached.

Process	Indicator	Expected value	Calculated value 2010
Instrumentation	Failure probability (number of repairs/instruments number)	<15%	23%
management	N. of Instruments found out of calibration	0	0
	MTBF (Mean time before failure, in days)	140	126
	MTTR (Mean time to repair, in days)	5	3
Maintenance	Outsourcing service dependency		
	$(N^{\circ} of maintenance operation requiring outsourcing$	<15 %	30%
	assistance / $N^{\circ}$ of maintenance operation)		

Table 2 – Maintenance Service performance indicators

#### 2.7 Human resources

As typical of research reactors, the number of personnel units is hardly enough to cover all the tasks / functions in the Organization. Currently, the staff on duty at LENA reactor consists of 16 personnel units and each section / unit of the Organization should usually be made by a head of section/unit and one or more employees. This is made possible by the involvement of staff in more than one task, namely the allocation of multiple responsibilities to the same person, of course with care to avoid any conflicts of interest. The allocation of LENA personnel units according the different functions is reported in the following tables:

LICENCED REACTOR OPERATION AND MANAGEMENT STAFF		
Technical Direction	3	
Reactor Supervisors	6	
Reactor Operators 6		

STAFF QUALIFICATION		
Ph. D.	3	
Master Degree	7	
High School Degree	4	
Middle School	2	

DUTIES		
Administrativ	e	1
Technical:	Top Management (Direction)	2
(15)	Reactor Operation & Management	12
	QA	2
	Radiation Protection Advisor	1
	Health Physics Section	7
	Mechanical Maintenance	2
	Electrical Systems & Instr. Maintenance	2
	Analysis	3
	Marketing & Customer	1
	Educational	1

DEGREE QUALIFICATION SUBJECTS		
Chemistry		1
Physics		5
Engineering		2
Economics		1
STAFF AGE		
<30	1	
<40 7		
<50 5		
<60	2	
>60	1	

#### Table 3: LENA Human Resources data report (2010)

This kind of staff organization, which stems from the need to better manage the limited human resources available, has the positive effect of increasing the skills of staff in the different plant activities, allowing staff

turnover and ensuring, in every time, the reliability of services related to nuclear safety, health protection and operation of the reactor itself.

#### 3. Summary and Outlook.

#### 3.1 Performance Indicators

As already mentioned above, during the QMS implementation process, LENA indentified, for each process involved, a set of *Performance indicators*, i.e. parameters that provide a measurement of processes efficiency and effectiveness. In particular, it is interesting to observe the values obtained for indicators related to LENA reactor operation process (Tab. 4) and how those indicators match with ones indicated in the publication *Optimization of Research Reactor Availability and Reliability: Recommended Practices* (IAEA Nuclear Energy Series No. NP-T-5.4) [2].

Process	Indicator	Expected value	Calculated value
Reactor operation	Reactor availability	70 - 80%	77%
	Reliability	70 - 80%	89%
	Reactor Utilization	>50%	52%
Quality Management System	Number of safety related NC detected	<b>→</b> 0	12
	Number of preventive actions	>1	4
	Auditing system efficiency		
	Number of corrective actions as a result of	>1	5
	an internal audit		

#### Table 4 - Performance indicators values related to reactor operation at LENA

#### 3.2 Operational data

After the major extraordinary maintenance activities described above, the TRIGA reactor of the University of Pavia shows a very good technical state and, at the moment, there are no political or economical reasons to consider a reactor shut-down. The following table 5 reports operational data of the last three years:

	Year 2008	Year 2009	Year 2010
Hours at 250 kW	388,88	321,51	229,91
MWD	4,05	3,34	2,39
Burn-up ( <sup>235</sup> U, g)	3,67	3,52	2,52

#### Table 5 – Operational data of the TRIGA reactor of the University of Pavia

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- [2] Optimization of Research Reactor Availability and Reliability: Recommended Practices IAEA Nuclear Energy Series No. NP-T-5.4

#### Experimental Measurement and MCNP5 Model of the Gamma Dose Distribution in the University of Utah TRIGA Reactor Pool

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#### Abstract

Gamma doses for the University of Utah TRIGA Reactor (UUTR) are estimated based on the MCNP5 model, and by measurements based on the thermo luminescent detectors (TLDs). In the experiment the gamma dose rate above the UUTR core was obtained by placing TLD-400s every 10 cm from the core top up to 150 cm measured from the surface of the UUTR core, and then every 50 cm up to 300 cm. The dose rate was also obtained at the pool surface level using a Ludlum model 19-survey meter. The measurements are obtained at a steady state thermal power of 90 kW. The MCNP5 model closely followed the experimental setup.

The MCNP5 values of the dose immediately above the core were just under 30 percent difference compared to the measured values, which is within the TLD response of  $\pm$ 30 percent. The TLD error is observed to remain fairly constant at around 5% of the mean. As the gamma dose approaches the lower limit for the TLDs (1 rem), the error rises almost exponentially reaching 80%. The difference between the simulated and measured gamma doses increases as the distance from the core increases. This difference at the surface of the UUTR pool is around 90 percent (the experimental gamma dose being 90% higher). The main source of the difference is due to statistical error of the MCNP5 calculation. The larger the size of the geometry, the more computational time is required to reduce the statistical error. Another source of error is the contribution of nitrogen-16 that is generated from (n, p)reaction with oxygen in the reactor pool water. The (n, p) reaction was not considered in our current MCNP5 model. Therefore, the MCNP5 model can be improved as follows: taking into account the fuel burn up that will correlate more accurately dose estimates near the surface of the pool due to the gamma emission from fission fragments, and taking into a consideration a production and transport of nitrogen-16 that will improve the dose estimates especially at the surface of the UUTR core.

Keywords: MCNP5, TRIGA, TLDs, dosimeter, gamma dose, neutron dose

#### **1. INTRODUCTION**

Radiation dose measurements are performed every few years at the University of Utah TRIGA Reactor (UUTR). The UUTR core is placed in a water pool that functions as a coolant for the reactor via natural convection and as a biological radiation shield in protecting personnel. The gamma dose measurements are recorded every time and used to benchmark computational simulations often based on using Monte Carlo N-Particle transport code, MCNP5/X. This UUTR simulation model is therefore verified every time, and it represents a useful tool for estimates of the dose change with the power up-grade, or the core configuration.



The UUTR core contains a mixture of old and new stainless steel and aluminum fuel elements. The fuel elements are arranged in a hexagonal grid. Reflector elements are located around the core perimeter. The UUTR has two different types of reflector, graphite and heavy water.

# 2. GAMMA DOSE EXPERIMENT AND THE UUTR MCNP5 GAMMA DOSE MODEL

The  $\gamma$  dose rate distribution above the core was experimentally determined using dosimeters placed in the pool at certain distances when the reactor was operated at thermal power of 90 kW (that is producing  $7.53 \times 10^{15}$  n/sec). The  $\gamma$  dose was measured for a 10-minute time interval once the reactor reached its steady state operating condition. Thermo-luminescent dosimeters (TLDs) were placed at various distances from the core surface toward the top of the tank. The  $\gamma$  dose rate distribution on the surface of the pool tank was obtained using a Ludlum model-19 survey meter. This instrument uses a sodium iodide scintillator.<sup>1</sup>

The  $\gamma$  dose distribution was calculated based on the UUTR comprehensive MCNP5 model of the UUTR and pool tank; the core thermal power was 90 kW.<sup>2</sup> The  $\gamma$  dose distributions were obtained along the same points where the measurements data were collected, namely from the core surface to the top of the pool. The computational values were then compared to the experimental data.

In order to reduce the MCNP5 simulation time and improve the accuracy, the UUTR pool was represented with 65 sections. The first section contained the core, irradiation facilities, water, and aluminum tank geometry up to 4 cm above the core surface. Subsequent sections were modeled for the next 10 cm increments above the core up to the pool surface. Geometry splitting and the use of cell importance IMP cards improved the variance and decreased the computational time. KCODE calculation of 50 million particle histories was simulated with 500 cycles with 10<sup>5</sup> particles. The dose rate distribution for each segment was obtained by placing a MESH tally (FMESH card) along the geometry divisions. The MESH tally was setup to obtain 2-cm<sup>2</sup> dose resolutions across the top of the core. The dose function DF and the dose energy cards DE were added to the tally in order to obtain normalized dose rate (rem/hr)/(particles/cm<sup>2</sup>-s) as a function of particle energy. The biological dose equivalent rate factors for DF cards were taken from ANSI/ANS-6.1.1-1977 data, Table H.2 from the MCNP5 manual for photons.<sup>3</sup> The dose tally was scaled by using the tally multiplier FM card which multiplied the normalized results by Eq. 1 giving a final dose rate in rem/hr:

$$D = T * \frac{P}{E * Fr * q} \tag{1}$$

D = dose rate (rem/hr)

T = normalized tally output (rem/hr)/(particles/cm<sup>2</sup>-s)

P = power (watts)

E = fission energy deposited in the core (eV/fission)

Fr = fission rate

 $q = 1.602 \text{ x } 10^{-19} \text{ coulomb}$ 

The TLDs used in this experiment were TLD-400s made of CaF<sub>2</sub>:Mn. These are a calcium fluoride crystal doped with manganese and have a wide response range from 1 mrem to  $10^5$  rem. However, these only have a linear response of about 10 percent of that range. The TLDs were read in a Teledyne Brown Engineering System 310 TLD reader. The TLD reader lists the dose in rems and is not calibrated below 1 rem. The resulting effective TLD range is between  $10^5$  rem at saturation and 1 rem. These limitations impeded dose measurement near the surface of the pool with TLDs. A Ludlum model 19 survey meter was used to obtain a dose distribution approximately 10 cm (4 in.) above the pool water. Figure 1 shows the geometry of TLD placement to measure  $\gamma$  dose in the UUTR pool tank.



Fig.1 Locations of TLD as placed in the UUTR pool

The TLDs were placed up to 300 cm measured from the surface of the UUTR core in thetank (Figure 1). A radial dose profile was obtained by placing five TLD packets in a line at 50 cm



measurements were taken 10 cm above the pool water while the UUTR was still operated at 90 kW.

#### **3. EXPERIMENTAL AND MCNP5 RESULTS**

The axial dose rate distribution above the core for measured and simulated results is shown in Figure 2 (MCNP5 simulation and TLD and Ludlum model 19 surface results in comparison).

#### Figure 2. Measured and MCNP5 estimated axial y-dose rates at the UUTR

As shown in Figure 2, the axial dose rates from MCNP5 calculation and measurement using TLD are very similar at the distance of 200 cm from the top of the core. At the distance above 300 cm from the top of the reactor core, the dose rate were not obtained because the TLD 400s cannot read a dose rate less than 1 rem/hr. The MCNP5 results are shown in Figure 2 as well, for the points above 300 cm. The dose rates at 10 cm above the pool water surface from MCNP5

calculation and Ludlum Model-19 detector did not match very well because the MCNP5 result has a large statistical error due to the geometry getting larger. More discussion is provided later on as well.

Each dosimeter packet contained five TLDs in order to achieve a reasonable statistical information on variance and error. The mean and standard deviation  $\sigma$  were calculated for each packet. The  $\sigma$  was observed to remain fairly constant until it approaches the lower limit for the TLDs (1 rem) where  $\sigma$  rises almost exponentially to around 80 percent. The standard response for TLD-400s dominates most of the trend with a response of  $\pm$  30 percent per batch.<sup>4.5</sup>

The MCNP5 results at the surface of the pool water were analyzed by dividing the water surface in several MESH tallies; each MESH tally was assigned an area of 100 cm<sup>2</sup>. The standard deviation  $\sigma$  for these tally samples ranged from around 5 to 35 percent. The 35 percent compares very well to the 30 percent TLD response.

Figure 3 shows the neutron and gamma dose rate from the MCNP5 calculations at the surface of the reactor core. Neutron and  $\gamma$  dose rates, both have their maximum values at the center of the core where it was expected. At the center of the UUTR core, the neutron and  $\gamma$  doses were  $9.7 \times 10^5$  rem/hr and  $2.68 \times 10^5$  rem/hr, respectively. As shown in the Figure 3, both doses decrease lower than  $1 \times 10^5$  rem/hr for  $\gamma$  and  $0.5 \times 10^5$  rem/hr for neutrons, at the edge of the reactor tank.



Figure 3. MCNP5 Neutron and Gamma Dose Distribution at the UUTR Core Surface across the UUTR Pool Water Tank

#### **4. FUTURE WORK**

There are three major areas that need to be addressed in order to create a more accurate UUTR model giving more accurate  $\gamma$  dose estimates. These include better instrumentation, inclusion of the decay  $\gamma$ -rays from long-lived fission fragments, and the production and transport of nitrogen-16 into the MCNP5 model as existing now.

The limited response range of the TLD (>1rem/hr) and the large response of  $\pm 30\%$  contribute to the error of the measurement. Also, different types of TLDs or detectors that can respond to the dose rate lower than 1 rem/hr will be beneficial to achieving more accurate measurements. To create more accurate MCNP5 model, knowledge of fuel elements burnup histories would be beneficial, as well as inclusion of nitrogen-16 generation and transport in the pool tank. Our current MCNP5 model did not include  $\gamma$ -rays that are generated from long-lived fission products. It is not a simple matter to include them as a source in the model. These fission products have different half lives and the distribution and quantity of fission product isotopes must be estimated. The  $\gamma$ -ray energy peaks and relative intensities need to be quantified and then added into the MCNP5 model as  $\gamma$ -ray sources.

The quantity of N-16 produced in and around the UUTR core can be estimated based on neutron flux and neutron capture cross-section of the oxygen-16 present in  $H_2O$ . A convection model needs to be used based on thermodynamic and physical properties of the core and coolant materials. The spatial distribution of nitrogen-16 can then be estimated based on coolant velocity and the 7-second half life of nitrogen-16 before its decay into oxygen-16. This source can then be added to a MCNP5 model.

#### **5. CONCLUSION**

The MCNP5 results correlated satisfactory well with experimental measurements of up to the distance of 300 cm from the top of the core surface. At the surface of the pool tank, the MCNP5 and Ludlum model-19 portable detector values did not match very well. More detailed MCNP5 model is required to obtain more accurate result. Improvements must include better instrumentation capable of measuring the full dose range between the core and pool surface. The tally multiplier FM card value from Eq. 1 can be adjusted to account for the delayed  $\gamma$  from fission product decay. In order to do so, a fuel burn up estimate is needed to quantify the inventory and ratios of fission products present in the fuel rods. The production and subsequent transport of nitrogen-16 before it decays should significantly improve the dose values estimated for the pool surface locations. The natural convection of the pool water is the primary mode of transport. The coolant flow and nitrogen-16 production rates need to be estimated in order to provide a spatial distribution of nitrogen-16 above the core. This may be incorporated into the model to provide more accurate dosimetry estimates in the UUTR pool tank.

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# THE USE OF GASEOUS RADIOACTIVE TRACER FOR THE DETERMINATION OF A PRESET TEMPERATURE IN A HIGH TEMPERATURE INDUSTRIAL OVEN

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#### ABSTRACT

A novel high precision detection method for the determination of the distillation end point of the coking process (usually in the 950-1100 °C range) has been developed, and patented. The system is based on the use of a metallic capsule that melts at a fixed temperature and releases a radioactive gas tracer (<sup>133</sup>Xe). A radioactivity detector, placed in the stack of a coke industrial oven, reveals the presence of <sup>133</sup>Xe in the distillation gas stream, indirectly marking the time in which the preset temperature has been reached in the centre of a coke moving pile, in an industrial high-temperature coke oven. The isotope was prepared by irradiating 1 to 20 cm<sup>3</sup> of natural xenon, contained in a high purity polyethylene vial. The irradiation was performed in the fast pneumatic transfer system facility of the TRIGA MARK II nuclear reactor of the Casaccia Institute, obtaining a total activity of 1-3 MBq of <sup>133</sup>Xe per irradiation. The radioactive gas was transferred inside a steel capsule, furnished with a calibrated membrane, that melted when the preset temperature was reached. A series of tests on a pilot oven confirmed the feasibility of the method on industrial scale. Extension of the coking process appeared also feasible.

#### 1. Introduction

Coke is a solid carbonaceous residue derived from low-ash, low-sulfur bituminous coal, from which the volatile constituents are driven off by baking in an oven without oxygen at temperatures of about 1000 °C, so that the fixed carbon and residual ash are fused together. Metallurgical coke is used as a fuel and as a reducing agent in smelting iron ores in industrial furnaces. Important characteristic of the coke are the test crush index, which convey the strength of coke during transportation into the blast furnaces, and the coke strenght after reaction, that represents coke's ability to withstand the violent conditions inside the blast furnace before turning into fine particles; depending on blast furnaces size, finely crushed coke pieces must not be allowed into the blast furnaces because they would impede gas dynamics. The preparation of coke is usually performed in large furnaces, in which the mass of coal is heated under high temperature in an oxygen deficient atmosphere, with the generation of coke oven gas and various chemical products. Conventional cokemaking is done in a coke oven battery sandwiched between heating walls. The temperature distribution and the elapsed time inside a coke coal bed produces critical effects on the final yield and quality of the product (grain size, volatile coal content, crack sensitivity, hardness, porosity). Above a critical thershold temperature, usually in the 500-700 C° range, the coke shrinks, and the rate of temperature drift and gas evolution  $(H_2)$  raises fast. At this stage the coke quality is critically affected. The reaching of a preset temperature, usually around 1000 C°, marks the coke distillation end point. Actually, the distillation end point is usually determined by the observation of the optical features of the distillation gases (vellow-to violet color change of the burning exhaust gases), or by the direct composition of gas composition or temperature on the oven stack, or chimney. (1-3) In the present paper a method is presented, that can mark the temperature of the distillation end-point into the inner of a mass of moving coal of a furnace for coke preparation. The method is based on the release of a gaseous inert tracer (<sup>133</sup>Xe), contained in a calibrated capsule, at a given temperature in the coke oven stream. The method has been patented, has been tested on an equipped industrial pilot plant, and is aimed to be implemented on the coke industrial production plants, for monitoring and optimizing the process parameters (4).

#### 2. Tracer selection

A null retention on coal mass and plant walls, a high specific activity and a suitable half-life are fundamental properties for a safe and effective use of a gas radiotracer. The neutron irradiation of natural xenon allows an easy detection by Nal(TI) detectors of a gamma emission at the energy of 81 keV from <sup>133</sup>Xe. A spike activity as low as 100 MBq can be safely released into the 200 000 m<sup>3</sup>/h exhaust stream of an industrial blast coke furnace, still allowing the puffing to be monitored with high accuracy. For 133 Xe, a specific actaivity of about 0.25 MBq/cm<sup>3</sup> of natural Xenon can be obtained, by irradiating for six hours a sample by a neutron flux of 2.6 n/cm<sup>2</sup>/s, for 6 hours.



Fig 1. Gamma spectrum of 133Xe by a Nal(Tl) detector. The 81 keV and 31keV peaks belong to  $^{133}$ Xe ( $\beta^-$ , $\gamma$ )  $^{132}$ Cs, and to the X-ray conversion of  $^{132}$ Cs, respectively. The 233 keV peak originates from  $^{133m}$ Xe (IT, $\gamma$ )  $^{133}$ Xe.

# 3. Capsule design

The capsule was machined by starting from a hollow stainless steel cylinder (see Figure 2). In the bottom side a circular V shaped notch was made, to act as a stress raiser. The notch radius and the thickness of the circular metallic membrane so obtained, were calculated in order to to obtain its rupture when a predetermined pressure was reached. To avoid early gas release, the metallic membrane has to be substained and reinforced by a metallic plug, designed to melt at a preset temperature. After the introduction of the radioactive gas into the capsule, the upper metallic lid is welded. The pressure from the gas contained in the inner chamber near the preset temperature should be sufficient to break the metallic membrane, while the metallic plug, pressed outside against it, prevents the membrane rupture. When the local temperature reaches the metallic plug melting point, the inner pressure, no more balanced by the metallic plug, breaks the membrane, and a sudden

gaseous radiotracer puff is released into the furnace. The capsule body and the metallic membrane were made by using an AISI 316 steel composition (high-molybdenum, low carbon), due to the corrosion resistance and soldering easiness of this material. The characteristics of the alloy are reported in Table 1

Mo content	Melting point	Failure load	Elasticity	Yield point,
		kg/mm <sup>2</sup>	module, kg/mm <sup>2</sup>	kg/mm <sup>2</sup>
2,3%	1370-1400 C°	12,6 (at 927 C°)	19.700	21-28 (18 C°)
				4-5 (927 C°)

Tab 1. Relevant propeties of AISI 316 stainless steel



Fig 2. Schematic design of the capsule. The upper lid is arc welded after the introduction of the radiotracer vial.

The plug material melting point must be a neat, sharp, highly reproducible first type transition; pure metals and true intermetallic compounds should be used. A Cu/Mn eutectic alloy (m.p. 1143 C°), and pure copper (m.p. 1083 C°) gave the best results. A membrane thickness of 0.12 to 0.15 mm was machined, and a calculated amount of ammonium carbamate (1 to 5 grams) was added in the capsule. Above 500C° this material is completely transformed into CO2 and NH3, that further raises the inner pressure of the capsule. 10 cm<sup>3</sup> of natural Xenon were irradiated in a high purity polyethylene vial, by using the fast pneumatic transfer system of ENEA Casaccia TRIGA Mark II nuclear reactor. The irradiation flux was always 1.3  $10^{13}$  n/cm<sup>2</sup>/s, while the irradiation times varied from 10 minutes to 2 hours. The irradiated gas was transferred under controlled condistions into a thin Al

container, that was crimped, and put inside the steel capsule, together with the ammonium carbamate. The steel lid of the capsule was arc welded.

## 4. Use in the industrial furnace

The system was repeatedly tested in the industrial pilot coke of CSM (Rome), a furnace equipped with moving walls, temperature, flux and stream gas chemical sensors, designed for the study and optimization of the parameters in coke preparation. A Nal(TI) scintillation detector, connected to a Silena multichannel analyzer, was inserted into a cooling jacket cylindrical container, and faced against the oven stack. The potopeack efficiency, in spite of the shielding cooling apparatus, was considered quite satisfatory. After coke blend loading in the furmace (from 500 to 1000 kg per batch), a capsule containing the radiotracer was inserted into the center of the coal mass, in near proximity of two termocouples, for the calibration and check of temperature response. Some heating run lasting 18 to 24 hours, simulating the industrail process, havr been carried out. As the capsule reached the end point temperature (in our case 1046 ± 30 C°, as resulting from the termocouple data) the capsule mebrane suddenly broke, and a neat radioactive puff was revealed in the oven stack. Some examples of the obtained (Count rate at the oven stack) vs (Temperatue in the inner of the coal mass) as shown in Figure 4. The proposed method offers a low cost, safe, highly precise alternative to indirect methods for temperature evaluation, and can be used both in static and in dinamic furnaces. With a suitable choice of different metallic plugs and radiotracers, a finer study can also be performed, allowing the investigation of temperature kinetics and distribution in the mass of coal, so optimizing process stages other than distillation end point.



Fig 3. Experimental setup in the coke industrial pilot oven.



Fig 4. Count rate of the Nal(TI) scintillator in the oven stack vs temperature of the inner of the cal in the furnace.

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# PREPARATION, IRRADIATION AND CLINICAL USE OF HOLMIUM CONTAINING MICROSPHERES OBTAINED BY NEUTRON IRRADITION IN TRIGA REACTOR

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#### ABSTRACT

A radioactive composition for use in therapy, containing radioisotopes immobilized on biocompatible and bioabsorbable poly(lactic acid)-based microspheres was prepared, and internationally patented. A 10% of an holmium organic complex was previously embedded within the matrix of the microspheres, that were successively irradiated in a TRIGA reactor for 1-to 3 hours 2 hours at a flux of  $1,25.10^{13}$  n cm<sup>-2</sup> s<sup>-1</sup>, obtaining a total activity of 5-200 mCi of <sup>166</sup>Ho per irradiation. The irradiated microspheres were certified sterile at the end of the irradiation, due to the gamma and neutron flux of the reactor. The radioactive microsphere were dispersed in a biocompatible and bioabsorbable matrix consisting of a hypotonic gel, and were used in the therapy of neoplastic diseases, by application of radioactive sources in direct contact with the tumor tissues or within the same (brachytherapy). Patients with II and III grade glioblastoma multiforme were treated, by hypodermic injection in the Ommaya reservoir placed under the patient's scalp, visualising the introduction of the microparticle-containing gel in the brain cavity by computed tomography (CT) and gamma scintigraphy. It has thus been ascertained that the introduced substance forms, at the outset, an approximately spherical deposit, and that the gel is later gradually absorbed in the surrounding tissue, thus leaving the dispersed radioactive particles in contact with all of the interstices of the brain cavity. Patients with solid hepatic or splenic tumours, that were considered untreatable by surgery due to bad general conditions or advanced age, have been treated by injecting intratissue the microspheres in the tumour lesions, under stereotaxy and ultrasound or radiographic visualisation. Patients with solid hepatic, adrenal or renal tumours, that were considered untreatable by surgery due to bad general conditions or advanced age, have been treated by injecting intra-arterially the microspheres in the tumour lesions, under stereotaxy and radiographic visualisation. The gel preparation according to the invention insures that that all the microspheres are homogeneously expelled by the syringe during the injection in the tumour lesions, which also allowed contains a controllable and uniform deposition of radioactive particles.Partial and, in some cases, total remission were obtained in many of the treated patients; without any toxic or radiotoxic effects, after 1 to 3 applications of the microspheres.

#### 1. Introduction

As it is known, there are many forms of cancer against which the conventional treatments, substantially based on surgery combined with external radiotherapy and/or with chemotherapy, are often ineffective. In some cases the conventional treatments are scarcely applicable in view of the location and diffusion of the lesions, or in view of the general conditions of the patient. Among such forms or cancer, brain tumors and liver tumors, both primary and originated from metastases, are particularly critical, have a high grade of mortality and are only occasionally treated successfully. The most lethal of the brain cancer forms cited above is glioblastoma multiforme a rapidly growing infiltrating tumor, that develops within the cerebral mass with branched projections similar to pseudopods. As the other brain tumors, glioblastoma is treated first of all with surgery, in every case where this is

possible. However, local recurrence is very frequent, owing to the practical impossibility of eliminating, even by a surgical exeresis macroscopically appearing guite radical, all of the tumor cells infiltrated in the surrounding healthy tissues. After the surgery, therefore, the patients undergo one or more cycles of irradiation of the tissue surrounding the original tumor site with electron beams or with gamma radiation. Such therapeutic approach, however, even when applied with the most sophisticated techniques, likefractionated stereotactic radiotherapy, has shown some efficacy only in the lowest grade forms. In order to limit as much as possible the damage induced by the irradiation on the healthy tissues, the postoperative treatment of brain tumors may resort to administration techniques that use the local-regional route, exploiting the principles of brachytherapy. In the present paper is described a radio-pharmaceutical product particularly suitable for the intracavitary therapy of neoplastic diseases, which is capable of irradiating in a uniform and selective way the interior of a cavity such as that resulting from the surgical removal of tumoral masses, reaching with its radiation all of the interstices that may be present within such a cavity and being gradually absorbed after some time, while not dispersing the radioactive agents in the surrounding tissues.A particular reservoir is used for such purpose, known as the Ommaya reservoir, which is permanently placed close to the brain lesion, after the surgical excision. The Ommaya reservoir substantially consists of a small silicone bag ending in a tube with a radioopaque terminal having a plurality of holes. The bag is placed under the patient's scalp, so as to be accessible through a simple hypodermic injection, while the perforated tip is inserted in the brain cavity left by the excised tumoral mass. Such system is presently used to introduce in a periodically repeated and non-invasive way chemotherapeutic agents or also labelled monoclonal antibodies into the brain. A layer of tissue some millimetres thick around the cavity has to be selectively irradiated without appreciably damaging, at the same time, the surrounding tissue. To that aim, the introduction of a liquid agent would be ineffective (unless such liquid was provided with means for selectively and reliably binding some given tumor antigens), as in a very short time the liquid would diffuse through the tissues, dispersing and indiscriminately irradiating wide zones of the body. A second form of administration, that can be used in forms like liver or kideney tumour, is the intratissutal direct injection into the tumor or by intraarterial infusion, through catheterisation of the blood vases.

The product described in the present paper consists in microsphere, made of poly(lactic acid), a polymer of lactic acid having a molecular weight in the range from 1000 to 60,000, that may be metabolised by the human body according to the normal metabolic routes up to lactic acid, a common physiological constituent. The microspheres contains a beta emitter isotope into its matrix (166Ho). Contrary to gamma-emitters, beta-emitters have a useful emission range not exceeding 1 cm. Such low penetration capacity, while being required in order to avoid an excessive damage to the healthy tissue close to the lesion, requires, on the other hand, a closer contact between the tissue to be treated and the radioactive sources.

# 2. Product preparation and irradiation

The microspheres were prepared by a solvent evaporation process. Briefly, 100-1000 mg of acid poly(lactic acid) and 10-100 mg of holmium acetylacetonate are dissolved in 2 ml of dichloromethane. The obtained solution is rapidly injected in 10-100 ml of an aqueous solution containing 1-5% by weight Polyvinyl alcohol of different MW's120,000 kept under rapid stirring. The Ho-containing microspheres formed are separated and dried. The dimensions of the microspherese is comprised between 2 and 50 mm. The Ho-containing microspheres (50 to 300 mg) were irradiated in high purity polyethylene vials, after mixing with mannitol powder in a ratio 1:3 to 1:10. Neutron irradiations were performed in the pneumatic rabbit system of ENEA Casaccia TRIGA Mark II reactor, for 0.5 to 2 hours, with a thermal neutron flux of 1.3 × 10<sup>12</sup> n cm-2 s-1. Holmium-166 (half-life = 26.8 h  $\beta$  and  $\gamma$  emission~94% and ~6%, 1.85 MeV Emax of energy beta particles) was obtained after the irradiation.

After the irradiation the particles are mixed with a biocompatible and bioabsorbable gel containing polyvinylpyrrolidone and agar, that is able to hold the microparticles in stable suspension at the body temperature, having at such temperature the consistency of a jelly. The gel can be easily mixed with the radioactive particles and administered to the patient by means of a syringe, and it can readily and homogeneously fill up the concerned cavity, or the chosen tumor mass. In this way the radioactive particles employed can strike with their emission the tissue surrounding the cavity, and provide a high absorbed radiation dose of radiation in a limited, definite tissue thickness. The particles sterility has been measured by incubating the particles with a growth media for bacteria; the material has always been found sterile, due to the high dose received in the reactor irradiation. The particles, put into contact with human serum, gave a loss of radioactivity <0.01% after 15 days of contact.



Fig 1. Microspheres preparation apparatus Fig 2 Intrartherial microsphere injection

# 3. Product administration

The microspheres after the irradiation have been suspended in the gel above described in a shielded glove box, and injected to the patients. A first administration route was by the use of selective catheterization of tumor vases; an example is shown in Figure 2, in which a contrast media has been mixed with the radioactive product. In Fig 3 is shown the selective radioembolization of a kidney, and in Fig 4 the corresponding scintigraphy, immediately after injection. In Fig 5 is shown the scintigraphy obtained after a selective radioembolization of a liver sector, obtained 3 days after the injection. In Fig 6 is shown the scintigraphy obtained after a selective radioembolization of the spleen, obtained 8 days after the injection. A second administration route was used for brain tumors, by using the above described Ommaya reservoir. Fig 7 shows the administration drawing, and Fig 8 shows a scintigraphy from a patients, immediately after injection into the Ommava reservoir. A third administration route has been the direct intra-tissue injection into tumor mass, under ultrasound or computer tomography guidance. In all injected patients no toxic effect have never been demonstrated; in 40 to 70 % of cases, a partial or a complete remission have been shown in intrartherially or intra-tissue administration. In the patients affected with glioma brain tumours (n=6), a significant life extension has been reached, and one complete remission has been achieved.



Fig 3 Selective radioembolization of a kidney

Fig 4 the corresponding scintigraphy, immediately after injection



Fig 5 Scintigraphy after a selective radioembolization of a liver sector, obtained 3 days after the injection.

Fig 6 Scintigraphy obtained after a selective radioembolization of the spleen, obtained 8 days after the injection





Fig 8 Scintigraphy from a patient, immediately after injection into the Ommaya reservoir.



Fig 9 Liver metastasis before treatment (left), and three months after a single intratissue injection of holmium microsspheres (about 10 mCi).

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## FUEL MANAGEMENT STRATEGIES FOR A POSSIBLE FUTURE LEU CORE OF A TRIGA MARK II VIENNA

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#### Abstract

The Vienna University of Technology/Atominstitut (VUT/ATI) operates a TRIGA Mark II research reactor. It is operated with a completely mixed core of three different types of fuel. Due to the US fuel return program, the ATI have to return its High Enriched Uranium (HEU) fuel latest by 2019. As an alternate, the Low Enrich Uranium (LEU) fuel is under consideration. The detailed results of the core conversion study are presented at the RRFM 2011 conference. This paper describes the burn up calculations of the new fuel to predict the future burn up behavior and core life time. It also develops an effective and optimized fuel management strategy for a possible future operation of the TRIGA Mark II with a LEU core. This work is performed by the combination of MCNP5 and diffusion based neutronics code TRIGLAV.

#### 1. Introduction

The Atominstitut (ATI) hosts the 250 kW TRIGA Mark II research reactor employing a completely mixed core of three different types including HEU FLIP fuel. This FLIP fuel is a SS clad 70% enriched fuel. Due to the US Fuel Return Program, the Vienna University of Technology-ATI is obliged to return its 9 HEU FE(s) by 2019. Most of the 102-types FE(s) (20% enriched and AI-clad FE(s)) in the current core are close to achieve their maximum burn-up values. Therefore, ATI thinks to return the FE(s) with maximum burn up numbers along with HEU fuel. Until now, there is no final decision on any further utilization of the Vienna research reactor beyond this limit of 2016. The three different possible scenarios for the future core have been studied at ATI [1]. Most attractive option is to utilize a mixed core of two types of fuels i.e. 104 and 108 type fuels. Both are Zirconium Hydride fuel with 8.5 w/o and 20 w/o uranium fuel respectively. This core contains total 54 FE(s). The 40 FE(s) are 108-type and needed for criticality. The remaining 14 FE(s) are 104-type.

The good core management strategy needs to achieve high discharge burn up ensuring the safe and efficient rated power operation. This requires safe implementation of planed strategies for fuel reshufflings and reloading schemes. In this study, the burn up calculations are performed to formulate an optimal fuel management strategy for maximum possible utilization of the fuel loaded to the possible future core. For this purpose, TRIGLAV computer code [2] is used to test various schemes for high burn up and reasonable flat thermal flux distribution.

#### 2. Methodology

#### 2.1. The ATI reactor MCNP Model

A detailed three dimensional MCNP [3] model of the TRIGA mark II reactor is developed and verified through local consistent experiments at ATI [4]. The confirmed model has been modified for the proposed mixed core to perform its neutronics calculations. The model considers only the structural importance for the parameters. For example, irradiation holes inside the core due to their least effect on the core reactivity are neglected in this model. Similarly, the exact geometry of the top and bottom Aluminum fixing is simplified into simple cylindrical geometry. The MCNP model is equipped with JEFF 3.1 neutronics data libraries. It incorporates all the necessary core components e.g. FE(s), central irradiation channel, three control rods, graphite elements, Be-Am source element inside the core. While annular graphite reflector, four beam tubes, thermal column and radiographic collimator are modeled outside the reactor core. All these components inside the core are shown in Figure 1. The top view of the MCNP model of the proposed mixed core has been shown in Figure 1.



Figure 1: Top or XY-view (left side) and Side or YZ-view (right side) of the MCNP model of the TRIGA Mark II reactor.

#### 2.2. Criticality Calculation

The criticality calculation gives confidence to perform burn up calculations for the proposed core. These calculations are performed by MCNP and the TRIGLAV for the core loaded with 104 and 108 type FE(s) at thermal power 250 kW. Table 1 presents the criticality results of the proposed mixed core [1].

Table 1: The criticality analysis of mixed core of the TRIGA Mark II reactor.

Case	FE(s)	U235 (kg)	K <sub>eff</sub> MCNP5	K <sub>eff</sub> TRIGLAV
Mixed core	40	3.940	$1.00081 \pm 0.00021$	1.00090

#### 2.3. Burn up calculations

The computer code TRIGLAV is applied to calculate the maximum achieved burn up for each ring of the core. In this TRIGLAV model, the core is loaded with 54 FE(s) including 6 burned fuel elements. The burn up of each burned FE is assumed as 1MWD. Without any reshuffling, the mixed core can be operated at 250 kW to achieve maximum burn up numbers as shown in the Table 2 which shows that operator has to reshuffle the core when these burn up limits are achieved.

Table 2. Duill	Table 2. Duffi up fluttibers of a filixed of				
FE position	Mixed core				
	Burn up (MWD)				
B-ring	13.50				
C-ring	10.73				
D-ring	9.10				
E-ring	*8.23				
_	**5.25				

Table 2: Burn up numbers of a mixed core.

\*108-type Fresh FE, \*\*104-type fresh FE

#### 2.4. Thermal Flux Density Distribution

The thermal neutron flux distribution in the core has significant importance. The theoretical/measured flux values are applied for the calibration of nuclear channels,

assessment of absolute power, power distribution in the core, identification of hot spots and calculation of fuel burn up etc. Since the flux density distribution is proportional to the power, therefore by knowing the thermal flux distribution, the power density can be calculated or the vice verse.

The MESH capability (F-4 tally) of the MCNP5 is applied to calculate the thermal flux distribution in the core as shown in the Figure 2. The thermal flux looks symmetrical as maximum in the centre and decreases along the radial dimensions of the core. The model applies 5x105 source size with 200 neutron life cycles.



Figure 2: MESH tally plot of the thermal flux distribution in the TRIGA reactor core.

#### 2.5. Power Pea kings

The power density distribution is deduced from the thermal flux distribution. It is seen that power peaking values (maximum value of power per FE) are usually found in fuel rods which are inserted near the core centre. In the present case, the hot spot is B02 which has power peaking of 1.454. The average power peaking values for B, C, D and E-rings are 1.442, 1.256, 0.989 and 0.746 respectively showing that new mixed provides a safe and uniform flux within the safety limits.

## 2.6. Core life time

The calculation of the keff and its relationship with core burn up (or full power days) days is of primary importance to determine the core lifetime. First the excess reactivity (Keff) for the beginning of the core life is calculated by TRIGLAV at 250kW reactor power and found to be 5.1596 \$ (keff = 1.0391455). Then the code is run for each optimal time interval (10 days) for first three successive keff calculations. Initially, the small step of 10 days is taken to see Xenon build. After Xenon achieves its equilibrium, this burn up time step is increased to 50 and then 100 full power days for fixed core calculation until it no longer influenced the core lifetime. These burn up calculations are performed for the proposed LEU TRIGA core without changing the loading pattern until the excess reactivity fell to zero.

The variation of the keff along the full power days (burn up) is shown in Figure 3 which shows that, theoretically, the proposed can be operated for about 2080 full power days.



Figure 3: The core excess verses burn up (reactor operation full power days).

#### 2.7. FM Strategy

For the maximum possible utilization of the TRIGA core, the in-core fuel management schemes are described here. For these calculations, the core is burnt without changing the loading pattern until the excess reactivity fell to zero and the results are plotted in Figure xx. It is seen that during first 10 full power days operation, the reactivity decreases sharply due to initial Xenon build up in the core. After steady state Xenon production, the excess reactivity decreases uniformly which can be seen in the Figure. After 2080 full power days operation, the burn up interval is increased to 100 full power days and this keff decreases and approaches to unity on 2380 full operation days. As a strategy, the core life can be significantly increased by reshuffling at an appropriate time when the decreasing curve becomes relatively sharper. This means that this is the nominal point for core life before the initial reloading step is needed. At this point the initial core requires added reactivity to remain operational at full power. This additional reactivity can be gained by simply replacing fuel elements by least burned elements. And the core Burn up should be continued until excess reactivity again fell to zero.

#### 2.8. Conclusion

The burn up and in-core fuel management strategies for the new LEU proposed core are investigated to optimize the core life for better utilization and safe future operation. The fuel elements should be kept in their respective ring positions up to their maximum burn up limits. It is also determined that reloading or reshuffling is needed to keep the reactor operation at full power before an excess reactivity approaches to zero. The combination of MCNP5 and TRIGLAV is applied for this study.

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## MODELLING THE FIR 1 TRIGA CORE FOR INCREASED POWER AND TILTED POWER DISTRIBUTION FOR FURTHER IMPROVEMENT OF BOTH THE EPITHERMAL NEUTRON BEAM INTENSITY AND THE CAPASITY FOR ISOTOPE PRODUCTION

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## ABSTRACT

Preparations for an improved core loading and a power upgrade of the currently 250 kW FiR 1 TRIGA reactor are made. Increasing the reactor power will allow to increase the capacity for BNCT cancer patient treatments by more than a factor of three, meeting the prospects of increased demand for these treatments, while still keeping the current capacity for radioisotope production, education and training.

A tilted core loading using 11 new fuel elements with 30 mass % of uranium in the UZrH matrix is considered. These would be placed into the core section facing the BNCT irradiation facility. Detailed reactor physics and thermal hydraulics calculations using the most advanced codes in use at VTT are performed to evaluate this option.

The power distribution is calculated using a detailed model in the MCNP5 code. The new SERPENT code developed at VTT is used for the burnup and radioactive nuclei inventory calculation. SMABRE thermal hydraulic system code is used for the core cooling calculations.

#### 1. Introduction

FiR 1 -reactor is a TRIGA Mark II type research reactor manufactured by General Atomics (San Diego, CA, USA) and started operation in 1962. Reactor power was increased in 1967 from 100 kW to 250 kW. Boron Neutron Capture Therapy (BNCT) work dominates the current utilization of the reactor. The weekly schedule allows still three days for other purposes such as isotope production, neutron activation analysis and training. [1]

FiR 1 has a special irradiation facility for BNCT [2,3]. The neutron moderator block of the BNCT station is used as an epithermal neutron source to treat tumours. For BNCT the FiR 1 core loading was made asymmetric in order to increase the intensity in the epithermal neutron beam without increasing the 250 kW maximum steady state power. Further improvement of the treatment quality and capacity by either shortening the treatment time or increasing the therapeutic effect of the BNCT neutron beam are aimed at by improving the core loading and/or by increasing the reactor power. In earlier evaluations performed by VTT doubling of the power from 250 kW to 500 kW has been set as a target.

To support these changes and to improve the general knowledge at VTT about the properties of the TRIGA reactor a modelling campaign for FiR 1 is currently underway. The power distribution is calculated using a detailed model in the MCNP5 code. The new SERPENT [4] code developed at VTT is used for the burnup and radioactive nuclei inventory calculation. SMABRE [5] thermal hydraulic system code is used for the core cooling calculations.

## 2. Improving the epithermal neutron beam by using high uranium concentration fuel

Increased neutron beam flux is sought for the patient irradiations to become shorter bringing increased patient comfort and positioning accuracy, as well as more optimized irradiation timing with rapid kinetics boron carriers. Higher flux at the beam exit window would be useful and even necessary for irradiation with a beam extension cone to be used with tumours in difficult locations; the cone attenuates the beam by 50%. Superficial tumours are generally irradiated with only one field and beam durations of the order of 40 minutes causing sometimes discomfort for the patient and long waiting times for the personnel; also in these cases increased beam intensity would be of benefit both to the patient and the personnel. Increased beam intensity would also allow to use a lithium filter for hardening of the neutron spectrum in the beam for deeper treatment penetration, compensating for the attenuation of the beam by the filter.

A tilted core loading using new fuel elements with 30 mass % of uranium in the UZrH matrix is considered. These would be placed into the core section facing the BNCT irradiation facility. Detailed reactor physics and thermal hydraulics calculations using the most advanced codes in use at VTT are performed to evaluate this option.

#### 2.1 The MCNP5 model

The power distribution and control rod values are calculated using a detailed model in the MCNP5 code. The reactor core with the graphite reflector and the surrounding water as well as the BNCT epithermal beam installation are included in the model. Model cross-section in the horizontal plane is presented in Figure 1.



Fig 1. MCNP model of the current FiR 1 core with detailed description of the fuel elements, graphite reflector (dark blue), Fluental<sup>™</sup> neutron moderator (light blue) and bismuth gamma shield (purple) as exit window of the epithermal neutron beam.

The FiR 1 core consists currently of aluminium clad elements with 8 w% of uranium and steel clad elements with either 8,5 or 12 w% of uranium. The enrichment of U-235 is 20%. New core configurations with 30 w% uranium containing fuel elements have been studied. The idea is to put more uranium in the section of the core that is facing the epithermal beam

facility. In this case on the opposite side of the core fuel elements are replaced by graphite elements not to increase the power of the reactor. If power increase would be allowed more fuel elements would be left into the core. Two of the studied configurations are shown in Figure 2.



Fig 2. MCNP model of two core configurations with new 30 w% fuel (marked with 30). On the left (case 2) six new elements in the outer F-ring, on the right (case 3) six new elements in the outer F-ring and five more in the E-ring. Graphite elements are blue marked with C. The location of the regulating rod is also shown (Reg).

#### 2.2 MCNP5 results

Compared to the current situation the intensity of the epithermal beam increases in case 2 by 34% and by 54% in case 3; assuming 250 kW reactor power in all cases.

The powers of the new 30 w% U fuel elements are about 60% to 70% higher than those of the 12 w% fuel in these positions currently. The highest powers per fuel element do not increase that much; currently the highest power is 5,7 kW and in case 2 the highest fuel element power is 6,5 kW and in case 3 7,1 kW.

The radioisotope production rate in the central tube will remain the same in all cases 1 to 3.

The total reactivity of the regulating rod is in case 1 1\$, in case 2 0,6\$ and in case 3 where it is located closer to the center 1,2\$.

#### 2.3 Thermal hydraulic modelling

SMABRE thermal hydraulic system code is used for the core cooling calculations. The first model produced is very simple and the goal was mainly to verify that the SMABRE code is suitable for modelling the natural circulation in the TRIGA reactor.

In the model the tank was divided radially into two parallel channels, one of which describes the core and areas below and above it. The second channel represents the part of the tank, which is outside the horizontal cross section of the core. Both channels were divided into ten sections axially. The channels in the tank were connected to each other with junctions to enable cross currents. The reactor core was modelled using 13 radial hydraulic channels. Axially the core was divided into 12 parts, of which 10 are active fuel zone and two in the graphite part of the fuel element at each end. In the core the model does not include cross currents. The nodal structure is shown in Fig. 4. The thermal power structures were described based on the MCNP5 model results.

The first results indicate that the current SMABRE model allows for the creation of a natural cycle in the tank and the temperature distribution in the tank and in the core are as expected. Model has to be further developed in detail before it can be used in real analysis. The model should also be validated by comparing calculation results with measured values in order to ascertain the reliability of the results.

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Fig 4. The nodal structure of the SMABRE model for FiR 1.

#### 3. Burnup calculations

For burnup and detailed nuclei inventory calculation the Serpent code is used. The final goal is to obtain an accurate inventory taking into account the reactor operation from the beginning in year 1962 until today. In the work of Clément Barray [6] each fuel rod is composed of eight cells in the 3D model. Each cell contents is considered separately. The use of many cells for each fuel rod permits to have a model close to reality. It was demonstrated that Serpent is a suitable and powerful tool for this task. The preliminary results are mostly close to the earlier results produced using the CASMO code at VTT.

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## FAST SAMPLE TRANSPORTATION SYSTEMS FOR INAA AT TRIGA REACTORS

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The facilities of short-time neutron activation analysis at the TRIGA Mark-II (250 kW) reactor of Atomic Institute-Vienna were completely reconstructed to implement the new generation of digital gamma spectrometers, to facilitate the analysis of large samples, to enhance the sensitivity and the quality of measurements, to develop modern and fast control units, to implement moveable neutron filters for thermal-/epithermal irradiation, to implement moveable counting chambers for accurate analysis at high count rates and to develop software packages for fully-automatic analysis. The quality and performance of the facilities were tested using radioactive sources and standard reference materials. The results indicate the effective and dynamic operation of the new irradiation-counting facilities.

#### Introduction

NAA requires a strong thermal neutron source such as nuclear research reactors (NRR). There about 270 NRR are operating in the world. Since 60 years, NRR act as centres of excellence in science and technology and one of the main activities is the elemental chemical analysis using the neutron activation analysis (NAA) method. NAA is an analytical method able to investigate about 65 elements in all types of materials /1-2/. The most powerful advantage of NAA is the capability to apply different techniques to optimise the analytical conditions for a certain group of elements. This gives the NAA to be very selective, sensitive and accurate. Some of these techniques require complicated technical facilities to facilitate their applications for example, sample transportation systems (STS) for short-time activation. STS is required to transfer the samples into the core of the reactor for irradiation then sending them back to the measuring stations. The fully-automatic STS are used for irradiation and counting the samples at optimal conditions, including the implementation of samples exchanger, optimizing the sample detector distances and managing the measurements by using multi-detectors system. The short-time activation analysis facilities at Atomic Institute were reconstructed to modify the old facilities which had been installed since at least 30 years,

#### Experimental

#### 1. Sample exchangers

Fig. 1A shows the construction and components of a sample exchanger for loading large number of samples of different sizes. It consists of a sample magazine, a loading device, an expelling unit, a special pneumatic cylinder and a Pb-shield. The sample magazine consists of four sample holders (containers), which are connected together. The magazines were fabricated from a Plexiglas. Three different magazines were designed to hold three distinct sample sizes: 20 ml, 5 ml and 0.5 ml. The total numbers of sample that can be loaded are 80, 120 and 200, respectively. The magazine is connected with a special pneumatic cylinder, which provides 4 positions to sequentially change the holders. Fig. 1B shows the construction and the main components of a sample exchanger (C) was constructed for 100 samples of 12 mm in diameter. The third sample exchanger (C) was constructed for a capsule diameter of 16 mm. It consists of a sample magazine with a separation unit, a sliding device to introduce only one sample. The unit is suitable to analysis 20-25 samples of 5 cm long.. The unit consists of three sample magazines; each is a 1 m long. The second part is three separation units to introduce only one sample into the system, each unit is equipped with two pneumatic cylinders.



Fig. 1: Construction and main components of sample exchangers

#### 2. Loading devices and sliding units

To guarantee the fully-automatic procedures it is necessary to develop and fabricate many small devices and sliding units. These units are usually fabricated from Plexiglas, polyethylene, and polyamide materials to avoid any contamination from metallic components. The figure shows three directions sliding device (A) which is powered pneumatically, using special cylinder to give three positions. This unit was constructed to direct the sample into the counting chamber; to the depot, or to another counting system. The figure shows the construction of a two directions sliding device (B) and a separation unit (D) to receive the sample after irradiation and then transferring it with clean pressurized air to the counting chamber.



Fig. 2: Construction and main components of sliding and loading devices

#### 3. Control units

The first control unit is based on an I/O card installed in a PC (40 digital I/O channels, an electronic unit, consists of a power supply 24 V), fast relay units and the connection interfaces for valves and optical sensors /3/. The second control is based on a programmable logic controller (PLC) and a group of interfaces and adapters to manage irradiation-measurement procedures. The unit was constructed to facilitate the control of two valve islands (VM10, each 8 valves) through two interfaces and 6 external higher flow rate valves and six sensors.



Fig. 3: Main components of two control units (based on A: I/O Card, B: PLC)

#### 4. Counting chambers

Fig. 4a shows the construction and main components of three counting chambers. The first was constructed for a sample size of 16 mm /3/. The second chamber was constructed in the way to accommodate and centre one of three sample sizes (0.5, 5, 20 ml). The third unit was constructed for 12 mm sample diameter. Each unit contains an optical sensor to start the measurement automatically.



Fig. 4a: Main components of the second control unit (based on PLC)

The counting chamber was further developed /4/ by introducing a special pneumatic cylinder which is connected to move it between 7 positions (1 cm steps) in front of the detector. Therefore, it is possible to set the optimal short-sample-detector distance according to radioactivity of the analyzed samples (Fig. 4b).



Fig. 4b: Construction and main components of the a moveable counting chamber

#### 5. Irradiation chambers

Fig. 5 shows two irradiation chambers. The first (5a) is installed inside the core for irradiating a sample of 12 mm in diameter /5/. The second system (5b) was constructed for irradiating a sample size of 15 mm facilitating to move the system between the F-ring and the centre of the core for irradiation at high neutron flux. The system will be installed after testing and ensuring an arrangement on the bridge of the reactor, to guarantee the safe installation and movement. The third system /6/ was constructed for a horizontal irradiation beam or to be used for non-reactor irradiation facilities (i.e., microtron).



Fig. 5: Construction and main components of two irradiation chambers

#### 6. Software

Software packages were developed to manage the irradiation –measuring procedures at optimal analytical conditions. The analytical procedure can be semi-automatic, automatic or a combination between the two techniques. Automatic procedure allows the analysis at certain shaping-time and a sample-detector distance. The main interface (Fig. 6) of the software indicates online the analytical information such as the sequence number, measuring time, geometry, shaping time, and dead time of the measured spectrum/6/. The software packages were designed to control several equipments (two detectors, two sample exchangers, an irradiation unit, a digital spectrometer valves and optical sensor) for a complete fully-automatic short-time activation analysis.

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Fig. 6: Main interface for a software package (based on I/O card)

#### **Results and Discussion**

Fig. 7 shows the arrangement of the main five units (e.g., irradiation-, pneumatic-, control, counting and PC/software package) of a complete irradiation-counting facility for short-time activation analysis. The system consists of two irradiation facilities and multi-detectors system. The facilities optimize the irradiation-counting conditions for small and large samples. Several measurements were carried out using radioactive sources such as <sup>137</sup>Cs and <sup>60</sup>Co to test the reproducibility of the measurements at different analytical conditions (count rate; dead time, sample detector distances, shaping times). The standard deviation of 84 measurements was found to be less than 3%.



Fig. 7: Schematic and arrangement of the irradiation –counting units

#### CONCLUSION

Short-time activation analysis is a promising technique for analysing a large number of elements in all type of materials. However, the technique requires dynamic irradiation and counting facilities for sensitive and accurate analysis. This work indicates that a moveable counting chamber and a optimal throughput/ resolution are the basis of accurate analysis, at high count rates. The new irradiation and counting systems therefore, provide real dynamic conditions to optimise sample analysis independent of their matrices, weights, and the sensitivity as well as the half-lives of the investigated nuclides.

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# Proposal of LDR Ir-192 production in the TRIGA Mark II research reactor

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#### ABSTRACT

The TRIGA MARK II research reactor in Vienna provides some irradiation positions with different flux distribution. In this regard, a case study is under investigation to appraise the possibility of medical radioisotope production in Vienna. For this purpose, neutron flux mapping and the axial neutron flux distribution are calculated by MCNP5 for the TRIGA Mark II core. This paper describes the feasibility of Low Dose Rate (LDR) <sup>192</sup>Ir production in the core of the low power research reactor.

#### 1. Introduction

TRIGA MARK II research reactor located in Vienna, is a small research reactor for research purposes. This research reactor has a thermal power of 250 kW<sub>th</sub> which is under operation since 1962. It utilises a completely mixed core of three types of fuels including high enriched FLIP fuel. Fast and thermal neutrons are generated in the empty positions of TRIGA MARK II core with the orders of  $10^{12}$  to  $10^{13}$  [n/cm<sup>2</sup>sec] (see table 1). These flux values are not high enough to produce radioisotopes with high activity levels. In this case, the production of radioisotopes for medical purposes and lower activity levels are proposed. Neutron flux fluctuation and axial neutron flux distribution are the important parameters for radioisotope production in low power research reactors.

	Neutron flux [n/cm2sec]		
Positions	Fast spectra	Thermal spectra	
	>10 keV	<0.21 eV	
Central irradiation position	1.2 x 10 <sup>13</sup>	1.0 x 10 <sup>13</sup>	
E-Ring	6.4 x 10 <sup>12</sup>	4.1 x 10 <sup>12</sup>	
Pneumatic transfer system (F-Ring)	3.5 x 10 <sup>12</sup>	4.3 x 10 <sup>12</sup>	
Rotary specimen rack (uninstalled)	2 x 10 <sup>12</sup>	2 x 10 <sup>12</sup>	

Tab 1: Neutron flux values of a TRIGA MARK II research reactor at 250 kW steady state operation [General Atomic (GA)]

In order to appraise the production of radioisotopes in the empty positions which can be used for irradiation of targets, knowledge of flux levels and spectra are needed. For this purpose, Monte Carlo techniques based computer code MCNP5 is applied in order to select the best positions with high neutron fluxes, appropriate neutron spectra and to calculate the axial flux distribution in the selected positions. The next step will be the activity level determination of the targets which are loaded into the selected positions.

#### 2. TRIGA MARK II core flux mapping

As it is mentioned in previous section, the TRIGA MARK II - Vienna consists of a mixed core and the data regarding the neutron flux distribution has been published in the references [T. Stummer et al] and [R. Khan et al]. This mixed core is planned to be operated until 2016 and its neutron flux data is applied for the first approximation of the LDR <sup>192</sup>Ir source production in the central irradiation position of the core.

One of the possible option for the future reactor operation is to employ fresh 20 w/o LEU fuel with 20% enrichment. The new core would provide more positions for irradiation proposes because of new high density fuel. In this case, the axial flux distribution (see figure 2.1) inside the central irradiation position of the new core is calculated by using Monte Carlo technique (MCNP5). In addition, a target assembly (see figure 2.2) designed for LDR <sup>192</sup>Ir sources is modelled and neutron flux depletion is studied.



Fig 2.1. Axial neutron flux distribution of bare central irradiation position



Fig 2.2. Axial neutron flux distribution of filled central irradiation position

The analysis of the data of the figure 2.1 provides the information regarding the average neutron flux (thermal and epithermal components) deviation of the axial distance of 12 cm. The flux variation through 12 cm axial distance is found from 3.15 to 6.73 % reflecting the possible flat flux.

By inserting the target assembly made by aluminium, the depletion of the thermal and epithermal flux is also calculated and presented in figure 2.2. The data show that the thermal neutron flux depletion is higher than the epithermal component.

#### 3. LDR Ir-192 wire production feasibility

Iridium is a hard metal of the platinum family which has the density of 22.56 g/cc. It has two naturally occurring, stable isotopes, <sup>191</sup>Ir and <sup>193</sup>Ir, with natural abundances of 37.3% and 62.7%. By neutron irradiation of metallic iridium, two isotopes such as <sup>192</sup>Ir (half-life of 73 days) and <sup>194</sup>Ir (half-life of 19.2 hours) are produced (cross-section data provided in figure 3.1).

The isotope <sup>192</sup>Ir is used as a Brachytherapy source for high/low dose rate (HDR/LDR) applications [Nath et al]. The neutron flux values for the production of HDR <sup>192</sup>Ir sources is not sufficient enough. In this case, the feasibility of LDR <sup>192</sup>Ir source is investigated here.



Fig 3.1. Total absorption cross-section of natural iridium [Source: ENDF/B-VI-4, MT-27]

LDR <sup>192</sup>Ir wires are produced by irradiation of commercially available Ir-Pt (20/80 or 30/80) wires (0.3 to 0.5 mm in diameter) or iridium wires (0.1 to 0.3 mm) which are coated with pure platinum. The activity level of the LDR <sup>192</sup>Ir sources varies respectively from 0.8 mCi/cm (30 MBq/cm) up to 10 mCi/cm (370 MBq/cm).

The activity levels for the iridium production is calculate using activation formula with neutron self shielding correction stated in the reference [TECDOC-1340]. The calculation includes the enrichment and target burn up. As flux values, thermal and epithermal fluxes are used with the knowledge of thermal cross-section and resonance integral obtained from the literature (Atlas of Neutron Resonances: Resonance Parameters and Thermal Cross Sections, S. F. Mughabghab, Elsevier Science; 5 edition, April 2006). Pure iridium wires with diameter 0.1 and 0.3 mm are selected for this study.

The results for different flux levels are presented in figure 3.2 and 3.3. The results obtained from the above mentioned investigation, are sufficient enough to start series of experimental investigation for the production of LDR <sup>192</sup>Ir sources. The thermal neutron flux levels of  $1 \times 10^{13}$  and  $5 \times 10^{12}$  [n/cm<sup>2</sup>sec] are selected and applied as two scenarios for activity level calculation of the mentioned iridium wires.

For the first scenario, the maximum activity of 10 mCi/ cm (370 MBq/cm) can be achieved by irradiation time of 60 hours. For the second scenario the mentioned activity is also reached by selecting of 140 hours for irradiation.



Fig 3.2. Ir-192 activity calculation for thermal neutron flux of 1x10<sup>13</sup> [n/cm^2sec]



Fig 3.3. Ir-192 activity calculation for thermal neutron flux of 5x10<sup>12</sup> [n/cm^2sec]

## 4. Conclusion

This paper gives feasibility idea of LDR <sup>192</sup>Ir production by using the actual core of the TRIGA MARK II – Vienna for local supply. In case of core converted to new LEU core, there will be more positions for irradiation. The next step of this work will be the experimental investigation for <sup>192</sup>Ir production which is under consideration.

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## AN OBJECT ORIENTED APPROACH TO SIMULATION OF TRIGA MARK II DYNAMIC RESPONSE

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### ABSTRACT

This paper deals with the development of a model for the nuclear research reactor TRIGA Mark II operating at University of Pavia. The purpose of the modeling is to reproduce the dynamic behavior of the reactor on the entire operative power range, i.e. 0+250 kW, using the *object oriented* approach, implemented by the Modelica language. The main advantage is the a-causal formulation of the model, based on equations instead of statement assignment. Equations do not specify which variables are inputs and which are outputs, thus the causality in the model is unspecified and is fixed only when the corresponding equation system is solved. In this way, equations can be solved according to the data flow context in which the solution is computed. The model describes the entire plant, including the heat removal system. The component representing the reactor core contains a series of subcomponents linked together through rigorously defined interfaces: in this way, it is possible to consider the interactions between the different physical aspects of the system. Equations governing natural circulation have been implemented in a component which defines the mass flow rate through the core, according to the temperature difference at the ends of the channels. Secondary and tertiary cooling loops are modeled using a simplified heat exchangers configuration: concentric tube version is adopted, which allows recreating the heat exchange dynamics without a great modeling effort. The developed a-causal model has been validated through the comparison with experimental data collected on the site, concerning three different power transients at 100 kW. 50 kW and 1 kW. A corresponding causal model has been referenced as concerns fuel and coolant temperatures evolution during the transients. The predictions of the two main approaches to dynamic modeling have been compared. A very satisfying accordance is found as discrepancies observed on the coolant temperature are comprised between 0.5% and 1% for the three transients.

#### 1. Introduction

Key features of a dynamic system simulator appear: "*Modularity*" - the virtual plant should be built by connecting its components models, which have to be written independently on the context where they are employed; "*Openness*" - the code should be easily readable and close to the original form of equations; "*Efficiency*" - the simulation code should be fast running; "*Tool support*" - the simulation tool should be based on reliable, accurate and tested software. Few simulation environments fulfilling all these characteristics seem to be currently available for nuclear applications. Recent advances in object-oriented modeling, and in particular the development of the Modelica language [1] [2], represent a viable path to achieve the above-mentioned goals. Introduced in 1997, Modelica is employed for the modeling of general physical phenomena, described by sets of algebraic and differential equations. The main advantage is the *a-causal modeling* approach: the system behavior is described in terms of conservation laws, that, combined with components constitutive equations, determine the overall system of equations to be solved. With the a-causal formulation, equations of each model are written independently on the actual boundary conditions and it is not necessary to establish a-priori which variables will work as inputs and which as outputs. Model causality of the model is determined automatically by the Modelica model interpreter or compiler at the aggregate level, when a system model is assembled out of elementary ones. In this way, models are much easier to write, document and reuse, while the burden of determining the actual sequence of computations required for the simulation is entirely left to the compiler. In addition, the multi-physics approach of the Modelica language must be considered. General in scope, it provides modeling primitives such as generic algebraic, differential and difference equations, and is not tied to any specific physical or engineering domain, such as mechanics, electrical engineering, or thermodynamics. Thus it is guite straightforward to model multi-disciplinary systems, e.g. the reactor core, where several physics (neutronics, heat exchange and fluid dynamics) interact with each other. For these reasons, Modelica language has been selected to simulate the nuclear research reactor TRIGA Mark II operating at University of Pavia, aimed at reproducing its dynamic behavior on the entire operative power range, i.e. 0+250 kW. TRIGA Mark II is a nuclear research reactor widely used as non-power nuclear reactor in many applications, including production of radioisotopes, non-destructive testing, basic research on the properties of material, and for education and training [3]. The present work is organized in two parts. The first one is focused on model development, describing the objects employed to recreate the primary loop and the cooling system. As simulation environment, Dymola (Dynamic Modeling Laboratory) has been adopted, as it allows the hierarchical composition of models and offers wide libraries of pre-defined components. The second part is focused on the model validation process and in the evaluation of the reactivity feedback coefficient due to fuel temperature.

#### 2. The reactor model

In this work the *component based approach* has been followed: a single model has been adopted for each one of the required components. Once single models are available, they can be connected in a variety combinations [4]. All the components adopted are taken from the *ThermoPower* and *NuKomp* libraries [5] [6]. In Figure 1, the model of the entire plant is represented. It is composed of three main loops: on the right, the reactor core and the primary cooling system, which removes the heat from the water tank. Heat is then transferred to the secondary water cooling system, through a shell and tube heat exchanger. Proceeding to the left, the secondary cooling system, composed of the secondary and tertiary loops, receives the heat from the primary one and discharge it to the sewerage system, which works as heat sink [3].



Figure 1. Model of the entire plant of the nuclear reactor TRIGA Mark II, operating at University of Pavia, and the respective heat removal system.

The component describing the reactor core and its respective detailed view are represented in Figure 2. It is composed of three sub-systems, each one describing a particular physical aspect of the model. The component Kinetics determines the neutron density, based on point reactor kinetic model, with one energy group and six delayed neutron precursor groups [7]. The component Fuel Rods describes the behaviour of the fuel elements: once defined five spatial concentric regions within the element, the time-dependent Fourier equation is applied, neglecting heat axial conduction since the length-over-diameter ratio is much greater than 10. Finally, the component Core Moderator describes coolant flow through the core channels. Only gravitational and distributed pressure losses are considered. Form pressure drops caused by area contractions are neglected, since flow velocity due to natural circulation is small. Convective heat transfer coefficient, which depends on coolant flow condition as well as on core geometry, is estimated using the Dittus-Boelter correlation [8], valid for turbulent flow in straight channels, updating its value in agreement with the mass flow rate. All the subsystems are connected through rigorously defined interfaces or "connectors" corresponding to the physical interaction with the outside: in this way, it is possible to describe the influences between neutronics and thermal-hydraulics, represented by fuel and moderator feedback temperature coefficients.



Figure 2. Model of the component *Core* and its respective detailed view, showing the sub-systems involved.

Equations governing natural circulation have been implemented in a component, situated upstream the reactor core. The natural circulation determined by the presence of a density gradient between core inlet and outlet sections is reproduced by a dedicated component, similar to a hydraulic pump. Coolant mass flow rate through the core is determined according to temperature difference at the ends of the channels [9]. The component is taken from the *ThermoPower* library. A suited modification has been introduced in order to have as input the coolant temperatures and as output the corresponding mass flow rate, determined by the buoyancy effect, in accordance to Boussinesque approximation (Figure 3).



Figure 3. The modified component  $G_{in}$  describing the natural circulation effects and its respective detailed view.

#### 3. The cooling system model

Several strong simplifying hypotheses have been adopted when modelling the cooling system, in particular regarding the heat exchanger configuration. That is a component representing a shell and tube heat exchanger on the whole requires a great and unnecessary modeling effort. Being the aim to reproduce the dynamic behavior of the system, a simplified version can be adopted, based on concentric tubes, with counter-current flows. The geometry has been reproduced adopting different tube models connected together. Moreover, all the processes involved in the heat transfer have been described by several appropriate components and then lumped together in a single model representing the thermal interaction between primary and secondary fluids [11], as it is shown in Figure 4. As in reactor core, the convective heat transfer coefficient depends on the flow condition and it is estimated by the Dittus-Boelter correlation both for internal and external tubes. Moreover, additional models have been introduced to describe wall conductive resistance and thermal fluxes and temperatures vectors as well, to take into account the counter current configuration of the heat exchangers.



Figure 4. The global heat transfer model in the adopted heat exchanger and its respective detailed view.

#### 4. Simulation Results and Validation Process

The developed a-causal model has been validated through the comparison with experimental data collected on the site, concerning three different power transients at 100 kW, 50 kW and 1 kW. The model reproduces the real behavior of the system in a very satisfying way: the error on power response simulation is less than 0.44% for the first transient and less than 0.3% for the other two transients. In addition, fuel and coolant temperatures evolution during the transients have been compared to the results of a corresponding causal model. While Modelica language allows a 1-dimensional description of system components, the corresponding causal model is 0-dimensional and describes the reactor core, taking into account the coupling between neutronics and thermal-hydraulics in natural circulation, by means of fuel and moderator temperature feedback coefficients [10]. In this way, the predictions of the two different approaches to dynamic modeling have been compared (Figure 5). A pretty fair accordance is found as the discrepancies observed on coolant temperature are comprised between 0.5% and 1% for the three transients. However, a similar comparison is not possible regarding the fuel temperature, due to the different definitions adopted for the mean temperature value.



Figure 5. Results of the validation process: from top to bottom, the comparisons between powers during the three different power transients, i.e. 100 kW, 50 kW, 1 kW, are presented.



Figure 7. Results of the validation process: from top to bottom, the comparisons between fuel temperatures during the three different power transients, i.e. 100 kW, 50 kW, 1 kW, are presented.



Figure 8. Results of the validation process: from top to bottom, the comparisons between coolant temperatures during the three different power transients, i.e. 100 kW, 50 kW, 1 kW, are presented.

#### 5. Conclusions

In this paper the development of an object oriented model simulating the characteristics of the research reactor TRIGA Mark II operating at University of Pavia (L.E.N.A.) has been described. Model predictions for reactor power have been compared with experimental data collected at LENA: the model accurately reproduces the behavior of the reactor within the entire operating power range (0÷250 kW), as the discrepancies observed are less than 0.5% in all the three transients analyzed. The discrepancies observed can be ascribed to the

several simplifications introduced: Boussinesque approximation to describe the buoyancy effect, assumption of material properties to be independent on temperature, Dittus-Boelter correlation used for heat transfer computation, the radical simplifications of heat removal system structure. Finally, the developed model has been validated by reproducing the dynamic behavior of the reactor and relative heat removal system at different power levels. Moreover, the possibility of realizing a one-dimensional description of the system permits to study the evolution of space dependent variables, such as the fuel temperature field. The proposed description of the plant constitutes a reliable tool to develop the control system for this kind of reactors.

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## THERMAL NEUTRON FLUXES CHARACTERIZATION IN THE IRRADIATION CHANNELS OF THE IPR-R1 USING MONTE CARLO METHOD

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## ABSTRACT

The IPR-R1 is a reactor type TRIGA, Mark-I model that is installed at Nuclear Technology Development Centre (CDTN) of Brazilian Nuclear Energy Commission (CNEN), in Belo Horizonte, Brazil. It is a light water moderated and cooled, graphite-reflected, open-pool type research reactor. IPR-R1 works at 100 kW but it will be briefly licensed to operate at 250 kW. It presents low power, low pressure, for application in research, training and radioisotopes production. The fuel is an alloy of zirconium hydride and uranium enriched at 20% in <sup>235</sup>U. This reactor has a Rotary Specimen Rack, RSR, outside the reactor, and it is composed by forty irradiation channels in a cylindrical geometry. In this work the IPR-R1 was simulated using the code MCNPX2.6.0 (Monte Carlo N-Particle Transport eXtend). The goal is to evaluate the neutronic flux in a sample inserted in the RSR channels. In each simulation the sample was placed in a different position, totaling forty positions around the RSR. The results obtained from the calculation show good agreement in relation to with experimental data.

#### 1. Introduction

Since 1960 the IPR-R1 reactor operates at Nuclear Technology Development Centre (CDTN) in Belo Horizonte, Brazil. This reactor is a TRIGA MARK-1 type which has the main nuclear applications related with neutron activation analysis (NAA), training of operators for nuclear power plants and experiments in neutronic and thermal-hydraulic. The NAA is an induction process of artificial radioactivity where the samples are subjected to neutron flux. In this way, the characterization of neutronic flux behaviour is important in this practice to application of k<sub>0</sub> method which is applied in NAA [1]. Experimental works [2, 3] have been measured the neutronic flux inside the cylinder in the RSR which contains the samples to irradiation. Other study has calculated this neutronic flux using the nuclear code MCNP4B (Monte Carlo N-particle Transport - version 4B). The present work simulates the IPR-R1 using two MCNP versions: MCNP5 and MCNPX 2.6.0 (Monte Carlo N-particle Transport eXtend - version 2.6.0). The models configured using these codes are based in previous studies [4] and some improvements in the geometry were performed. The goal is to compare the calculated values of thermal neutronic flux with the experimental results for the process of the model validation. Moreover, the values calculated previously with MCNP4B by MENEZES [2] were compared with experimental results. Therefore, three models of different MCNP versions are evaluated in this paper: Model 1 (simulated with MCNP4B), Model 2 (simulated with MCNP5) and Model 3 (simulated with MCNPX 2.6.0).

#### 2. Methodology

#### 2.1. Some Characteristics of Simulated Reactor

The IPR-R1 is light water moderated and cooled; graphite-reflected, open-pool reactor which works at 100 kW but it will be briefly licensed to operate at 250 kW. It presents low power, low pressure, for application in research, training and radioisotopes production. The fuel is homogeneous mixture of zirconium hydride and Uranium 20% enriched in <sup>235</sup>U isotope. The core has a radial cylindrical configuration with six concentric rings (A, B, C, D, E, F) with 91 channels that are surrounded by coolant (water). This core contains 59 fuel elements cladding with aluminum, 4 fuel elements cladding with stainless steel, 23 radial reflectors elements, 3 control rods (shim, safety and regulating), 1 central channel, and 1 neutron source. The fuel elements have three axial sections, upper and lower reflector (graphite), and the central portion filled with fuel (U-ZrH). The radial reflectors elements are covered with aluminum and they have the same dimensions of the fuel elements but are filled with graphite. The control rods have boron carbide and also cladding with aluminum. In the center of the reactor there is an aluminum tube (Central Thimble) to irradiation of the experimental samples. This tube is removable and, when it is not in use, the reactor pool water occupies its volume. Furthermore, the core has an annular graphite reflector which is cladding with aluminum layer. This annular reflector has a radial groove where a Rotary Specimen Rack (RSR) is assembled for insertion of the sample to irradiation. In such RSR is possible to place the samples in 40 different positions around the core. Moreover, tangent to annular reflector, there is a Pneumatic Tube where the samples also can be inserted to irradiation. Therefore, the IPR-R1 has three facilities for sample irradiation: the Central Thimble, the Rotary Specimen Rack and the Pneumatic Tube. Fig. 1 shows the radial and axial core configuration.



Fig. 1. Design of IPR-R1 core.

As described before, the reactor model was developed using MCNP5 and MCNPX codes where 500 active cycles were calculated with 1000 neutrons per cycle. The simulations executed in MCNPX code, considers 1.0 hour of irradiation (time of sample irradiation) with 0.1 MW (currently TRIGA reactor power). The simulated model was based on previous studies [4] where the reactor core has the same features of the IPR-R1 geometry described before. The configured geometry is the same to MCNP5 and the MCNPX 2.6.0. The core was configured considering a cylinder containing water, fuel elements, radial reflectors, central tube (or central thimble), control rods and neutron source. Each rod has a coordinate value. They were filled according to their individual characteristics. Around the core there is the RSR which has groove to insert the samples to irradiation. The configured core is inside the pool where water surrounds the core and the RSR. However, the model of this work presents improvements:

- 1<sup>st</sup>) In the RSR, the sample is placed inside three cylinders which are made of aluminum, polystyrene and polyethylene. Inside of aluminum cylinder is the polystyrene tube which contain the polyethylene cylinder where the sample is inserted (see Fig.2). The assembly of the three cylinders can be in 40 different positions (see Fig.1) in the RSR to sample irradiation.
- 2<sup>nd</sup>) The groove in which the RSR is assembled was configured with an aluminum layer (thickness of 1.6 cm).
- 3<sup>rd</sup>) The thickness of aluminum layer that covers the reactor core was increased (from 0.64 cm to 1.0 cm).

The Table 1 presents the cylinders dimensions which are inside of RSR. In addition, Fig. 2 illustrates the axial and radial view of the simulated model.

Tab 1. Dimensions of the simulated cylinders inside Rotary Specimen Rack.					
Cylinder Material	Inner Radius (cm)	Radial Thickness (cm)	Height (cm)		
Aluminum	1.50	0.10	20.0		
Polystyrene	1.10	0.30	7.90		
Polvethvlene	0.48	0.07	0.55		

Tab 1. Dimensions of the simulated cylinders inside Rotary Specimen Rack.





#### 3. Results

The assembly of the three cylinders was positioned in 40 different positions of the RSR to calculate the thermal neutron flux (energies of 0.5 eV or smaller) inside of the polyethylene cylinder that contains the sample. The Fig. 3 shows the thermal neutron flux simulated by MCNP5 and MCNPX codes. It is possible to see clearly the behaviour of the thermal neutron flux around the core reactor. The neutron fluxes vary at each position and changes over the RSR. In spite of the good agreement of the most of the calculated points some differences were observed. These differences must be justified by a theoretical analysis and using a statistical program.



Fig. 3. Axial and radial view of the reactor TRIGA IPR-R1 simulated in MCNP5 and MCNPX codes.

Although the neutron flux value was calculated in 40 positions in the RSR, the reference work [2] presents only 11 experimental data. Table 3 presents the values of thermal neutron flux calculated by the codes MCNP-4B, MCNP5 and MCNPX 2.6.0. The results are being compared with experimental data and the error is presented in percentage. Eight positions present differences smaller than 10% and three positions present differences smaller than 21%. In this way, it is possible to conclude that the most of the calculated results are in good agreement with the experimental measurements.

	Prev	vious Studie	S	Present Study				
Position	Experimenta	<u>Model 1</u> (MCNP- 4B)		Model 2	(MCNP5)	Model 3 (MCNPX)		
RSR	I Value*	Value*	Error (%)	Value*	Error (%)	Value*	Error (%)	
1	6.69	6.77	1.18	6.11	8.67	7.14	6.30	
3	6.55	6.65	1.50	6.60	0.76	6.50	0.76	
7	6.35	6.67	4.80	5.79	8.82	6.32	0.47	
10	5.99	6.90	13.19	6.44	6.99	6.24	4.01	
24	6.94	6.98	0.57	6.33	8.79	6.97	0.43	
25	6.45	6.86	5.98	6.91	6.66	6.54	1.38	
29	7.32	6.86	6.28	6.57	10.25	6.77	7.51	
34	7.30	6.73	7.81	5.90	19.18	5.77	20.96	
35	7.18	6.72	6.41	7.00	2.51	6.29	12.40	
38	6.58	6.80	3.24	5.76	12.46	5.58	15.20	
40	6.16	6.73	8.47	5.91	4.06	6.51	5.38	

Tab 2. Thermal neutron flux  $(n/cm^{-2}s^{-1})$ 

\*Value in the order of 10<sup>11</sup>

#### 4. Conclusions

The results presented in this work show that there is good agreement between the experimental data and the values calculated by the used codes. The errors presented by the most of estimated flux are smaller than 10% and three positions presented differences between 10 and 21%. The tool employed by MCNP5 and MCNPX 2.6.0 estimate the flux average inside of the cylinder which contains the sample. However, the used codes have other method to calculate the neutron flux as Forced Collision, Point Detectors, Spherical Detector, etc. These tools may improve the results decreasing the differences between experimental data and the calculated values. Future woks will simulate the IPR-R1 employing other method to flux calculate.

The information about neutron flux predicted by MNCP5 and MCNPX 2.6.0 can improve NAA where the sample activity can be estimated knowing neutron flux. Furthermore, these codes can characterize the neutron flux in other parts of the reactor where experimental measuring is difficult to be obtained.

#### Acknowledgments

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## NEUTRONIC PARAMETERS CHARACTERIZATION OF THE TRIGA IPR-R1 USING SCALE6.0 (KENO VI)

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#### ABSTRACT

KENO-VI is a Monte Carlo based transport code used to calculate the criticality of a nuclear system. A model built using this code with the SCALE6.0 code system was developed for the characterization of the neutronic parameters of the last cycle of the IPR-R1 TRIGA research reactor. A comparison with experimental values and those calculated with a MCNP code model could be then attained with the purpose to validate this methodology.

Key Words: KENO VI, TRIGA research reactor, SCALE6.0, Neutronic parameters.

#### 1. Introduction

TRIGA (Training, Research, Isotopes, General Atomics) reactors are research reactors developed by General Atomics and widely used in the world. Situated in the UFMG (Universidade Federal de Minas Gerais) campus, the IPR-R1, a TRIGA MARK 1 type reactor, operated by the CDTN/CNEN (Centro de Desenvolvimento da Tecnologia Nuclear / Comissão Nacional de Energia Nuclear), reached criticality for the first time in November of 1960 and it has been operational since then. Because of its long operational life, it is impractical to simulate the whole burnup history in details. For matter of simplicity, the history of this reactor was summarized in different cycles, the sixth one being the actual configuration.

In the present work, a Monte Carlo model of the IRP-R1 has been developed, through the ORNL (Oak Ridge National Laboratory) computer software system SCALE6.0 (KENO VI), with the intent of characterize the neutronic parameters of this reactor, such as the effective neutron multiplication factor, the neutron flux in the reactor irradiation devices and the reactivity of the core and of the control bars. In addition, isotopic characterization of the burned fuel, among other burnup data can be obtained for each operational cycles of this reactor with the use of the SCALE6.0 transport and depletion sequences (TRITON). The model, thus, could be validated by comparison with experimental results, as well as with results obtained with MCNPX, a Monte Carlo method based transport code.

#### 2. Materials and methods

The IPR-R1 core, with diameter of 109 cm, contains 91 elements arranged in six concentric rings (A to F) around its centre [1]. This core is located at the bottom part of a 192 cm diameter and 662.5 cm deep water pool. The pool structure consists of five coaxial cylinders, the innermost being 10 mm thick, made of aluminium, followed by a 72 mm of concrete, 6.3 mm of steel, 203 mm of concrete and the outermost, 6.3 mm of steel.

The IPR-R1 has its core immerged into a demineralised light water pool, which works as coolant, neutron moderator and reflector, and as a radiation shield. There are 91 positions in the active part of the core and each one can be filled with fuel or control rods, graphite "false" elements, neutron source or irradiation devices. Initially, the IPR-R1 operated at a constant power of 30 kW. Currently the maximum operational power reached 250 kW. In this simulation we considered the configuration of the sixth cycle during year 2004, in which the IPR-R1 operated with 63 fuel rods at 100 kW. The core configuration for this cycle can be seen in Fig. 1.



Fig 1: Core configuration.

There are two types of fuel rods currently being used by CDTN. First type rods have aluminium cladding, while the second type ones use stainless steel as clad (Fig. 2). Composition of fuel elements consists of an homogeneous mixture of metallic uranium and zirconium hydride, being 8 - 8.5% of the weight composed by uranium enriched to 20%. Both types have an axial graphite reflector on both ends of the fuel. Moreover, the aluminium cladding rods have two burnable poison disks between the fuel and the reflector, made of samarium trioxide in an aluminium matrix. The stainless steel cladding rods have in their centre a zirconium rod, same length of the fuel and with a 6.35 mm diameter.



Fig 2: Fuel rods.

The core central rod is an aluminium tube with a 1.69 cm inner radius and a thickness of 215 mm. The interior of the tube is filled with water and can be used to insert samples for the acquisition of experimental data. There are identical three control rods called regulator, control and safety, respectively, made of Boron Carbide bars 38.1 cm long and 1.9 cm diameter, surrounded by an aluminium coating of 0.145 cm thick. The guide tube is made of aluminium with inner radius of 1.6 cm and with a thickness of 3 mm. The core is surrounded by layers of aluminium and graphite coaxial cylinders. There is also a cylindrical sample table where different types of materials can be placed to be irradiated. In this simulation, this table is filled with air.

A 3D model of the IPR-R1 was built with the Monte Carlo criticality sequence KENO-VI of the SCALE6.0 computer code, to obtain the neutron multiplication factor for two different positions of the each control rod, in and out. Figure 3 shows the geometric configuration of the IPR-R1.



Fig 3: The active core of the IPR-R1

KENO-VI calculates the criticality parameters using the Monte Carlo method and is capable of building a large amount of different volumes. Although its primary purpose is to determine  $k_{eff}$  [3], the usual parameter employed in the reactors field is the reactivity, which measures how far from criticality is the system. We define the excess reactivity of the core,  $\rho$ , by

$$\rho = \frac{k_{eff} - 1}{k_{eff}} \tag{1}$$

where  $k_{eff}$  is the effective neutron multiplication factor of the core with all control rods out. The reactivity worth of each control rod is obtained by

$$\Delta \rho = \frac{1}{(k_{eff\,1})} - \frac{1}{(k_{eff\,2})}$$
(2)

where  $k_{eff1}$  and  $k_{eff2}$  are the neutron multiplication factor for the rod out and in respectively. With this parameters it is possible to characterise the importance of a given control rod.

For these calculations it was used the 238-group ENDF/B-VII cross-section library, available in the SCALE6.0 package. The problem dependent cross-sections calculations were made by the codes CENTRM/PMC (Continuous Energy Transport Module/Pointwise Multigroup Cross-sections).

#### 3. Results and analysis

Table 1 shows the results in terms of the neutron multiplication factor with the respective deviation in each case calculated by KENO-VI and MCNPX, along with the reactivity of the core and the reactivity worth of each control rod.

	KE		MCNPX			
	k <sub>eff</sub>	ρ (pcm)	$\sigma_{p}$	k <sub>eff</sub>	ρ (pcm)	$\sigma_{p}$
All rods out	1.01859 ± 0.0021	1825.1	203.4	1.01891	1855.9	46.2
Regulator control rod	1.01476 ± 0.0027	-370.5	470.4	1.01523	-324.9	92.8
Control control rod	0.99829 ± 0.0025	-1996.4	453.2	0.99685	-2141.1	97.6
Safety control rod	0.99629 ± 0.0024	-2197.5	443.1	0.99832	-1993.4	95.4

Tab 1: Values of  $k_{eff}$  and  $\rho$ 

The standard deviation for the excess reactivity and the reactivity worth were calculated by the partial derivative method.

A comparison between the two methods and the experimental data available is displayed in Table 2. The differences between the methods and experimental values are shown in Table 3.

	KENO-VI	MCNPX	Experimental
All rods out	1825 ± 203	1856 ± 46	1825
Regulator control rod	-371 ± 470	-325 ± 93	-411
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Control control rod	-1996 ± 453	-2141 ± 98	-2410
Safety control rod	-2197 ± 443	-1993 ± 95	-2212

	Tab 2:	Values	in	pcm	of t	he	reacti	vity
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	KENO-VI	MCNPX
All rods out	0	31
Regulator control rod	40	86
Control control rod	414	269
Safety control rod	15	219

Tab 3: Comparison of the methods with the experimental values

Deviations in reactivity values are very large in every calculation, even though deviations in the k-eff are less than 0.3%. Nevertheless, the reactivities obtained are close to the experimental ones and there is virtually no difference between the excess reactivity of the core measured and calculated by KENO-VI. Slightly larger discrepancies in the control rods results can be observed, but still they have a good approximation.

#### 4. Conclusions

The model built in KENO-VI shows a good concordance with the ones calculated by MCNP and measured experimentally. A development of this model can be achieved by changing the calculation parameters and adding geometry elements that were left either because of lack of source or for simplification of the code. After refining this model, the results obtained can be used in other works in order to give not only parameters values but also their characterization.

In tandem with the validation of this model, a method to generate collapsed and homogenized cross-section libraries by two energy groups from a continuous energy library, using the NEWT sequence in SCALE code, is being developed. NEWT provides lattice physics parameters for the Purdue Advanced Reactor Core Simulator (PARCS) code. These parameters could be, then, applied in 3D diffusion codes for coupled transient calculations (RELAP/ PARCS).

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# INVESTIGATING THE ORIGIN OF ART-HISTORICAL LIMESTONE VIA NEUTRON ACTIVATION ANALYSIS

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# ABSTRACT

Preserved stones of antique ruins and ancient objects give historians and archaeologists insight into life in former times. Knowledge of the origin of a stone allows information to be gained on old trade and transport routes, as well as providing indications of the historical relevance of the object itself. A useful technique to shed light on this is the Instrumental Neutron Activation Analysis (INAA), which is a classical application for research reactors, such as at the TRIGA Mark II of the University of Mainz. INAA is a quasi non-destructive, multi-element analysis method in combination with a high precision. The aim of this work is to find characteristic trace element concentrations ("fingerprints") in the deposits of historical objects. Furthermore, the identified fingerprints will help to relate an object to its primary deposit, so that questions about the historical context can be answered.

In the first part of the project, the contamination of samples due to the abrasion of the drill head for taking samples was analysed. Other problems are inhomogeneity, contamination or general changes of the surface caused by handling and storage of the stone over the centuries. Using the example of arthistorical limestones, the possible contamination effects of different drilling units were systematically analysed via INAA. At the same time the homogeneity of the stones was proved and the composition of the limestones was compared. This will be the basis for further investigations. First analyses of a recently sampled quarry in France are in progress.

# 1. INTRODUCTION

Since the history of many archaeological materials and their origin are unknown, INAA is of interest to museums, art historians and archaeologists. The compositional characterisation of archaeological materials by INAA can be used in conjunction with stylistic and petro-graphic<sup>1</sup> criteria to infer the geographic origin and the attributes of the material. Compositional data answers questions about groups of material belonging together, since a common origin relating materials of unknown origin to a region or a quarry that may have furnished the raw material for its production, gives information about trade relations in the archaeological time period [2]. INAA is one of the best methods used presently for the analysis of many archaeological materials for several reasons: its precision and accuracy allows for the testing of very small samples, it determines many elements simultaneously and its sensitivity permits the determination of constituents present in very small concentrations.

The actual project aims to determine the origin, through analysis, of spoils produced by limestone from the Roman period in Rhineland-Palatinate and which were used previously in buildings and later re-used, for example, for grave stones. First investigations are focused on sampling, because for origin analysis, it is necessary to collate as many trace elements as possible to receive a high accuracy for the data analysis. In this respect, a considerable difference between the results of sampling by drilling and sampling by breaking off pieces of the material was observed – that suggests a contamination by the drill. Of course, valuable and rare archaeological objects could only be sampled by less-destructive methods such as drilling at non-visible locations in the object. Various drills were investigated to find the best way of sampling with the lowest risk of contamination.

In a further step, the feasibility of origin determination using INAA has to be tested. The necessary conditions are homogeneity within every spoil – just as the homogeneity of the quarry where the material of the spoil had once been gathered from. Also, a good distinctness of different objects of different origin or quarries is naturally imperative. For this purpose a quarry in Metz, France [3], has already been sampled in 2010. The measurements on these samples are currently in progress.

# 2. METHOD

Two ancient stones were sampled by drilling at non-visible sites to determine the concentration of the elements – the gravestones "Maura" and "Randoaldo". First, suitable drill bits were selected to sample archaeological objects with a low risk of contamination. Due to conservation reasons, the damage to the archaeological objects has to be minimised – quarrying out pieces of the samples is forbidden, even if this method would cause no contamination of the sample material. In respect to sampling the limestone, different drill bits were tested to evaluate the effect of contamination caused by abrasion – a core drill, a carbide drill and a masonry drill bit were analysed. Finally, a drill bit using a high purity tungsten electrode was used to reduce possible contamination to a single element.

The INAA was carried out at the TRIGA Mark II reactor of the University of Mainz. Irradiations were performed in the steady state mode of the reactor at a power of 100 kW using the pneumatic transfer system with  $\Phi = 1.6 \times 10^{12} \text{ n/(cm}^2 \text{ s})$  or the rotary specimen rack with  $\Phi = 0.7 \times 10^{12} \text{ n/(cm}^2 \text{ s})$ . For a quantitative analysis appropriate standards were used – for example, the certified stone GBW 07113, the inorganic marine sediment NIST SRM 2702, the reference material BCR-142R and miscellaneous liquid standards. The samples are analyzed using  $\gamma$ -spectroscopy with a high-purity germanium detector (HPGe) and evaluated using special software.

<sup>&</sup>lt;sup>1</sup>Petro-graphic and geochemical methods used for origin analyses of limestone from the Rhine-Main area are described in the work of Stribrny [1].

1 min-irradiation	6 h-irradiation
Al, Ca, Mg, Mn, Sr, Ti, V	As, Ba, Ce, Cs, Co, Cr, Eu, Fe, Hf, K, La, Lu, Na,
	Nd, Rb, Sb, Sc, Sm, Ta, Tb, Th, U, W, Yb, Zn

Tab. 1: List of detectable elements for different irradiation times

For each sample, three measurements are conducted to detect and exclude a possibly undesirable contamination by natural inclusions in the sample material. The sample masses, in PET vials, are 10-20 mg each for the short-time irradiation and 150-200 mg each for the long-time irradiation. The exposures were performed at a reactor power of 100 kW<sub>therm</sub>. For the short-time irradiations for studying the trace elements with short-lived activation products, the samples were irradiated using the pneumatic transport system (one-minute irradiation). For the long-time exposures, an irradiation time of six hours is chosen, in which the reactor's carousel is used to ensure a constant neutron flux by rotating. The samples of the short-time irradiation were measured for a time period of 10 min after a decay time of two minutes and for 30 minutes after 45 minutes. The samples of six-hour irradiation were measured after two to three days for one hour and after two to four weeks for eight hours. The time-delayed analysis is necessary to be able to determine isotopes with similar energies and different halflives. A total of 32 elements can be detected (Tab. 1).



Fig. 1: Gravestone "Randoaldo" sampled by different drill bits



Fig. 2: Sampling by the tungsten drill

# 3. RESULTS

At first, the three commercially available drills mentioned above were tested. Initially, two early medieval gravestones were sampled to perform comparative measurements and to analyse potential contamination. The "Maura's gravestone" was sampled with the carbide and the masonry drill bit, the "Randoaldo" with the carbide and the core drill.

In the investigation on "Randoaldo", irregular values were found for the same object sampled by different drill bits. This was with regard to several elements, so it can be suggested to be a drill-related contamination. The size of this contamination is not constant, which indicates a variable abrasion of the drill. A comparison of the core drill and the carbide drill is shown in (Fig 1).

A similar situation emerges when comparing the carbide and the masonry drill bit on the basis of Maura's gravestone. While some elements (Mn, K, As) have comparable levels of concentrations in all sampling holes, this does not apply to other elements (W, Zn, Hf, Ti), which confirms again the assumption that the drill bits cause contamination by abrasion. Due to the reason that the contamination is not constant, an evaluation of these elements in the samples is not possible. Thus, none of the actual drills is likely to obtain evaluable samples.

In addition to the investigation of the drill bit contamination, it could be shown that the Randoaldo gravestone is, in fact, not even limestone, as declared beforehand.

	core drill bit	carbide drill bit	masonry drill bit	tungsten elecctrode
AI	Х			
Со		Х		
Cr	Х			
Cs			Х	
Fe	Х	Х	Х	
La	Х			
Mn	Х	Х	Х	
Та	Х	Х	Х	
Ti	Х		Х	
۷	Х			
W	Х		Х	Х
Zn	Х			

Tab. 2: Possible contaminations by different drill bits

To minimize the contamination of different drill bits, a drill bit was fabricated from a pure tungsten electrode, such that the contamination should be limited to tungsten only. The powder from the sampling with the tungsten drill and a broken piece of the sampled object were analysed. The sampled object was a fragment of pillar of a grave monument near Neuwied.

A small piece was broken off from the pillar fragment (Neuw I-1). Immediately adjacent to the breaking edge, two test holes were made with the tungsten drill (Neuw I-2, Neuw I-3), to minimize the risk of a possible inhomogeneity within the limestone.

The studies show that the tungsten drill causes only a tungsten contamination and that the other elements are not detectably affected (Fig. 2). Hence, with this method all measurable elements except tungsten are available for evaluation. In Table 2, possible contamination of the sampled material is summarised.

Using the tungsten drill, the issue of homogeneity within a larger object of limestone should now be feasible. For this, the fragment of pillar from Neuwied and a spoil from the foundation of a Roman bridge in Trier were sampled at different points.

The analysis shows a largely homogeneous elemental composition within the particular findings, but significant differences in the lanthanide concentration of the two stones are observed (Fig. 3).



Fig. 3: Comparison of the homogeneity of the samples from Neuwied and Trier

### 4. SUMMARY AND PERSPECTIVE

The first studies with various drills demonstrate that the contamination, caused by abrasion during the sampling, poses difficulties. To reduce the contamination to a minimum of one single element, a modified tungsten electrode was used as drill bit. As a consequence, many elements become available for evaluation. Furthermore, it could be shown by means of comparing two different limestone objects, that the homogeneity within spoils can be proved, which is an important basis for further investigations. Finally, by comparing the two examined spoils from Neuwied and Trier, concerning the rare-earth elements, a first reference about the chemical distinctness of different limestones was found. These studies are the basis for

further research via NAA regarding the origin of ancient limestone spoils. In the next step, the homogeneity of quarries and the distinctness within a small geographical area should be determined. A first sampling of a quarry in France was accomplished and measurements are in progress. More samples of limestones will be taken soon. For statistical analysis, multivariate methods will be applied to deal with the high amount of data.

A long-term objective is to build up a database of the results and combine it with an existing database at the Metropolitan Museum of Art in New York [4], with the intention to have a good availability of data for archaeologists and art-historians. It should assist them to discover ancient trade connections and facilitate on questions of limestone origin.

It is also planned to initiate an international co-operation with the University of Utah (Salt Lake City, USA).

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### DETERMINATION OF THE NEUTRON FLUX DENSITY DISTRIBUTION INSIDE THE CENTRAL EXPERIMENTAL TUBE OF THE TRIGA MAINZ

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#### Abstract

Related to the analysis of 3d-metals in rods of directionally solidified silicon for the solar cell production, the relative neutron flux density in the vertical direction was determined for the central experimental tube of the TRIGA Mark II reactor at the Johannes Gutenberg-University in Mainz. The standard irradiation capsule was modified to achieve a fixed irradiation position. Nickel and iron were used as flux monitors. After normalisation of the activities, a profile of the neutron flux for thermal and epithermal/fast neutrons inside the central experimental tube is obtained.

### 1. INTRODUCTION

One important aim of the photovoltaic industry is the further reduction of manufacturing costs for solar cells. Silicon feedstock and wafering with their part of about 40% of the overall costs provides a good possibility to save expenses. An economic alternative to the mostly used high purified silicon can be given to so-called solar grade silicon (SoG-Si). SoG-Si is defined as silicon with acceptable purity grades, especially of the 3d-transition metals, which act as traps for charge carriers and form recombination centres and thus reduce the solar cell efficiency. In order to investigate different purification procedures for SoG-Si, such as directional solidification, Neutron Activation Analysis (NAA) is used to determine the 3d-metal content of solar grade silicon.

When investigating samples with dimensions that cannot be neglected in terms of varying neutron fluxes, the distribution of the neutron flux for a fixed irradiation position has to be determined beforehand. For the case here, the neutron flux distribution inside the central experimental tube of the TRIGA Mark II in Mainz was measured, as this is where the concentration profile of 3d-metals and arsenic in rods of directionally solidified silicon for solar applications [1-3], will be determined using Instrumental Neutron Activation Analysis (INAA).

To monitor the distribution of thermal neutrons, the reaction  ${}^{58}Fe(n,\gamma){}^{59}Fe$  of iron with thermal neutrons was used. Nickel and its  ${}^{58}Ni(n,p){}^{58}Co$  reaction was used to determine the distribution of the epithermal and fast neutrons.

# 2. MATERIAL

Irradiations are performed at the research reactor TRIGA Mark II at the Johannes Gutenberg-University in Mainz [4]. In the steady state mode, the reactor can be operated at power levels ranging from about 100 mW<sub>th</sub> up to 100 kW<sub>th</sub>, depending on the requirements of the different experiments. Pulse-mode operation is also possible. For in-core irradiations, the TRIGA Mainz has a central experimental tube, three pneumatic transfer systems and a rotary specimen rack with 40 positions which allows the irradiation of 80 samples at the same time.

Samples for monitoring the flux are high purity nickel and iron wire from *Goodfellow Cambridge Limited* with a diameter of 0.25 mm and a purity of > 99.99%. The detection system consists of a high purity germanium (HPGe) gamma-ray detector with an embedded pre-amplifier from Canberra Ind., surrounded by a heavy lead shielding. Recording and evaluation of the gamma-ray spectra is carried out using the program Genie2000, also from Canberra Ind. To increase operational capacity in measurement, a sample changer capable of handling up to 20 samples is used.

# 3. EXPERIMENT

With the aim to provide a fixed and reproducible irradiation position, the standard irradiation capsule for the central experimental tube was modified to stand on the base plate of the reactor core and thus fixed in the vertical direction. The cavity inside the capsule is designed to extend across the whole fuel matrix of the fuel element, where the neutron flux is expected to be highest. An illustration of the positioning of the irradiation capsule inside the core is shown in Fig. 1.

The high purity iron and nickel wire was cut into pieces of approx. 3.9 mg and 4.4 mg, respectively. All pieces were shrink-wrapped in strips of PE-foil, with a distance between each other of 10 mm. These strips were mounted on a PE-tube, which was shrink-wrapped again before it was put into the irradiation capsule. A nylon string was affixed to the capsule to lift it up after the end of the experiment.



Fig. 1: sketch of the reactor core and the irradiation capsule fixed by the

Irradiation of the samples was performed in the central experimental tube at a reactor power of 100 kW for 30 min. After irradiation and a decay time of approximately 15 h, all samples were repacked separately for later measurement via gamma-ray spectrometry.

For means of standardisation of the measured specific activities generated by Genie2000, each activity is divided by the highest measured activity of the respective dataset. By this, ratios ranging from 0 < RATIO < 1 are obtained. For iron, the weighted average of the activities calculated for the energy lines at 1099 keV and 1291 keV is used.

# 4. RESULTS & DISCUSSION

The relative neutron flux distribution of thermal neutrons is shown in Fig. 2. *Dataset 1* shows an increasing neutron flux density but no clear maximum. Despite a known plan of the reactor core, it seems likely that the modified irradiation capsule was not long enough to extend across the whole fuel matrix. A second irradiation was performed, but for this measurement the irradiation capsule was raised 20 cm to be sure to extend across the whole fuel matrix and even further but also to have an overlap with *dataset 1* for internal control. Results are shown as *dataset 2* in Fig. 2. Data recorded show a similar trend compared to *dataset 1*, but a displacement in height. The same behaviour could be observed for the measurement of the epithermal/fast neutron flux density, as measured via <sup>58</sup>Ni(n, p)<sup>58</sup>Co.



Fig. 2: Relative neutron flux density of thermal neutrons measured with the reaction  ${}^{58}\text{Fe}(n,\gamma){}^{59}\text{Fe}$  as a function of the distance from the bottom plate of the reactor core

There could be a few possible explanations for the observed displacement in height:

- a displacement of the inner sample holder inside the irradiation capsule
- the capsule slipped down before it could be fixed after it was lifted up
- an elongation of the nylon-string during the second irradiation
- a fluctuation of the neutron flux density distribution

Nevertheless, it could be shown that the sample cavity inside the irradiation capsule can be designed to fix the sample holder in the vertical direction. Therefore, displacement of the inner sample holder is very unlikely. Also during the performance of the experiment, the core itself was unchanged and the irradiation was done in the same way. There is no obvious reason for an alteration of the neutron flux to cause the measured displacement of *dataset 2*.

A third measurement was performed to investigate possible slipping of the capsule and a possible elongation of the nylon-string. The data of the third measurement is also shown in Fig. 2 as *dataset 3*. As for the second dataset, there is a displacement in height. A similar behaviour of the epithermal/fast neutron flux density was observed. Furthermore, the third measurement has proven that an elongation of the nylon string by 1 cm occurred, but this only can serve as an estimate. The elongation probably is underestimated, because the

freely floating capsule will stretch the string even more. A slipping of the irradiation capsule can be excluded.

The characterisation of the central experimental tube has shown that the position of the irradiation capsule will vary unless the capsule is fixed in height. To ensure a constant and reproducible irradiation geometry for future irradiation, a newly designed capsule should stand on top of the bottom plate of the reactor core, and thus is fixed in height.



Fig. 3: Relative neutron flux density of thermal neutrons measured with the reaction  ${}^{58}\text{Fe}(n,\gamma){}^{59}\text{Fe}$  as a function of the distance from the bottom plate of the reactor core. *Dataset 2* is shifted by  $\Delta h = -3$ ;  $\Phi_{exp}(h)$  being the fit of *dataset 1* and 2

To be able to adjust the measured activities along the silicon rod according to the neutron flux distributions of *dataset 1* and *dataset 2*, the relative neutron flux density of thermal neutrons is shifted by  $\Delta h = -3$  (see Fig. 3), and fitted with the program Eureqa [5] to give the function  $\Phi_{exp}(h)$  shown below:

$$\Phi_{exp}(h) = 0.46439853 + 0.50553173 * \cos(2.3672993 + 0.81167862 * h) - 0.097538292 * \cos(3.444473 + 0.084239304 * h) * \cos(-0.11537161 * h - 2.8771236)$$

Knowing the position of samples or sample segments relative to the core base plate and the associated neutron flux, it is possible to normalise measured activities according to the neutron flux distribution inside the central experimental tube.

## 5. SUMMARY

After the performed experiments, the central experimental tube of the TRIGA Mainz has been characterised according to the profile of the thermal and epithermal/fast neutron distributions. This knowledge is fundamental regarding further measurements of rods of directionally solidified silicon for solar applications. Quantitative measurements of activity and consequently concentration are now possible even for samples with dimensions that cannot be neglected in terms of varying neutron fluxes.

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