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PUBLICAÇÃO IPEN 43 IPEN - Pub - 43	JUNHO/1982

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CENTRO DE OPERAÇÃO E UTILIZAÇÃO DO REATOR DE PESQUISAS - COURP

INSTITUTO DE PESQUISAS ENERGÉTICAS E NUCLEARES SÃO PAULO - BRASIL

# Série PUBLICAÇÃO IPEN

**INIS Categories and Descriptors** 

E36 A30

EXPERIMENTAL REACTORS: IEAR - 1 reactor IEAR - REACTOR: Experimental reactors RESEARCH REACTORS: IEAR - 1 reactor IEAR - 1 REACTOR: Research reactor BRAZIL: IEAR - 1 reactor IEAR - 1 REACTOR: Brazil IEAR - 1 REACTOR: Nuclear Physics IEAR - 1 REACTOR: Radiochemistry

COURP

Aproved for publication in February 1982,

Writing, orthography, concepts and final revision are of exclusive responsibility of the Authors.

Received in October 1981.

# EXPERIENCE AND RESEARCH WITH THE IEA-R1 BRAZILIAN REACTOR\*

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### ABSTRACT

The IEA-R1 reactor of the Instituto de Pesquisas Energéticas e Nucleares, IPEN, of São Paulo, Brazil, a lightwater moderated swimming-pool research reactor of MTR type, went critical for the first time on September 16, 1957. In a general way, in these twenty four years the reactor was utilized without interruption by users of IPEN and other institutions, for the accomplishment of work in the field of applied and basic research, for master and doctoral thesis and for technical development. The present work describes n a general view Some works performed and the renewal programme established for the IEA-R1 research reactor in which several improvements and changes were made. Recent activities in terms of production of radioisotopes and some current research programm in the field of Radiochemistry are described, mainly studies and research on chemical reactions and processes using radioactive tracers and development of radioanalytical methods, such as neutron activation and isotopic dilution. It is also presented the research programmes of the Nuclear Phisycs Division of IPEN, which includes: nuclear spectroscopy studies and electromagnetic hyperfine interactions; neutron diffraction; neutron inelastic scattering studies in condensed matter; development and application of the technique of fission track register in solid state detectors; neutron radioactive capture with prompt gamma detection and, finally, research in the field of nuclear metrology, and productive field of the technique of fission track register in solid state detectors; neutron radioactive traces and the field of nuclear metrology, and application field and application and, finally, research in the field of nuclear metrology.

### I - INTRODUCTION

The IEA-R1, a lightwater moderated swimming-pool research reactor of type MTR, went critical for the first time on September 16, 1957. After surpassing the main problems and difficulties in the initial cycles of operations<sup>(9,14,21)</sup> the reactor has been operating for about twenty years without a single accident with respect to its safety at the Instituto de Pesquisas Energéticas e Nucleares, IPEN (formerly-IEA), of São Paulo, Brazil. The current fuel is uranium enriched at ninety-three per cent in the isotope U-235. The reactor, which was described in details in a previous paper<sup>(18)</sup>, uses graphite reflectors and was designed to operate at a power of five megawatts, but currently has been operating at 2 Mw.

A crew of three to four men permits reactor operation on an eight-hour per day schedule five days per week. The total operating staff consists of seventeen people of which four are graduate supervisors and thirteen are technicians. The team that operates the reactor belongs to the Operation and Maintenance Research Reactor Division (AOMR), which is also in charge of the water treatment and purification, orientation and supervision of the utilization of the irradiation facilities.

In a general way, in these twenty years, the principal aim to be reached with the reactor utilization has been the development of academical research performed mainly by the two Divisions that have their staffs permanently located near the reactor facilities: the Radiochemistry Division (ARQ) and the Nuclear Physics Division (AFN). The setting of the three mentioned Divisions compose the Center of Operation and Utilization of the IEA-R1 Research Reactor (COURP). The Center, which has a total staff of 80 persons, belongs to the Executive Directory I of IPEN. Nevertheless, as a by-product of these research activities it was possible to reach a condition where services to the scientific community and also to industry, can be offered by the Center. The reactor has been used practically without interruption by users of the IPEN, of other Institutes and of industrial organizations as well. The several scientific papers developed in the field of basic and applied research have been used for personnel

Presented at the Seminar On Research Reactor Operation and Use held by International Atomic Energy Agency, in co-operation with the Goverments of the Federal Republic of Germany and the United States of America, from 14 to 18 of September 1981, Jülich, Federal Republic of Germany.

training purposes such as master and doctoral thesis publications. More specifically, since the beginning of the reactor operation, besides sealed sources production for industrial applications, sample irradiations were carried out for the production of radioisotopes to be applied in the IPEN researches and for external supply; neutron activation analysis of several materials were frequently made. A large number of scientific papers were published on the field of Radiochemistry<sup>(3,8,11,17,23)</sup> and also in Neutron Physics<sup>(1,2,5,15,16,2,2,24,25)</sup>, in this case using neutron beams collimated through the reactor beam holes.

### II - OPERATIONAL EXPERIENCE AND PROBLEMS

Besides the reactor maintenance and the setting in compliance with the more recent safety regulatory guide, the renewal and power upgrading programmes of research reactors must aim the obtention of higher and better quality neutron fluxes, in order to attend the evaluation of the fifferent user's needs.

The majority of the world's research reactors, which were designed and built up fifteen or twenty years ago, are still being used; it is abvious that many components had to be replaced since they became obsolete. The development in the control and instrumentation systems were particularly rapid, leading to the greatest "modifications in the existing instalations. Nevertheless, more than the simple change of the individual components through the years, the subjects to be addressed when considering either a new research reactor, the modernization or the upgrading of an existing plant are much wider than simply the selection and design of the modification or extension itself. Ideally, the opportunity should be taken to review the safety basis of the system as a whole, in order to ensure that not only the operational parameters are improved, but also the current Safety Criteria and integrity of the whole system. It is of great importance to consider a more extensive renewal programme for the installation as a whole in order to have economic and safe reactor operations for another twenty years or more.

Some results of the first years of operation of the brazilian swimming pool reactor have already been described in previous publications<sup>(14,21)</sup>. The following subitens describe some improvements and changes introduced in the renewal programme established for the IEA-R1 research reactor and elaborated after an accumulated operational experience of almost twenty years:

### II.1 - Pool Liner

The reactor swimming pool inner linning, formerly consisting of tile structure, were changed to stainless steel casing due to water infiltration. The tile substitution was done after twenty years of utilization of the IEA-R1 reactor and this fact can be considered as an uncommon case, since in some other similar reactors the substitution had to be done a few years after beginning of operations.

To perform the pool lining work, the IPEN preliminary established a partnership with a brazilian engineering company (PROMON) and afterward they both contacted the foreign nuclear centers that had experience in this field. The elaboration of a basic project was established between PROMON and the Dr. J. D. Randall from Texas A & M University that had already accomplishe similar work at that University; this part of the job was finished in December 1976. After the project detailing the work was initiated on December 1977 and it was concluded after six months. The reactor returned to operation in October 1978 and the results may be considered as highly satisfactory.

## 11.2 - Control Rod Drives

The drive mechanism of the reactor safety rods and the magnet of the neutron absorbing rod coupling were substituted by a new in line drive mechanism type. In the old one the magnet was put it

in a position very near the reactor core and it was immersed into the water, so it presented problems due to radiation damage. An end course hydraulic weakening system was developed by the IPEN personnel for the neutron absorber rod and coupling haste assemblage.

#### II.3 - Pool Surface Activity

A light water moderated pool type reactor is favoured for ease of access to the core are a allowing quick irradiated sample retrieval, and flexibility for locating further irradiation rigs in and around the core area. However, depending on the power level and the operating cycles, there is in these types of reactors the limitation of the radiation levels mainly near the swimming pool surface.

The increasing of the IEA-R1 reactor power level to values higher than 2Mw, is included in the future programme of the IPEN reactor utilization. For this reason, some experiments and calculations<sup>(12)</sup> were performed with the aim to pre ent against radiation levels much higher than the p-esent ones, when the reactor will operate at higher power. It was tried to maintain an artificial layer of hot water over the swimming pool surface, but it was not possible to estabilize this layer, due to the high turbulence of the water returning from the reactor cooling system, even after setting a deflecting plate just above the water returning diffusor. Theoretical calculations indicate that if the level of the surface of water in the swimming pool is raised 30 cm it is possible to operate the IEA-R1 reactor at 5 Mw with radiation levels at the water surface similar to the present ones (12 mR/h at 2Mw)<sup>(12)</sup>. During the pool ling the opportunity was taken to rise in 30 cm the swimming pool water surface.

### II.4 - Pneumatic Tubes

During the pool lining, after the swimming pool was emptied, the original pneumatic sample irradiation system was re-installed. This original system, which has access to the core through the pool bottom, had been eliminated 10 years ago when leakage problems were detected. During this time the system was substituted, with disadvantage, by another one with access through the top og the pool. In the renewal of the old system some improvements were made, such as: change of the terminai positions near the reactor core of the four pneumatic transfer systems, in order to obtain an homogenous flux distribution between the four positions at the relfector of the reactor; substitution of the original pneumatic aluminium tubes by stainless steel tubes; new design of the plate that connects the four systems at the swimming pool bottom, in order to obtain an absolute independence of the pneumatic circuits; renewal of the pneumatic systems drive mechanics with total substitution of wires and components.

# 11.5 - Console

In 1976 a new instrumentation for the control of the reactor was put in service for a continuous mode of operation. The central console for neutron flux control and the radiation monitoring system were purchased from General Atomic and the additional auxiliary system was developed and constructed by the IPEN personnel. The job in the auxiliary system consisted in remaking the drive mechanisms for the hydraulic pumps of the reactor primary cooling system, the alarm and fire systems of the reactor building, and the air circulating system. A moto generator group of no break type was installed to supply the central console and the hydraulic pumps of the reactor primary cooling system. Two moto generator groups of the conventional type with ten seconds of starting delay were installed to supply the additional auxiliary system.

The former instrumentation for the reactor control had to be completely substituted because the reposition components were missing, since most part of the old electronic modules still used electron tubes.

#### II.6 - Cooling System

The dissipation capacity of the reactor cooling circuit was enlarged from 5 Mw to 10 Mw. A fly wheel was added to the hydraulic pump of the primary cooling system. The lack of this wheel had been the cause for operating at 2 Mw instead of 5 Mw during all the past years, because an eventual pump break off would cause a water reflux with the consequent overheating of the reactor core.

A retention tank for the <sup>16</sup>N decay was also intalled in the primary cooling system and now access is allowed to the machinery room in the reactor building basement during the reactor operation.

#### 11.7 - Air Circulating System

An improvement on the ventilation system was performed. Presently the recirculation of three fourths of the building volume air is allowed and one fourth new air is introduced after filtering. An emergency circuit with absolute and active cool filters was added to the exhaustion system. A waiting-room with a logic pneumatic system for the control of the doors opening was built to allow the access to the reactor building interior, since it is kept in depression with relation to the outside.

### II.8 - Emergency Cooling System

The possibility of an accidental swimming pool emptying in the case of a collimator rupture is considered in the Safety Analysis Report<sup>(7)</sup> performed in connection with the reactor modifications. An emergency water system was installed with the aim of a rapid reposition of water and a draining of the leaked water. In case of accident the water supplying is made from a reservoir with 600 m<sup>3</sup> and the water draining from the reactor building is collected in retention tank via the hold up tank.

### 11.9 - Removal of the Inferior Through-port

Two of the several beam ports available for neutron physics experiments, were horizontal through-ports disposed in a vertical plane. During the pool lining the opportunity was taken to remove the inferior through-port, since it did not present suitable safety conditions for reactor operations at higher power levels. This decision was taken due to the following reasons: the through-port was installed only two centimeters above the lower part of the reactor core and so too near the core; it had a large inner volume and two exits to the swimming poc! outside. An accidental rupture of the inferior through-port would leave the reactor core 67% exposed. From performed calculations it was concluded that this alarming situation would be critical for the case of reactor operations at 5 and 10 Mw.

Recently, the demand for higher flux level for neutron physics experiments and radioisotopes production are motivating the upgrading of the reactor power level to 10 Mw. Under the new modifications described, the physical and thermohydraulic calculations indicate that it is possible to obtain an operational power level of up to 12 Mw without sub-cooled boiling effect at the surface of the aluminium cladded fuel plate.

However, due to the impossibility to find high enriched uranium (93%) fuel elements at the international market, the IPEN is at the present moment occupied in ordering a new core load with low enriched uranium (20%). After solving this immediate problem, making an outlook for the future, the purpose of the IPEN is to continue the profitable previous reactor utilization in various fields and also to develop irradiation facilities of the in pile loop type for material testing and fuel element prototype.

Neutron radiography for non destructive testing, neutron activation analysis with ultra rapid sample transfer, delayed neutron counting analysis and sealed source production enlargement will be also carried out.

# III - EXPERIMENTAL FACILITIES

The schematic diagram of the reactor core and the positions of some experimental samples irradiation facilities are shown in. Figure 1.



#### III.1 - Facilities in the Reactor Core

The reactor pool is ten meters deep and was designed to be located inside a building 24 meters high provided with a basement and three floors. The reactor is equipped with a pneumatic irradiation system (rabbit system) which has its blower and associated electromechanical equipment placed in the basement. Four pneumatic transfor systems are installed at different positions on the side of the core near the reflecting zone of the reactor. Two of these rabbit systems 1 and 2 are inside the reactor building: the top of the pool reaches the third floor where the station 1 is located; station 2 is in the ground floor where there is ample space around the reactor with 12 beam hole doors for experiments. The receiving posts of the two other rabbit systems are located in the Radiochemistry laboratories, another building that is attached to the reactor. The four rabbit systems are identical and the rabbits are transferred using vacuum in the dispatching tubes. The rabbits have an external diameter of 30 mm and a height of 75 mm. The time required for the return of the rabbit from the irradiation position to the receiving posts, are 6 sec for station 1 and 2 and 10 sec for stations 3 and 4. All these rabbit systems, were installed for small sample irradiations with time periods smaller than 20 minutes. The systems can work with preset irradiation time from 1 sec to 20 min tile dispatching and returning of the rabbit being made automatically by the use of a special time-switch. The thermal flux at the irradiation position of the rabbits is of the order of  $2-4 \times 10^{11}$  n/cm<sup>2</sup>.s.

The pneumatic rabbits have been mailly used for activation analysis and production of tracers for radiochemistry research. On the other hand, almost all radioisotopes used for industrial and medical applications have been produced from irradiations of materials in different types of sample holders located on the grid plate near the reflecting zone. The irradiation facilities are vertical aluminium, squared cross section tubes, with external dimensions similar to the fuel elements, placed at the reflector in the grid plate. These boxes called "irradiation elements" are being used for long irradiations ( $t \ge 8$  hours) in a rather high neutron flux, of the order of  $10^{1.3}$  n/cm<sup>2</sup> is mainly for sealed sources and radioisotopes production. There are several types of irradiation elements installed in the IEA-R1 research reactor.

EIRA – Water Cooled Irradiation Element consisting of two concentric aluminium tubes. The external guide tube ( $\phi = 1.1/4''$ ) has holes that permit the water passage through the interior. The sliding internal tube ( $\phi = 1''$ ) has 8 receiving positions to lodge standard water proof aluminium cans, called "AOMR capsules" with  $\phi = 3/4''$  and L = 70 mm. Installed in the grid plate is one irradiation facilities called "EIRA", which is used mainly for permanent irradiation of elemental tellurium, sulphur, metallic gold and some other eventual irradiation.

**EIRA** · C – A irradiation facility similar to that described above. The differences are in the external tube which is divided in two parts: the upper part in aluminium ( $\phi \approx 1.1/4''$ ) and the lower part in stainless steel with 80 cm length cased internally with cadmium blades of 0.5 mm thickness. The interior tube has 5 receiving positions for standard AOMR aluminium cans. This unique irradiation element is used for rock irradiations, mainly in a topaze irradiation programme in progress for studies of color change.

EIFS – Dry Irradiation Element. Also with construction similar to those described previously, but in this case with a water proof external guide tube and with the sliding internal tube having still 8 positions for receiving standard aluminium cans. Only two elements of this type are used for eventual irradiations made with special care since the sample cooling in the EIFS is less efficient than in the EIRA facilities.

MUDI – Multidisc Element. Consisting of a rectangular profile of aluminium  $(2^{"} \times 1^{"})$  that has inside a slinding shelf with 26 double positions. In the shelves, it is possible to lodge two magazines having 10 discs of metallic. In or metallic Co in each one. The MUDI is lodged in the interior of a control fuel element fixed in the grid plate, in a similar way as a neutron absorbing rod. There are two MUDI installed near the reactor core, with 52 positions for discs in each one.

EIGRA – Water Cooled Graphite Irradiation Element. A rectangular aluminium profile  $(3'' \times 1 1/4'')$  with a sliding shelf in the interior having 24 receiving positions for AOMR capsules. The EIGRA is fixed

in the grid plate using another rectangular aluminium profile ( $3'' \ge 1.1/2''$ ) which is filled with graphite that acts as reflector optimizing the neutron flux over the samples. Three EIGRA are available to be used mainly for permanent irradiation of elemental tellurium for  $1^{3.1}$  production.

EIFIR – Element for Irradiation of Iridium. Consisting of two concentric tubes of aluminium, the external one being ( $\phi = 1^{"}$ ) used as guide and the internal tube ( $\phi = 7/8^{"}$ ) with 16 shelves where the assembled iridium sources are lodged. These Ir sources are stainless cilindrical capsules ( $\phi = 1/4^{"}$ ) having Ir discs in the interior. There are three EIFIR in the grid plate with 16 positions each one.

EICO - Element for Irradiation of Cobalt Rods. Identical to the EIFIR, but used only for cobalt. Three irradiation facilities are fixed directly in the grid plate using a fixing plug.

GRESIL – A device constructed under an ancient Grenoble (France) - Brazil Cooperation, used for metallic alloys irradiation in studies of order - disorder in crystal lattices with the conjugated action of neutron irradiation and thermal heating. It is fixed directly in the grid plate.

G! – Irradiation Guide. Consisting of a cilindrical aluminium tube ( $\phi \approx 3^{"}$ ) that is fixed at the position 32 in the grid plate where thermal neutrons are prevailing with a flux of about 6 x  $10^{12}$  neutrons/cm<sup>2</sup>.sec. Only one GI is available and it is used for sample irradiations with periods smaller than 8 hours.

Most of the radioisotopes production is concentrated on <sup>131</sup>I, <sup>32</sup>P, <sup>198</sup>Au, <sup>51</sup>Cr, <sup>24</sup>Na, and <sup>82</sup>Br although several other raioisotopes for special purposes have been produced as well. A great part of this production is distributed among several research Institutions or hospitals in Brazil.

Concerning the sealed sources production for industrial application the main interest is in <sup>60</sup>Co and <sup>192</sup> Ir production. In a general way the <sup>60</sup>Co sources have activities in the 100 mCi range and for <sup>192</sup> Ir the preparation of sources with approximately 20 Gi is possible.

For radioisotopes production almost all samples have been irradiated in the different types of sample holders located in the grid plate; pneumatic rabbits have also been used especially for the production of tracers for radiochemistry research which is also subject of this work.

One can estimate the reactor rate of utilization by observing that about ten thousand samples are yearly irradiated by the reactor operation team, in the described irradiation facilities in the reactor core and also in the beam holes that will be described in a next section. On the other hand, any special sample irradiations order is accepted by the staff of the Operation and Maintenance Research Reactor Division of IPEN, provided that the irradiation procedures are under the current safety norms.

## IV - REACTOR UTILIZATION

### IV.1 - Radiochemistry Research

The Radiochemistry Division of IPEN is in charge of the development of experiments in several branches of research in the fields of Radiochemistry and Radiation Chemistry. Its staff is, presently, constituted by 15 researchers being five Doctors, ten Masters in Science and two candidates for a M. Sc. degree. The Division is installed near the IEA-R1 reactor building and utilizes almost exclusively two of the four available pneumatic rabbit stations for sample irradiation. The Division was pioneer concerning the IEA-R1 research reactor utilization because since the beginning of the reactor operations it has been presenting profitable scientific performance as can be seen by ouserving the large number and the quality of the scientific papers prepared during the last twenty years<sup>(3,8,11,17,23)</sup>.

Some current research programmes of the Radiochemistry Division, using the IEA-R1 reactor, are:

IV.1.1 -- Studies and research on chemical reactions and processes using radioactive tracers, especially radioanalytical applications of tetracycline as extracting agent<sup>(17)</sup> for some metals and fission products. Among the experiments the following can be quoted:

a) Separation of <sup>233</sup>Pa from <sup>233</sup>Th for <sup>233</sup>U obtention.

In previous studies results about extraction by a solution of tetracycline in benzyl alcohol of the elements protactinium, thorium and uranium, in the presence of masking agents, were presented. Now, a extraction of uranium using a solution of <sup>233</sup>U tracer was made instead of using a natural uranium solution. Results show that it is possible to perform a radioanalytical separation of <sup>233</sup>U from Thorium and <sup>233</sup>Pa.

- b) Determination of lead, as polutant, using tetracycline as extracting agent and cooalt ethylenediamino tetraacetate as radioactive reagent. The radioreagent method used in this study consists in the lead determination by measuring the radioactivity of the <sup>60</sup>Co displaced by the lead from the Co-EDTA complex. <sup>60</sup>Co is extracted into the organic phase by using a solution of tetracycline in benzyl alcohol and its radioactivity is proportional to the lead concentration in the sample.
- c) Systematic study of the tetracycline behaviour as extracting agent of some fission products. The main interest is in the extraction of elements such as: zirconium, ruthenium, cesium, niobium, strontium, technacium, molybdenium, barium and iodine which are present in a solution obtained from the dissolution of irradiated uranium oxide.

Other studies using radioactive tracers can be cited:

- d) Study of protactinium adsorption in the surface of glass using radioactive tracer. The aim of the study is the knowledge of the adsorption behaviour of some elements of interest in nuclear technology and the attack of glass and plastic surfaces by chemical reagents.
- e) Synergic effect study for the extraction of uranium in the system constituted by tri-n-butyl phosphate (TBP) and di-(2-ethylhexyl) phosphoric acid(HDEHP). The aim is to establish the conditions of the extracting systems that lead to a better uranium extraction by the conjugated use of the TBP and HDEHP complexing agents.
- f) Study of synergic systems as extracting agents in the liquid-liquid extraction and in extraction chromatography, applied to yttrium and lanthanides separation.
- g) Study of the application of isotopic and ion exchange in radiochemical separations. The goal is to establish the best conditions for the separation of interfering elements in uranium and thorium determinations by the <sup>239</sup>U and <sup>233</sup>Th activities, by means of isotopic exchange ssociated to ion-exchange.

IV.1.2 Research and development of radioanalytical method, mainly neutron activation analysis and isotopic dilution. The Radiochemistry Division of IPEN has been offering and developing neutron activation analysis (NAA) for almost twenty years, having a good tradition in this kind of work. Some recent NAA studies include:

a) Trace element composition of coal samples coming from several mines in Brazil. The work consists in the determination of the composition of brazilian coal concerning the concentration of its micro-constituents. From the economic point of view there is interest in the recuperation of some micro-constituents, such as uranium, for instance; from the

environmental point of view it is of interest to examine the concentration of toxical elements that would pass to the atmosphere as a result of the coal burn-up. Analysis are being made in samples coming from seven coal mines located in different regions of the brazilian territory. By irradiating samples with thermal neutrons the following elements were determined: U, Th, Ce, Lu, As, Sc, Rb, Co, Eu, Br, at the ppm range; Fe and K at the per cent range. By means of analysis with epithermal neutrons it was possible to find other elements: Sm, Yb, La, Hf, Ba, Cs, Tb, Ta, Na, Cr, Sb at the ppm range; Fe and Ca at the per cent range. All these results are being collected in order to prepare a report about this subject.

b) Determination of trace elements in sediments of the Santos bay (São Paulo, Brazil) for a study of the silting up of the bay. This work is being performed in cooperation with the Institute of Earth Sciences of the University of São Paulo and it has the purpose to find out the origin of sediments which cause the silting up of the bay and the harbour of Santos. In the 66 samples analyzed by neutron activation analysis(NAA) associated to gamma spectroscopy using Ge-Li detectors, the following elements were found: Sm, La, Br, Th, Cr, Sc, Co, Sb; for all these elements the concentration is in the ppm range. As to aluminium and iron, the concentration was between 1.2 and 9.6 per cent. Zinc was determined after radiochemical separation, and its concentration was in the ppm range. Based on the results obtained for the elements which are present in the collected samples a map is being made of sedimentary facies associated to the distribution curve of the analyzed elements; the resulting figures will be helpful to the determination of the origin and dynamics of the sediments.

Another facility, installed for fission delayed neutron counting, is being used for the determination of uranium concentration and for the  $^{235}$ U/ $^{238}$ U ratio in uranium compounds. More specifically, three works are presently in development:

- c) Determination of the isotopic composition of uranium in rocks of the Brazilian northeastern region. The work consists in examining the isotopic composition in rock samples using NAA with thermal neutrons followed by fission delayed neutron counting or followed by high resolution gamma spectroscopy. The aim is to find possible anomalies in the <sup>235</sup>U content of these rock samples collected in a region of the Brazilian territory which is supposed to have been connected to the Oklo region, in Gabon(West Africa), where a natural fossil nuclear reactor was found in 1972. Signs of burned uranium were searched for in samples from the Estância Area (State of Sergipe) and from the Garanhuns Area (State of Pernambuco). For the location of the geological regions and for the collection of samples, researchers from the Federal University of Pernambuco and also from the Earth Sciences Institute of the University of São Paulo have participated in cooperation with the personnel of the IPEN Radiochemistry Division. An article about this matter<sup>(10)</sup> was presented in the VI<sup>th</sup> Modern Trends in Activation Analysis Conference held in Toronto, Canada, in June 1981.
- d) Determination of the <sup>235</sup>U/<sup>238</sup>U ratio in uranium compounds with different degrees of uranium enrichment. The method of activation analysis followed by high resolution gamma spectroscopy was already applied in the previous work about <sup>235</sup>U content determination in rocks of the northeastern region of Brazil. In the present work the aim is to establish calibration curves using standards and then to apply the method to the analysis of real samples. In order to obtain the calibration curves, standards with several degree of <sup>235</sup>U contents, from 0.518% to 9.993%, were prepared. For enrichments greater than 10% the application of this method is not suitable because the peaks corresponding to the fission pruducts of <sup>235</sup>U are so intense that the peaks of <sup>239</sup>Np, related to the <sup>238</sup>U content, become masked, For isotopic composition determination in

samples with unknown uranium content, it is necessary to plot the 28 curves obtained for each ratio between the <sup>235</sup>U fission products and the <sup>239</sup>Np peaks, in function of the <sup>235</sup>U/<sup>238</sup>U ratios of the standards. The final result is an average of the 28 independent results.

e) Extensions of the method of activation analysis by fission delayed neutron counting. The main interest is in the enlargement of the application field for the facility of fission delayed neutron counting that is permanently installed in the IPEN Radiochemistry Division and then to apply the detection and counting systems for the analysis of uranium and thorium present in several kind of materials, such as: biological materials, nuclear metallic alloys, sediments of rivers and seas, etc. This study aims also to develop <sup>235</sup> U isotopic composition analysis in minerals containing uranium and to perform analysis of transuranium elements, such as <sup>239</sup> Pu and <sup>233</sup> U obtained from <sup>232</sup> Th.

Besides these mentioned direct utilizations of the IEA-R1 research reactor, made by the IPEN Radiochemistry Division, another research branch concerning Radiation Chemistry is also in development. In These kind of experiments not the reactor but a  $^{60}$ Co irradiator is used in order to provide gamma dose (about 2.5 x  $10^5$  rad/h in June of 1981) in samples, with the aim of studying fundamental processes in chemical systems submitted to gamma radiation.

#### IV.2 - Nuclear Physics

The staff of the IPEN Nuclear Physics Division has under its responsibilities the development of experiments and studies in the following main fields of research: Neutron Physics, Nuclear Reactions and Nuclear Metrology.

**IV.2.1** – Studies and research is the Neutron Physics branch. The experiments are made by utilizing almost exclusively the reactor experimental beam ports. The Division has installed several experimental apparatus in front of the beam holes doors at the ground floor of the reactor building.

At the beginning an effort was made to develop a important programme of total cross sections measurements in rare earth elements<sup>(25)</sup>, as well as in hydrogenous liquids<sup>(2,16)</sup>. The total neutron cross sections of praseodymium, ytterbium lutetium, erbium, holmium and thulium have been measured by transmission within the neutron energy grange of 0.001 to 1.0 eV, with special attention given to obtain values at the thermal energy of 0.025 eV. A crystal spectrometer coupled to a mechanical nonochromator and a slow chopper, all of them built at the workshop of IPEN, were used. Powder samples of the oxides were supplied in high in high purity with particular care to eliminate contamination by the rare earths which have high neutron cross sections. Effects of nuclear resonances and of the atomic paramagnetic scattering were considered. In the case of lutetium, the parameters of the first resonance were presented. In the cases of thulium and holmium, experimental values of the paramagnetic scatter ng cross sections were compared with those expected from calculated electronic wave functions.

In the mean time, in a parallel programme three other neutron spectrometers were built at the workshop of IPEN: a) Time-of-flight spectrometer; b) Neutron Diffractometer and c) Triple-Axis neutron spectrometer.

IV.2.1.1 – Measurements with the conventional cold neutron time-of-flight spectrometer. This instrument is installed in a radial beam hole of the reactor. Inside the beam hole a polycrystalline filter cooled with liquid nitrogen transmits a neutron spectrum with a sharp cutoff at 3.95 Å (5.2 meV) and with a mean energy of 3.5 meV and a width of 2 meV; a Pb monocrystal filter is used to reduce the  $\gamma$  ray background. Neutrons Scattered by the sample are pulsed by a curved slit slow neutron chopper and are detected by a bank of <sup>3</sup>He detector after an evacuated flight path of 3.15 m. Scattered neutron

spectra are recorded with a multichannel time-of-flight analyser. Several experiments about proton motions in alcohols by cold neutron scattering were performed<sup>(1,15,24)</sup> by using this spectrometer. Recent activities include studies of molecular dynamics of hydrogenous compounds and of hydrogen motions absorbed in metals and intermetallic compounds.

IV.2.1.2 – Structural studies by neutron diffraction. A double axis-neutron spectrometer with a Pb(220) crystal monochromator with  $2\theta = 38^{\circ}$ , is used to perform the experiments. The diffractometer operation including the data collection is made in an automatic way by using a computer. The instrument has also an automatic goniometer with four axis that permits the study of single crystals samples and polycrystalline samples that present some texture. Presently, one cryostat for liquid helium is being installed in order to study at low temperatures some polycrystalline samples. Some current research programmes are: a) determination of magnetic structures in metallic alloys and compounds, mainly studies of Heusler alloys; b) determination of crystalline textures in materials that have suffered some mechanical process of conformation<sup>(22)</sup>; c) development of the multiple neutron diffraction as analytical technique for the crystalline state.

**IV.2.1.3** – The triple axis neutron spectrometer. The triple axis neutron spectrometer is the most convenient apparatus for experimental determination of dispersion relations for the lattice vibrations with wave vectors along the symmetry directions in a crystal; the spectrometer operation is based on the coherent inelastic scattering of thermal neutrons by a crystalline sample. As part of the neutron inelastic scattering programme of IPEN Nuclear Physics Division, a spectrometer was built. It has a copper crystal monochromator selecting a fixed neutron wavelength of 1.436 Å and as the analyzing crystal it is being used the (002) planes of a pyrolitic graphite crystal with d = 3.354 Å. Details about the project, construction and main characteristics were object of a previous paper. Measurements of the known dispersion relations of copper and studies to obtain) focusing conditions were also performed in order to verify operational conditions and performance of the instrument<sup>(5)</sup>. After these studies the routine procedure for operating the triple-axis spectrometer was established and so the instrument is in conditions to be operated for lattice dynamics studies.

### IV.2.2 – Programme of research in nuclear reactions.

**IV.2.2.1** – Neutron radioactive captule with prompt gamma detection. The method is used for non destructive analysis in geological and biological material and other material that has interest for nuclear technology<sup>(13)</sup>. The analysis is made by counting the prompt gamma resulting from the capture of the thermal neutrons counting from the reactor. An experimental apparatus of internal target geometry type is installed in the 6-inches through-tube which transverses the pool tangentially to the reactor core. The detection system consists of a pair spectrometer constituted by three detectors: one Ge-Li crystal with 42.5 cm<sup>3</sup> of active volume and two Nal(TI) crystals of 5 by 6 inches. With this system a factor of about 6 in the sign to background ratio is obtained. In the region occupied by the target a neutron flux of  $5 \times 10^{11}$  n/cm<sup>2</sup> s is available. Presently, the method is being used to perform analysis of rare earths (Gd, Sm, Dy) in ThO<sub>2</sub> and UO<sub>2</sub> samples; afterwards biological samples will be analysed. Recent activities include the installation of another experimental apparatus of external target geometry type, in a radial beam hole of the reactor. Several tests using filters of single lead crystals and beryllium polycrystalline, were made in order to obtain a thermal neutron beam practically free from fast neutron and gamma background,

IV.2.2.2 – Nuclear spectroscopy studies and electromagnetic hiperfine interactions using the gamma-gamma angular correlation technique. The aim of these fundamental researches made at the Nuclear Physics Division is the study of nuclear structure by means of the experimental determination of nuclear parameters<sup>(19)</sup>, such as: spin and parity of nuclear levels, multiple mixing ratios of the gamma transitions, nuclear momenta, half life of the excited states, etc. In this research branch, studies of electromagnetic hyperfine interactions in metallic alloys and chemical compounds of interest in the field of applied physics, are also being developed<sup>(20)</sup>. Part of the programme consists in the systematic

study of nuclear structure of the nuclei in the mass region between  $70 \le A \le 140$ . The experiments are made from the beta-gramma decay of radioactive sources in general produced by  $(n, \gamma)$  reactions with neutrons of the IEA-R1 reactor or from isotopes obtained by chemical separation of  $^{23.5}$  U fission products. To perform the experiments an automatic spectrometer for the gamma-gamma angular correlation measurement is used with two or three detectors and also with another angular correlation spectrometer where an external magnetic field of 26 k Gauss can be applied to the radioactive sample. The electronic system adopted is the conventionally used in nuclear spectroscopy. Recent studies include the  $^{7.5}$  Se,  $^{12.7}$  Te and  $^{12.9}$  Te nuclei. In the field of hiperfine interactions the hyperfine magnetic fields are being studied in Heusler alloys of the type: Co<sub>2</sub> TiA1 (Ga, Sb, Sn).

**IV.2.2.3** – Photonuclear reactions and development and application of the fission track registration technique<sup>(6)</sup>. The photonuclear reactions near the threshold are studied by using photons from the thermal neutron capture. The experiments are directed towards the determination of cross sections for  $(\gamma, f)$  and $(\gamma, n)$  reactions, angular distribution of fission fragments and average number of neutrons  $\overline{(v)}$  emitted in the photofission. The experimental apparatus is installed in front of a radial beam hole door at the ground floor of the reactor building. Each gamma beam constituted by practically one unique gamma energy is obtained by putting one selected target near the reactor core and so for each target a discret gamma line is obtained covering the interval between 3.5 to 11 MeV. The  $(\gamma, n)$  reaction cross section and the  $\overline{v}$  value are measured by using a system for neutron detection constituted by a long counter with 60<sup>-3</sup> He detectors in a geometry of four concentric rings. A fission track detector of the type Makrofol KG is used for the  $(\gamma, f)$  reaction cross section measurements. The angular distribution measurements are made by using a set of three coupled chambers with a system that permits the sample rotation during the measurements in order to have an isotopic geometry. Two works are presently in development: a)  $(\gamma, f)$  and  $(\gamma, n)$  cross section measurements of  $2^{3.7}$ Np near the threshold; b) determination of the average number of neutrons emitted in the  $2^{3.8}$ U and  $2^{3.2}$ Th photofission.

**IV.2.3** – Activities of the Nuclear Metrology Laboratory. This laboratory is a branch of the Nuclear Physics Division and it has under its responsability all the determinations with high precision of radioactive sources produced at IPEN or other institutions in the country<sup>(4)</sup>. It is equipped with five counting systems for activity measurements and routine calibrations: 1)  $4\Pi \beta \gamma$  coincidence system; 2) ionization chamber open to ambient air; 3) a pressurized well-type ionization chamber; 4) a system for relative measurements by using a Ge-Li detector and 5) a system for relative measurements that utilize a Si-Li detector. With these mentioned calibration systems the laboratory has performed in the last year, 150 standardizations with accuracy better than 0.1%.

#### ACKNOWLEDGMENTS

The authors wich to thank the researches of the three Division (Nuclear Physicis, Radiochemistry and Reactor Operation and Maintenance) of the Center or Operation and Utilization of the IEA-R1 Research Reactor (COURP), as well as the technicians of the IEA-R1 operation team.

#### RESUMO

O reator IEA-R1 do Instituto de Pesquisas Energéticas e Nucleares, IPEN, de São Paulo, Brasil, um reator de pesquisa de piscina moderado a água leve do tipo MTR, ficou crítico pela primeira vez em 16 de setembro de 1957. De maneira geral, nestes vinte e quatro anos, o reator foi utilizado sem interrupção por usuários di IPEN e de outras instituições, para a execução de trabalhos nos campos da pesquisa básica e aplicada, para dissertações de mestrado e teses de doutoramento e para desenvolvimentos técnicos. O presente trabalho descrever em uma vista geral algumas pesquisas desenvolvidas e o programa de reforma do reator de pesquisa IEA-R1, no qual foram feitas diversas alterações e aprimoramentos. Estão descritas as atividades atuais relacionadas com produção de radioisótopos e alguns programas de pesquisas em andamento no campo da Radioquímica, especialmente estudos e pesquisa da frea de Efsica Nuclear do IPEN que incluem: estudos de espectroscopia nuclear e interações hiperfinas electromagnéticas; difração de neutrons; estudos de espalhamento inelástico de nêutrons na matéria condensada; desenvolvimento e aplicação da técnica de registro de traços de fissão em detectores de estado sólido; captura radioativa e néutrons com detecção de gamas prontos e, ainda, pesquisa no campo da metrologia nuclear. (  $\phi = \sqrt{t_{cont}}$ )

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