ENHANCEMENT IN THE UTILIZATION OF IEA-R1 BRAZILIAN RESEARCH REACTOR

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1. INTRODUCTION

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 science and engineering. The extent of the utilization of a research reactor is basically determined by it's power level, which in turn determines roughly the neutron flux available in the reactor, as well as on it's operational schedule . Small power level and relatively short operational cycle (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 level in conjunction with prolonged operational cycle (several weeks continuous operation) on the other hand result in high fuel consumption and require expensive maintenance program. Since small research reactors are generally operated and maintained through government funding, which are usually scarce, it is obvious that efforts must be made to optimize the utilization of such reactors and to determine the power level and operational regime on the basis of cost and benefit. Social benefits derived from the reactor utilization should usually be given priority however the cost of such benefits must be determined realistically.

Brazil has four operational research reactors and the Comissão National de Energia Nuclear (CNEN) is responsible for their operation and maintenance. These reactors fall into the following categories: two for research, one for training and one critical assembly. Some details about the characteristics of each reactor are presented in table 1.

Since the IEA-R1 is the only Brazilian research reactor operating at substantial power level (2MW) suitable for utilization in wide areas of research such as physics, chemistry , biology and engineering as well as in the production of radioisotopes for medical and other applications, we wish to review briefly general description of this reactor, it's past operational experience and current utilization. We shall also describe briefly about the future plans to optimize it's performance in the areas of research and development but especially it's use as a radioisotope producer.

Table I: BRAZILIAN RESEARCH REACTORS

	IEA-R1	IPR-R1	Argonauta	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 MW	100 kW	0.2 kW	
Enrichment	20-93%	20%	19.01-19.91%	4.3%
Supplier	Babcox & Wilcox	General Atomics	USDOE	Brazil

2. IEA-R1 RESEARCH REACTOR

The IEA-R1 research reactor is a swimming pool type reactor, using light water as moderator and coolant and graphite as reflector. The reactor was constructed by Babcok & Wilcox and achieved it's first criticality on September 16, 1957. Although designed to operate at 5 MW the reactor really never operated at this power level. It has been operating at 2 MW since early sixties on an operational cycle of 8h a day 5 days per week. The reactor core is assembled in a 8x10 grid plate with fuel elements reflectors and irradiation elements arranged as Shown in figure 1.

The original fuel load consisted of plane plate (18 plates per element with size 62cm x 7cm x 0.15cm) 93% enriched U-Al standard fuel elements. However following the international efforts, for core conversion to low enriched uranium (LEU) fuel, several elements have been replaced with 20% enriched fuel elements. Five such elements were initially purchased from NUKEN, Germany . IPEN at present produces it's own LEU plate type elements (U_3O_8 - Al, 20% enriched) and in about a year's time it is expected to have a complete LEU core using Brazilian fabricated fuel elements. The control elements are fork type (Ag-In-Cd) fabricated by CERCA, France. The maximum thermal neutron flux is of the order of 2×10^{13} n/cm^2.s. In figure 2 a horizontal cut of the present core configuration is shown.

The reactor building is located in the premises of the Instituto de Pesquisas Energéticas e Nucleares (IPEN) which is situated in side the campus of the university of São Paulo. IPEN is administratively subordinated to the Comissão Nacional de Energia Nuclear (CNEN) and is the most important center for nuclear research in the country. Presently it has a permanent staff of about 1.500 people including the scientists, engineers, technicians and administrative personnel. The main areas of research and development (R&D) which are under the responsibility of institute are: Nuclear fuel cycle & metallurgy, Nuclear reactors, Applications of nuclear techniques, and Health physics

The IEA-R1 research reactor can be considered as a multipurpose facility and has been used in all these years intensively for basic and applied research in nuclear, neutron and solid state physics, radiochemistry, radiobiology and material science. The reactor has also been used for the training purpose, for the production of some useful radioisotopes for applications in the industry and nuclear medicine, and also for some irradiation services. The reactor is used routinely by all the departments of

IPEN. It is also used occasionally by the faculty members and the students of the universities and other research institutions of the country for academic and applied research. By far the largest user of the reactor is however the Department of the Applications of Nuclear Techniques (ANT) of IPEN which is responsible for the R&D activity in the areas of nuclear physics and radiochemistry, radiobiology, industrial applications of radioisotopes and radiations and for the production and distribution of radioisotopes and radiofarmacuticals for medical applications.

The Department of the Applications of Nuclear Techniques has a permanent staff of 163 people including Physicists, Chemists, biologists, engineers, and technicians. In addition there are about 50-60 students attached to the department carrying out their graduate work towards M.Sc. or Ph.D. degree programs in nuclear sciences.

It may be worth while to mention here that in addition to the IEA-R1 reactor, which has been the major facility for the research and development work carried out by the staff members and students as well as for the production of radioisotopes, this department has other equipment and laboratories which are routinely used for the R&D work and services, important among these facilities are: A Linear electron accelerator with 1.5 MeV electron energy used for irradiating electrical cables for industries, a variable energy Cyclotron with 24 MeV protons and 28 Mev alpha particles used for producing some medical radioisotopes like ^{67}Ga, ^{123}I and ^{111}In, several high activity cobalt sources (5000-10000 C_i) for gamma irradiation and a number of radiochemical laboratories and hot cells for the processing of radioisotopes.

3. BASIC AND APPLIED RESEARCH

The IEA-R1 reactor is used as an intense neutron source where the neutron beams can be obtained through a number of beam holes provided and used in a variety of nuclear and neutron physics experiments. The reactor is also used for the in core irradiations in different applications for example irradiation of silicon for NTD, irradiation of plastic films for making the nucleopore filters and irradiation of samples for neutron activation analysis. The operational experience and the utilization of the reactor in basic and applied research has been quite intense in these almost four decades of its operation and has been described in some of the earlier publications [1-3]. Here we wish to mention only briefly some of the experimental facilities attached to the reactor which have been crucial to the research and development program of the department in all these years. The main characteristics of these instruments have been described in detail elsewhere [3].

The Beryllium filter - time of flight spectrometer has a liquid nitrogen cooled Be filter which provides the cold neutron spectrum with mean energy of 3.5 meV and FWHM 2.0 meV. The spectrometer is used for studying hydrogen vibrations in metal hydrides and other hydrogenated solids and liquids, atomic and molecular diffusion and electronic excitations in molecules.

The Triple axis neutron spectrometer uses a Cu111 monochromator diffracting at 1.4392 Angström, pyrolitic graphite as analyzer and ^{3}He detectors. Main areas of research interest include dispersion relation in monocrystalline solids, powder diffractometry and lattice dynamics in hydrogenated compounds.

The Neutron Diffratometer uses a copper single crystal oriented for reflection 200 as monochromator, has a beam cross section of 25 cm^2 and neutron flux at the sample of the order of 10^5 n/cm^2. s. The neutron detectors are ceramic window type BF_3 detectors. The diffratometer is totally automated with an on line micro computer. Current research areas of interest include texture analysis, order disorder studies in metallic alloys, study of the crystalline quality of single crystals, crystal and magnetic structure determination, neutron multiple diffraction as a method for analysis of crystal and magnetic structure.

Experimental arrangement for neutron radiography is used for material inspection such as reactor fuel elements, turbine blades, ceramics, corrosion in air craft structure. Application of real time neutrongraphy, and application of solid-state nuclear track detectors in neutrongraphy are under development.

The Experimental arrangement for the study of photonuclear reactions with thermal neutron capture gamma rays uses the tangential beam port where the target elements are placed near the reactor core and irradiated with thermal neutrons to produce monoenergetic gamma rays through (n,γ) reaction. These gamma rays are then collimated and extracted to induce photonuclear reaction on sample targets placed out side the beam hole. The main research interests are: Photofission cross section for actinides near threshold (5-11 MeV), Photoneutron cross section for light and heavy elements near threshold (5-11 MeV), Fission fragment angular distribution, Gamma ray scattering cross sections.

Neutron activation analysis is used as an analytical technique for studying the samples of interest in the areas of biology, geology, environmental science, food and agronomy. Some of the current research which is being carried out in this area includes determination of inorganic elements in Brazilian medicinal plants, dentine abrasion and composition of dental pastes ,trace element determination in geological samples, in rain water, and soil, heavy element analysis in river sediments, determination of mercury and methyl mercury in human hair. Besides these research areas service is offered for the routine analysis of a variety of samples to the universities, research institutions and also to private industries using NAA technique. Approximately 500 samples are analyzed each year.

4. SILICON IRRADIATION

Through a technical cooperation project sponsored by the International Atomic Energy Agency during 1989-91 IPEN developed the technique of silicon doping with phosphorus by neutron transmutation. A simple device for irradiating single crystals of silicon with up to 4" diameter and 40 cm length was installed in the reactor IEA-R1 at the end of 1991. The silicon irradiation rig shown in figure 3 is located in the graphite reflector (see fig.2) Details about the design of the irradiating device and it's performance can be found else where [4,5]. The important feature of the irradiation procedure is, that the excellent axial homogeneity in the distribution of neutron dose received by the crystal is achieved by irradiating two silicon crystals 20 cm long each simultaneously one on top of the other. After exactly 50% of the required dose to attain the final target resistivity the crystals are pulled out of the reactor and their positions are interchanged. The irradiation then continues till the remaining dose is complete. Precise detection of the 50% fluence is the key to the success of this method and is accomplished with the help of self powered neutron detectors placed close to the irradiation tube as well as the cobalt monitors which are irradiated with each silicon ingot. The radial homogeneity in the neutron dose received by the crystals is achieved by the usual procedure of rotating the crystals during the irradiation. Excellent doping uniformities and doping precision have been obtained by using this method. Considering the irradiation of just 4" diameter crystals and the target resistivity of approximately 50-60 Ωcm the irradiation capacity of the reactor at present is approximately 600 kg per year.

5. RADIOISOTOPES PRODUCTION AT IPEN

Beginning of the production of radioisotopes for medical use in IPEN dates back to late fifties when a small experimental production of ^{131}I was started. In 1961 with the IEA-R1 reactor operating at 2MW, 8h a day 5 days a week the production of ^{131}I was sufficient to meet the demand of existing hospitals and clinics practicing the nuclear medicine in the country. In the subsequent years the institute began the production of several other radioisotopes for use in nuclear medicine such as ^{32}P, ^{198}Au, ^{24}Na, ^{42}K, ^{35}S, ^{82}Br and ^{51}Cr as well as the preparation of labeled compounds. National demand for these isotopes increased rapidly during seventies and it was no longer possible to meet the requirement of hospitals and clinics with the existing production. IPEN therefore began the importation of ^{131}I, ^{32}P and ^{51}Cr which could not be produced in sufficient quantities and high enough specific activities with the domestic reactor. These isotopes were processed and distributed to the medical centers.

Until 1980 all the ^{99m}Tc generators used in the country were imported. Due to rapidly increasing demand of this important radioisotope and high cost of importation IPEN started making its own ^{99m}Tc generator kits, with automatic elution system, from the fission ^{99}Mo purchased from Canada. Today ^{99}Mo - ^{99m}Tc generators represent more than 90% of all the radioisotopes distributed to the hospitals and clinics in the country. The present national demand for ^{99}Mo is of the order of 4.4 TBq (~120 Ci) per week and is increasing at the rate of about 15% per year during the last few years. The present domestic requirement for other radioisotopes is: ^{131}I (6 $\mathrm{C_i}$ /week), ^{32}P (50 $\mathrm{mC_i}$ /week), ^{35}S (20 $\mathrm{mC_i}$ /week) and ^{51}Cr (70 $\mathrm{mC_i}$ /week). The demand for these radioisotopes is also increasing rapidly. The demand for ^{24}Na, ^{45}Ca is small and sporadic. The present annual cost of the importation of these medical radioisotopes is approximately US$1.8 million. It is estimated that at present the distribution of medical radioisotopes by IPEN provide benefits to approximately 600.000 patients per year through 165 hospitals and clinics all over the country.

A recent survey has shown that the projected demand for the medical radioisotopes in the country is likely to increase by a factor of two to three in the next 5-10 years due to rapidly increasing interest, in nuclear medicine, of many private hospitals and clinics in the country. In order to meet this demand the institute may have to further increase the level of importation of these radioisotopes with costs reaching to about 4-5 million dollars a year. This is some what cumbersome for institutions like ours whose funding comes exclusively from the federally approved budget. Further more, increasing reliance on practically a single supplier of this vital radionuclide will certainty be an strategic disadvantage.

Considering these factors, as well as the necessity to produce several other radioisotopes, for therapeutic applications for example ^{153}Sm, ^{165}Dy, ^{166}Ho, ^{169}Er and ^{186}Re, IPEN took an important decision in the beginning of 1995 to up-grade the reactor power from 2 to 5 MW and also to increase its operational cycle from 8h a day, 5 days a week to 120h continuous per week.

Initial plans to produce ^{99}Mo from fission had to be given up due to shortage of necessary financial resources as well as due to the technical problems associated with complex radiochemical processing and highly radioactive waste management. In view of the recent success demonstrated by China and Vietnam in producing the ^{99}Mo - ^{99m}Tc generators by (n,γ) reaction and gel technique it was decided to adopt and develop this method which is less expansive and does not produce highly radioactive waste. Under the new operational schedule of our reactor, that is, a continuous 120 hours operation per week, and thermal neutron flux of the order of $5x10^{13}$ n/cm^2.s expected at 5MW it should be possible to meet to a large extent the present domestic demand for this radionuclide with very little need for importation. In addition, the reactor will provide the possibility to produce radioisotopes like ^{131}I, ^{32}P,and ^{35}S in sufficient quantities and higher specific activities to meet the local demand. As a consequence of the new operational condition of the reactor, the radionuclides such as ^{153}Sm, ^{165}Dy, ^{168}Ho, and ^{169}Er, can also be produced in sufficient quantities to meet the present requirements.

The proposed up grading of the power level to 5MW as well as the continuos 120h per week operational cycle of the reactor was enthusiastically welcome by most of other users of the reactor who will be able to increase the productivity of their services or products. For example the presently estimated capacity for the production of NTD silicon which is about 600 kg per year could be increased to about 2500 kg per year based on just 4" diameter crystals and more if it is decided to install a new rig to irradiate 5" diameter silicon crystals. Installation of a 5" rig is presentlty under consideration. A production capacity of NTD silicon of this order of magnitude will certainly be more attractive to the international silicon producers. Similarly the production of ^{192}Ir, in the form of wire or discs, and ^{60}Co as discs, used as sealed sources in brachytherapy, ocular cancer and in industrial gamma radiography respectively will benefit either in terms of larger number of sources produced or sources produced with larger specific activity. The experimental facilities installed in the beam holes of the reactor for academic and applied research in nuclear and neutron physics will benefit largely due to increase in the, intensity of the neutron beams but also due to increased data acquisition time available per week. The quality of data in most of these experiments is mainly determined by the counting statistics. A good quality data could be obtained in shorter duration experiments thus reducing considerably the systematic errors.

6. UP-GRADING THE REACTOR POWER TO 5MW

To achieve the goal of up-grade of the reactor power to 5MW, and implement the 120h per week operational cycle, the project has been divided in three major areas, production of fuel elements, optimization of systems, structures and components of the reactor and optimization of the radioisotopes production.

Due to necessary time required to implement all the IEA-R1 reactor modifications, and licensing from the regulatory body, it was decided as a first step to change the operating cycle of the reactor to 48 hour a week while maintaining the power level to 2MW. This was done in the beginning of November 1995. The operational cycle has since been further increased and the reactor is presently operating 64 hours a week. This has already permitted to increase the production of radioisotopes in the country including a modest commercial production of ^{153}Sm used in the metastatic bone cancer therapy.

The required modifications in the reactor will increase the fuel element consumption to more than 10 elements per year instead of the usual consumption of 2 fuel elements per year. Just the 64 hours continuous weekly operation will increase the fuel consumption to about 4 elements a year. Efforts are being made to step up the production of fuel elements made at IPEN. It is also planned to increase the density of fuel elements (U_3O_8 - Al) from the present 1.8 g U/cm^3. In addition the institute will soon start to produce U_3Si_2 - Al fuel elements with a density of 3.1 g U/cm^3 , initially at experimental level to qualify the fuel element production but, with a goal to eventually replace the present U_3O_8 - Al fuel elements with indigenous U_3Si_2 - Al elements.

The operation of reactor at 5MW level will require changes in some of the reactor systems, replacement of some structures and introduction of some new systems, mainly those related with safety and design problems for example: reactor cooling system, ventilation system, electrical system, instrumentation and control, radiation monitoring system etc. A new emergency cooling system consisting of passive spray coolers and a hot layer to reduce the circulation of radioactive ions on the surface of reactor pool will also be installed. Optimization of all these systems is needed mainly to comply with the requirements of the licensing by CNEN. It is expected that the entire project will require a tittle more than one year and cost around US$ 1 million . Thus it is expected that the IEA-R1 reactor should be operating at 5MW and 120h continuous per week in the first quarter of l997.

The production of radioisotopes with the new operational conditions of the reactor will also require modifications and improvements in the processing laboratories. Some new hot cells will have to be constructed as well as the schielding of others may have to be optimised to handle larger activities. New equipment for producing ^{99m}Tc using gel method will be purchased.

7. CONCLUSIONS

To summarize, the up-grade of the power to 5MW and continuos operation of the IEA-R1 reactor will allow that it becomes actually a radioisotope producer and help meet to a large extent the demand for medical isotopes in the country. With reactor operating at 5MW and 120 hours per week and with the thermal neutron flux of the order of $5x10^{13}$ n/cm^2.s. it is estimated that the local production of ^{99}Mo with (n,γ) reaction will result in ^{99m}Tc generators with activities between 250 mC$_i$ and 1000 mC$_i$ and save the country about US$ 800,000 per year. An additional saving of approximately US$ 280,000 is expected from the local production of other radioisotopes like ^{131}I, ^{32}P,and ^{35}S.

As a consequece of the increase in the level of neutron flux other applications such as NTD silicon production, neutron radiography, and neutron activation analysis will have better performance. Basic and applied research will certainly benefit from this new operational condition of the reactor due to availability of more intense neuron beams. This will also stimulate renewed interest in other applications which are at present at an experimental stage for example boron neutron capture therapy (BNCT) and coloration of topaz Since Brazil is one of the most important world producer of topaz, special devices are being installed in the reactor to initiate the commercial irradiation of this semi precious gemstone to induce blue coloration.

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Figure Captions

Figure 1. General view of the reactor grid plate with fuel, control and reflector element

Figure 2. A cross-sectional view of the IEA-R1 reactor core. Si is the silicon irradiation position

Figure 3. Side view of the silicon irradiation rig located in the graphit reflector

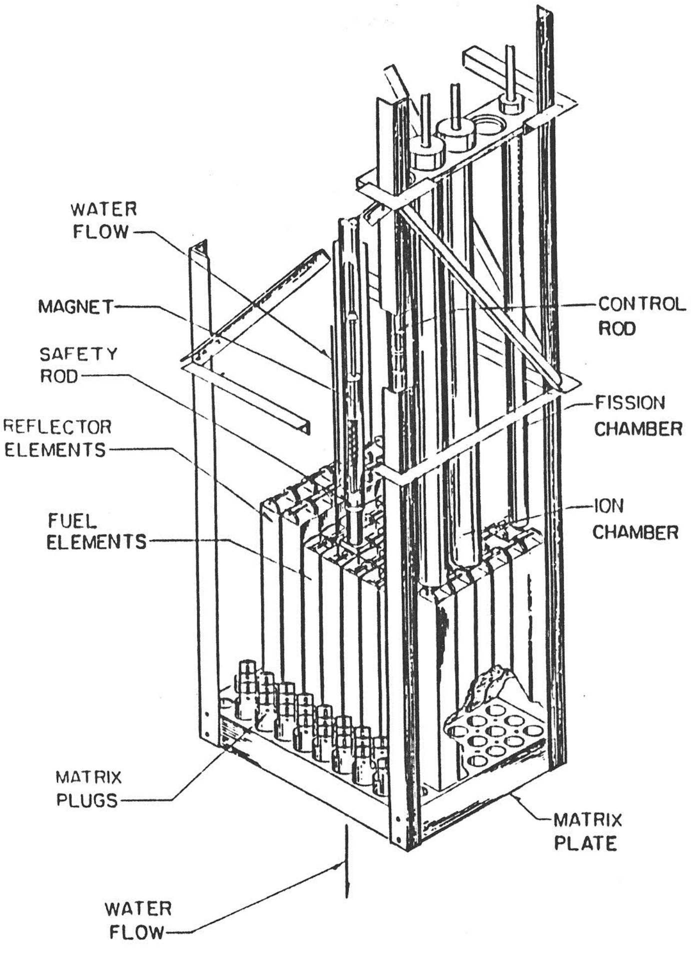

WATER FLOW
MAGNET
SAFETY ROD
REFLECTOR ELEMENTS
FUEL ELEMENTS
MATRIX PLUGS
WATER FLOW
CONTROL ROD
FISSION CHAMBER
ION CHAMBER
MATRIX PLATE

Figure 1

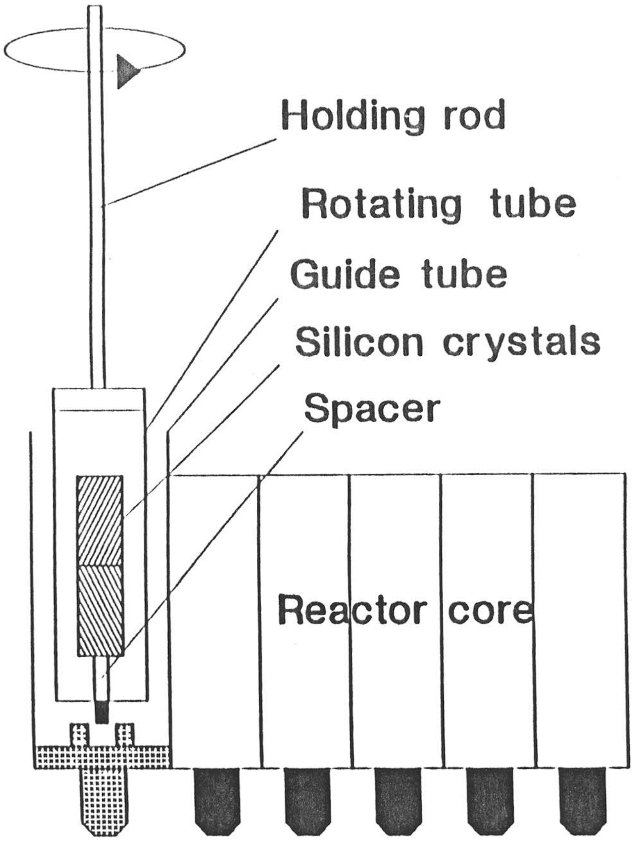
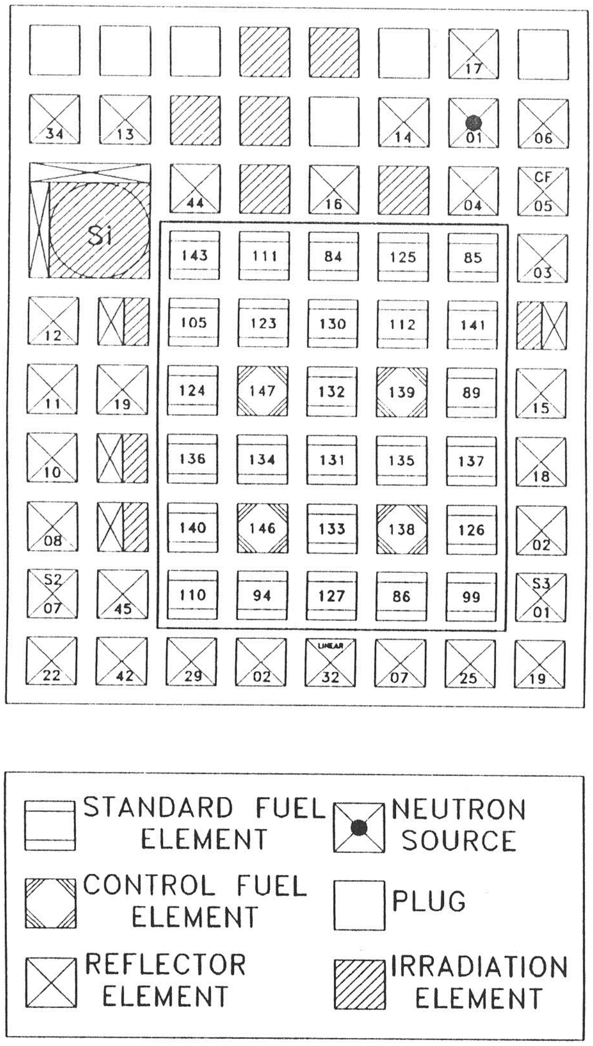

Holding rod
Rotating tube
Guide tube
Silicon crystals
Spacer
Reactor core

Figure 3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146
133
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02
S2
07
45
110
94
127
86
99
S3
01
22
42
29
02
LINEAR
32
07
25
19
STANDARD FUEL ELEMENT
NEUTRON SOURCE
CONTROL FUEL ELEMENT
PLUG
REFLECTOR ELEMENT
IRRADIATION ELEMENT

Figure 2