THE IEA-R1 RESEARCH REACTOR: 50 YEARS OF OPERATING EXPERIENCE AND UTILIZATION FOR RESEARCH, TEACHING AND RADIOISOTOPES PRODUCTION

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ABSTRACT

This paper describes almost 50 years of operating experience and utilization of the IEA-R1 research reactor for research, teaching and radioisotopes production. The current and future program of upgrading the reactor is also described. IEA-R1 research reactor at the Instituto de Pesquisas Energéticas e Nucleares (IPEN), Sao Paulo, Brazil is the largest power research reactor in Brazil, with a maximum power rating of 5 MWth. It is being used for basic and applied research in the nuclear and neutron related sciences, for the production of radioisotopes for medical and industrial applications, and for providing services of neutron activation analysis, real time neutron radiography, and neutron transmutation doping of silicon. IEA-R1 is a swimming pool reactor, with light water as the coolant and moderator, and graphite and beryllium as reflectors. The reactor was commissioned on September 16, 1957 and achieved its first criticality. It is currently operating at 3.5 MWth with a 64-hour cycle per week.

In the early sixties, IPEN produced ^{131}I, ^{32}P, ^{198}Au, ^{24}Na, ^{35}S, ^{51}Cr and labeled compounds for medical use. In the year 1980, production of ^{99m}Tc generator kits from the fission ^{99}Mo imported from Canada was started. This production is continuously increasing, with the current rate of about 16,000 Ci of $^{99m}T_C$ per year. The ^{99m}Tc generator kits, with activities varying from 250 mCi to 2,000 mCi, are distributed to more than 260 hospitals and clinics in Brazil. Several radiopharmaceutical products based on ^{131}I , ^{32}P , ^{51}Cr and ^{153}Sm are also produced.

During the past several years, a concerted effort has been made in order to upgrade the reactor power to 5 MWth through refurbishment and modernization programs. One of the reasons for this decision was to produce ^{99}Mo at IPEN. The reactor cycle will be gradually increased to 120 hours per week continuous operation. It is anticipated that these programs will assure the safe and sustainable operation of the IEA-R1 reactor for several more years, to produce important primary radioisotopes ^{99}Mo, ^{125}I, ^{131}I, ^{153}Sm and ^{192}Ir. Currently, all aspects of dealing with fuel element fabrication, fuel transportation, isotope processing, and spent fuel storage are handled by IPEN at the site. The reactor modernization program is slated for completion by 2009.

1. INTRODUCTION

A nuclear reactor is a strong neutron source for both thermal and fast neutrons and can be efficiently used for the production of radioisotopes with numerous applications in medicine, agriculture, and industry and for other irradiation services. It could also be used for academic and applied research in the areas of nuclear and neutron related sciences and engineering. The extent of the utilization of the reactor is basically determined by the reactor power, which determines the neutron flux available, as well as on its operational cycle. Small power levels and short operational cycles (only few hours of operation each day) are particularly inconvenient, both for high quality research and also for the production of useful radioisotopes. High power levels, in conjunction with a prolonged operational cycle (several weeks continuous operation), on the other hand result in high fuel consumption and require an expensive maintenance program.

The Brazilian research reactor IEA-R1 at IPEN, São Paulo, with a maximum power rating of 5 MWth is being used for basic and applied research in the nuclear and neutron related sciences and for the production of radioisotopes for medical and industrial applications. The reactor is also used for providing services of neutron activation analysis, real time neutron radiography, and neutron transmutation doping of silicon. In order to achieve the goals of reactor upgrading for safe and sustainable operation for radioisotopes production, IPEN undertook a major task of reactor system modernization, under a technical cooperation program funded by the International Atomic Energy Agency (IAEA) and Brazilian government agencies [1].

Brazil has four operational research reactors, all under the responsibility of Brazilian National Nuclear Energy Commission (CNEN). Some details of the characteristics of these reactors are summarized in Table 1. IEA-R1 is the only research reactor in Brazil operating at a substantial power level suitable for its utilization in wide areas of research such as physics, chemistry, biology, and engineering, as well as in the production of some useful radioisotopes for medical and other applications. The general features of this reactor, its past operational experience and current utilization, are reviewed briefly. The future plans to optimize its performance in the areas of research and development, specifically its role as a radioisotope producer, are described.

Table 1. Brazilian Research Reactors

	IEA-R1	IPR-R1	ARGONAUT	IPEN/MB-01
Criticality	September 1957	November 1960	February 1965	November 1988
Operator	IPEN-CNEN/SP	CDTN-CNEN/MG	IEN-CNEN/RJ	IPEN-CNEN/SP
Location	São Paulo	Minas Gerais	Rio de Janeiro	São Paulo
Type	Swimming Pool	Triga Mark I	Argonaut	Critical assembly
Power Level	2-5 MW	250 kW	200 W	100 W
Enrichment	20%	20%	20%	4.3%
Supplier	Babcock & Wilcox	General Atomics	USDOE	Brazil

2. IEA-R1 RESEARCH REACTOR

IEA-R1 is the largest power research reactor in Brazil, with a maximum power rating of 5 MWth. The reactor was commissioned on September 16, 1957 and achieved its first criticality. Although designed to operate at 5 MW the reactor operated only at 2 MW during the early sixties and mid-eighties on an operational cycle of 8 hours a day, 5 days a week. It is currently operating at 3.5 MWth with a 64-hour cycle per week. The reactor originally used 93% enriched U-Al fuel elements. It currently uses 20% enriched uranium (U_3O_8-Al and U_3Si_2-Al) fuel that is produced and fabricated at IPEN [2,3]. The reactor is operated and maintained by the Research Reactor Center (CRPq) at IPEN, São Paulo, which is also responsible for irradiation and other services.

The IEA-R1 reactor is a multidisciplinary facility and is being used extensively for basic and applied research in nuclear and neutron related sciences and engineering. The reactor has also been used for training, for producing some useful radioisotopes with applications in industry and nuclear medicine, and for miscellaneous irradiation services. Several departments of IPEN routinely use the reactor for their research and development work. Many scientists and students at universities and other research institutions in Brazil also use it quite often for academic and technological research. However, the largest user of the reactor is the staff of the Research Reactor Center with interest in basic and applied research in the areas of nuclear and neutron physics, nuclear metrology, and nuclear analytical techniques.

In the early sixties, IPEN produced ^{131}I, ^{32}P, ^{198}Au, ^{24}Na, ^{35}S, ^{51}Cr radioisotopes and labeled compounds for medical use. After 1980, IPEN started producing ^{99m}Tc generator kits from the fission ^{99}Mo imported from Canada. This production is continuously increasing, with the current rate of about 16,000 Ci of $^{99m}T_C$ per year. The ^{99m}Tc generator kits, with activities varying from 250 mCi to 2,000 mCi, are distributed to more than 260 hospitals and clinics in Brazil. Several radiopharmaceutical products based on ^{131}I, ^{32}P, ^{51}Cr and ^{153}Sm are also produced at IPEN.

During the past few years, a concerted effort has been made in order to upgrade the reactor power to 5 MWth. One of the reasons for this decision was to produce ^{99}Mo at IPEN, thus minimizing the importation cost and reliance on only one or two world suppliers. The reactor cycle will be gradually increased to 120 hours per week continuous operation [2, 3]. The reactor component refurbishment plans include the following during 2003-2009 [1]:

- Replacement of pool water treatment and purification system.
- Replacement of four reactor control elements.
- Replacement of primary heat exchanger.
- Installation of a new core grid plate.
- Replacement of the control panel.

3. NEUTRON IRRADIATION AND OTHER SERVICES

CRPq is making enormous effort to enlarge the scope of services and applications resulting from reactor utilization so that more benefits of these applications could be offered to the society. Some of the products and services offered by the Reactor Center find their way to petroleum industry, aeronautical and space industry, medical clinics and hospitals, semiconductor industry, environmental agencies, universities and research institutions. The reactor produces special radioisotopes such as ^{41}Ar and ^{82}Br for industrial process inspection, ^{192}Ir and ^{198}Au radiation sources used for brachytherapy, ^{153}Sm (EDTMP) for pain palliation in bone metastases, calibrated gamma sources of ^{133}Ba, ^{137}Cs, ^{57}Co, ^{60}Co, ^{241}Am and ^{152}Eu used in clinics and hospitals practicing nuclear medicine and research laboratories. Regular services of non-destructive testing by real-time neutron radiography, multi-element trace analysis and miscellaneous neutron irradiation of samples for research applications are also offered. Regular irradiations are carried out to produce some primary radioisotopes for the Radiopharmaceuticals Center at IPEN.

Neutron irradiation of silicon single crystals for doping with phosphorus was developed at IPEN in the early 1990's. A simple device for irradiating silicon crystals with up to 12.7-cm diameter and 50-cm long, located in the graphite reflector, was installed in the reactor for commercial irradiation. Details about the design of the irradiation rig and its performance may be found elsewhere [4].

4. PRODUCTION OF RADIOISOTOPES

Until 1980 all the ^{99m}Tc generators used in the country were imported. Due to the rapidly increasing demand and high cost of importation, the Radiopharmaceuticals Center started to make its own ^{99m}Tc generator kits with automatic elution system using fission ^{99}Mo purchased from Canada. Today ^{99}Mo-^{99m}Tc generators in the form of several kinds of radiopharmaceuticals, represent more than 80% of all the radioisotopes distributed to hospitals and clinics in the country. In 2006, more than 16,000 Ci of ^{99m}Tc generators were produced, with individual kit activity ranging from 250 mCi to 2,000 mCi and distributed to about 260 hospitals and clinics all over the country, benefiting more than 2,500,000 patients. In addition to ^{99m}Tc generators, the Radiopharmaceuticals Center also makes and distributes radioactive preparations for medical use based on ^{131}I (1,500 Ci per year), as well as smaller quantities of ^{51}Cr (2 Ci per year), ^{32}P (3 Ci per year). The shorter-lived radioisotope ^{153}Sm (40 Ci per year) is regularly produced in IEA-R1 and distributed in the form of ^{153}Sm (EDTMP) for treatment as pain palliation in bone metastases. These reactor produced radioisotopes, in addition to some of the cyclotron produced radioisotopes such as ^{67}Ga, ^{201}Tl and ^{18}F and radiopharmaceuticals used in nuclear medicine, represent annual receipts from sales of the order of 15-16 million US dollars for IPEN (estimates for the year 2006).

The importation costs for the reactor-produced primary radioisotopes such as ^{99}Mo and ^{131}I on the other hand are of the order of 5-6 million US dollars. Recent survey has shown that the demand for ^{99m}Tc generators and ^{131}I preparations is steadily increasing at the rate of 8% and 25% a year, respectively and expected to continue increasing in the years to come. In order to meet continuously increasing demand of these radioisotopes in Brazil, IPEN will have to face increasing importation costs easily reaching 6-8 million US dollars in just a couple of years. This is quite cumbersome for an institution like IPEN whose main funding comes from federally approved budget. Importation cost factor aside, increasing reliance on practically one or two foreign suppliers of vital radionuclides such as ^{99}Mo, is certainly a strategic disadvantage for the country.

Considering these factors, as well as the possibility of producing in the IEA-R1 reactor, radioisotopes such as ^{99}Mo, ^{192}Ir, ^{131}I, and ^{125}I among others, an important decision was made by the higher management a few years ago to upgrade the reactor power to 5 MW and to gradually increase the operational cycle to 120h continuous per week. Initial plans to produce ^{99}Mo from fission were given up principally due to lack of necessary funds required but also due to technical problems associated with complex radiochemical processing and the management and storage of highly radioactive waste generated. It was decided to produce ^{99}Mo by ^{98}Mo(n,γ)^{99}Mo reaction using natural Mo target and ^{99m}Tc generator using gel process. In order to achieve the goal of producing high specific activity sources of ^{99}Mo, resulting in the ^{99m}Tc generators in the range of 250mCi to 1,000 mCi activities, from IEA-R1, it was found necessary to raise the reactor power to 5 MW and to adopt an operational cycle of 120h continuous per week. Four main areas, received particular attention: 1) optimization of reactor systems, structures and components, 2) optimization of reactor fuel elements production, 3) optimization of radiochemical facilities using gel process, and 4) an effective program for spent fuel management. IPEN assigned definite priorities for these projects at the institutional level.

5. REACTOR UPGRADE AND MODERNIZATION

During the last several years many changes in the reactor system and components have been made in an effort to extend the lifetime of the reactor and secure its safe and sustainable operation. IEA-R1 is one of the oldest reactors of its kind in the world and has been operating for nearly 50 years with excellent safety records. Some of the important improvements made in the reactor systems during the last several years are:

- Modification of the reactor core from 6x5 to 5x5 using LEU fuel elements.
- Installation of a central beryllium irradiating element
- Replacement of 10 graphite reflectors with beryllium reflectors.
- Installation of four isolation valves in the primary cooling system.
- Repairs in the cooling tower and pipelines.
- Installation of a new ventilation and air conditioning system.
- Improvement in the control instrumentation.
- Replacement of the old radiation monitoring system.
- Installation of an emergency spray core cooling system.

With these modifications introduced in the reactor, a new safety analysis report was prepared and submitted to the regulatory body. An authorization from the regulatory body was obtained in the September 1997 for commissioning of the reactor at 5 MW. The reactor operated at 5 MW for six months. However, since other projects like optimization of fuel element production, chemical processing facilities for ^{99m}Tc using gel process, and spent fuel storage facility had not yet been implemented to full extent, the reactor power was reduced back to 2 MW. The reactor has since been operating at 2 MW on a continuous 64h per week cycle. The reactor power was increased gradually and attained 3.5 MW in 2001. During the course of 1998 to 2004, substantial progress was made to implement the reactor fuel production program. The Fuel Fabrication Center acquired the know-how and capacity to produce 15-18 fuel elements of the type U_3Si_2 (3.0 g/cm^3) per year. At the same time a chemical method for producing ^{99m}Tc generator by gel process was developed implemented. In the 1999, all the spent fuel elements stored in the reactor pool since its first criticality (a total of 127 elements) were transferred to USA under a bilateral agreement (DOE-IPEN/CNEN). Transfer of an additional batch of 40 spent fuel elements to USA has been negotiated and will take place in 2007 under the same agreement.

The reactor pool at present has storage space for more than 130 spent fuel elements. The available pool storage space should be sufficient for about 7-8 years of reactor operation at 5 MW: 120h/week. The fuel consumption is estimated to be 15-16 elements per year in this regime of operation. A new project for spent fuel management and storage was initiated in 2001 at IPEN to investigate the possibility of an alternate dry storage space. This activity received active support from IAEA in the form of a regional project. Prototype of a cask for dry storage of 21 spent fuel elements will be constructed and tested soon. Aging management and refurbishment program for the IEA-R1 reactor components and systems is a continuous and on-going activity of the Research Reactor Center. For example, the reactor pool water treatment and purification system was replaced in 2004. The older control and safety elements of the reactor began to show signs of aging and were replaced in 2004 with identical elements, (fork type Ag-In-Cd) fabricated at IPEN. The infrastructure modernization efforts include the following [1]:

- Modernization of the reactor fuel element fabrication facility in order to increase the production capacity to 15-18 U_3Si_2 type fuel elements per year.
- Optimization of radiochemical facilities to process ^{99}Mo using the gel process.
- Development of an effective project for spent fuel management and storage.

It is anticipated that these programs will assure the safe and sustainable operation of the IEA-R1 reactor for several more years, to produce important primary radioisotopes ^{99}Mo, ^{125}I, ^{131}I, ^{153}Sm, ^{177}Lu, ^{188}Re and ^{192}Ir. The production of these isotopes will result in less dependence on the world supply, and reduce importation costs. Routine radiation monitoring is carried out at 25 different locations in the reactor building. This regular monitoring practice has helped in maintaining the equivalent dose of reactor workers below the established limits [5]. Following the guidelines of IAEA an ALARA program is being implanted at the reactor and associated laboratories, which handle radioactive materials.

The Research Reactor Center had a Technical Cooperation (TC) project BRA/04/056: “Modernization of the IEA-R1 research reactor to secure safe and sustainable operation for radioisotope production,” supported by IAEA during 2005-2006. The project contemplated several training programs for the reactor operation and maintenance personnel as well as improving the technical infrastructure of the reactor. Some of the goals achieved through the TC project were:

- Replacement of some electrical and refrigeration systems; radiometric analysis system for water and air samples; reactor control instrumentation; radiation monitoring equipment.
- Neutron flux mapping facility using self-powered neutron detectors (SPNDs).
- Improved computational facility for neutronic calculations.
- A highly radioactive sample handling facility.
- Training of personnel engaged in electrical and mechanical maintenance, water chemistry, and irradiation services.
- Installation of a continuous vibration monitoring system for rotating machinery.

The rotating machinery in the IEA-R1 reactor system is primarily the water circulating pumps. As part of the reactor upgrade plan, a continuous vibration monitoring system has been installed. This will provide accelerometer data to a central processing unit that will monitor the changes in the vibration levels of the pump-motor system. Defects such as imbalance, misalignment, looseness, and bearing faults can be detected before a catastrophic failure occurs. Thus, incipient fault detection and diagnosis of rotating machinery will be an important feature of this upgrade. Currently, all aspects of dealing with fuel element fabrication, fuel transportation, isotope processing, and spent fuel storage are handled by IPEN at the site. Spent fuel assemblies are visually inspected routinely using underwater cameras. Seeping analysis is performed if there is an indication of fission product release in the pool water.

6. CONCLUDING REMARKS

The reactor modernization program, introduced several years ago at IPEN, and its effective implementation during all these years with solid investments, will guarantee safe and continuous operation of the reactor to achieve the goal of producing some of the important primary radioisotopes such as ^{99}Mo and ^{131}I among others in the IEA-R1 reactor [1,2]. Important economy in the foreign exchange is expected to result from the partial substitution of importation. Estimated figures are: US$400,000-600,000 for ^{99}Mo, US$400,000-500,000 for ^{131}I, and US$50,000-80,000 for ^{192}Ir. Reactor operation under new conditions will also permit the production of ^{125}I seeds (which are at present imported) used for the treatment of prostate cancer.

As a consequence of increased neutron flux available (maximum thermal neutron flux of 2×10^{14} n/cm^2-s at 5 MW power) and extended operation period, other applications and services such as silicon doping with phosphorus by neutron irradiation, neutron radiography, and neutron activation analysis will have better performance. The improved operational regime of the reactor will stimulate renewed interests in other applications, which are currently in experimental stages, such as boron neutron capture therapy (BNCT) and coloration of topaz. Neutron beam research will benefit due to availability of more intense neutron beams. A viability study has recently been made for the possibility of installing a low angle neutron scattering (SANS) facility at the reactor. This activity was supported by IAEA through a research contract. It should be emphasized that academic research and postgraduate teaching at the Reactor Center are very important programs in the effective utilization of the reactor. Research scientists, students, and professors from universities and other research institutions and their students have free access to the research facilities at the Reactor Center. The current phase of the reactor modernization program is slated for completion by 2009.

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