

Optimization on the core of IEA-R1 research reactor for enhance the radioisotopes production

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ABSTRACT

In this work a parametric study was carried out to increase the production of radioisotopes in the IEA-R1 reactor. One of the variables directly proportional to isotope production is the magnitude of the neutron flux in which some material is exposed, so the main objective of this work was to increase neutron flux, especially in the center of the reactor in the beryllium element irradiator (EIBe), within the operational and safety limits of the reactor. The study is initiated by defining a default configuration, in which core of the IEA-R1 reactor is modeled with all fresh fuel assemblies to ensure the reduction of variables that affect the data analysis, then para metric studies were performed evaluating, by comparative analysis of the behavior of the relation of neutron flux versus the fuel for the standard configuration. Therefore, another configuration was tested: the changes in the core of graphite reflecting elements for beryllium, as well as, the result due to reactor core compaction. Parameters such as the fraction of delayed neutrons (B_{eff}) and temperature reactivity coefficient are analyzed to ensure that the configuration has the minimum safety requirements for the reactor safe operation. The results of the study demonstrate a large increase in neutron flux magnitude and in particular in the center of the nucleus in the beryllium irradiating element.

1. INTRODUCTION

The radiopharmaceuticals produced at IPEN are mostly made from material irradiations carried out in the IEA-R1 reactor, an open-pool research nuclear reactor. The reactor building is located on the premises of the Institute of Nuclear and Energy Research (IPEN / CNEN-SP), in the USP, in São Paulo [2]. Although its core was designed for 5 MW, initially there was no defined operating regime for the IEA-R1 reactor, and the thermal power ranged from 200 kW to 2 MW. From 1961, the reactor started to operate at a defined power of 2 MW. Between 1971 and 1991, a number of modifications were introduced to the reactor to adapt its facilities to the latest safety standards. In 1995, IPEN / CNEN-SP optimize the IEA-R1 reactor to operate at 5 MW, its design thermal power. The reactor then underwent several renovations and modernizations which, completed in September 1997, increased the maximum operating thermal power to 5 MW [3] [4] [5].

Even though CNEN's production of radiopharmaceuticals, Brazil is not self-sufficient in its production, depending on other countries such as Canada to import the Tc-99m used, especially in areas such as cardiology and oncology. This radiopharmaceutical is used in more than 80% of nuclear medicine procedures worldwide, mainly for scintigraphy examinations [6]. In 2009, with the shutdown of the Canadian NRU reactor, there was a shortage of this radiopharmaceutical resulting in the shutdown of several medical procedures. This event was an indication that the expansion of radiopharmaceutical production is more than an economic issue, but also a public health issue [6].

These questions were some of the main justifications behind the Brazilian Multipurpose Reactor (RMB) project. RMB is a radioisotope nuclear research and production reactor capable of making the Brazilian market independent of the importation of radiopharmaceuticals. This reactor will also enable studies for various purposes such as nuclear fuel irradiation tests, structural materials used in reactors, conducting scientific research with neutron beams and a range of other applications [7]. However, the RMB is not close to having its construction completed and operation started, making the issue of meeting national demand for radioisotopes a current problem and of importance to be minimized until the RMB becomes operational.

Thus, IEA-R1 becomes a key player in the production of radiopharmaceuticals in Brazil. However, there is a need for an adaptation of the nucleus of this reactor to increase the neutron flux, which is essential for the generation of radiopharmaceuticals. This research project aims to improve the irradiation conditions in the IEA-R1 reactor and thus optimizing and contributing to the production of radiopharmaceuticals in the country.

2. METHODS AND DATA

2.1. IEA-R1 reactor

Actually the IEA-R1 reactor operates with a thermal power of 4.5 MW and the arrangement of its core can be seen in figure 1 below.

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| Tills and the second | B tHA A 03 | PILIS 04 | Alla OS | 5 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 | PIIIS 07 | Pillip 08 |
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| Grafite 21 Grafite 2 | 2 Grafite 23 | P A 24 | Grafite 25 | Grafite 26 MINI PLACAS | Grafite 27 | Grafite 28 |
| 31 U/A Grafite 31 Grafite 3 | 33 Be 33 | EIRA A | 34 Be 35 | 31/05/11 36 | 35 Be 37 | 46 s CIC Grafite 38 |
| 13A 42 | PADRÃO U3Si2-Al 3pt/or ² ARAMAR 03/06/13 | PADRÃO U3Si2-AI 350/ce ³ ARAMAR 03/06/13 | PADRÃO U3Si2-AI 198/08 ² ARAMAR 28/04/12 | PADRÃO U3Si2-AI 3,6/cs ¹ ARAMAR 03/10/16 | PADRÃO U3Si2-AI 1q1/or ⁸ ARAMAR 30/12/15 | 36 |
| Grafite 41 Grafite 4 | 218 | 224 14.56% 44 | 216 32,84% 45 | 234 8.46% 46 | 231 10.55% 47 | Be 48 |
| 47 Grafite 51 Be 51 | PADRÃO U3Si2-Al 3gl/cs ³ ARAMAR 12/12/18 239 0.00% 53 | BC U3Si2-AI 34/04/13 ARAMAR 14/01/13 229 31.03% 54 | PADRÃO U3Si2-AI 368/08 ARAMAR 03/10/11 215 36,25% 55 | BS#3 U3Si2-AI 3#/m ³ ARAMAR 24/04/17 242 8.45% 56 | PADRÃO U3Si2-AI 3pl/cm ³ ARAMAR 02/07/12 217 29.16% 57 | He 58 |
| 51 Câmara 35 | PADRÃO U3Si2-Al 3gl/cm ³ ARAMAR 20/09/17 | PADRÃO U3Si2-AI 3/10/11 | в | PADRÃO U3Si2-AI 30/00 ³ ARAMAR 06/06/11 | PADRÃO U3Si2-AI 341/m ³ ARAMAR 30/03/15 | 48 |
| Grafite 61 Grafite 6 | 2 236 3.61% B3 | 214 31.20% 64 | EIBe | 213 33.86% 66 | 226 12.841% 67 | Grafite 68 |
| 04A 43 | PADRÃO U3Si2-AI 398/cm ³ ARAMAR 27/10/14 | BS # 1 U3Si2-AI 3#/rm ³ ARAMAR 20/09/17 | PADRÃO U3Si2-AI 3;8/cm ³ ARAMAR 05/06/11 | BS#2 U3Si2-AI 3#/00 ³ ARAMAR 14/01/13 | PADRÃO U3SI2-AI 3gl/cm ³ ARAMAR 03/06/13 | RA 2 |
| Grafite 71 Grafite 7 | 2 223 16,46% 73 | 243 5.90% 74 | 210 38.07% 75 | 230 30.37% 76 | 225 21,20% 77 | Be 78 |
| Safety 2 52 | PADRÃO U3Si2-Al 3gl/cm ³ ARAMAR 30/12/15 | PADRAO PLR U3Si2-AI 34/cm ³ ARAMAR 22/12/08 | PADRÃO U3Siz-Al 198/08 ³ ARAMAR 18/12/15 | PADRÃO U3Si2-Al 3,8/m² ARAMAR 03/10/16 | PADRÃO U3Si2-Al 361/01 ⁸ ARAMAR 12/12/18 | Safety 3 47 s |
| Grafite 81 Grafite 8 | 2 227 9.65% 83 | 238 2.03% B4 | 232 13,05% 85 | 233 8.94% 86 | 240 0.00% 87 | Grafite 88 |
| 49 20A | . 30 | 29 | 27 | 31 | 32 | 28 |
| Grafite 91 Grafite 9 | 2 Be 93 | Be 94 | Be 95 | Be 96 | Be 97 | Be 98 |
| GANHO DE REATIVIDADE CALCULADA = 11 | IS.00 PCM GANHO | I REATIVIDADE MEDIDO | = 1258,66 PCM | ENTRADA DOS ECP | - 239 e 240 | DGS 2018 |
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Figure 1: IEA-R1 reactor configuration – 263 [8].

The IEA-R1 reactor core consists of 30 MTR type fuel assemblies (26 standard and 4 control) with graphite reflector. All radioisotope irradiation elements and material tests are located on the reflector.

Standard fuel assemblies consist of 18 fuel plates. Figure 2 shows the fuel assemblies manufactured by IPEN. The external dimensions of the two assemblies are the same. The dotted lines surrounding the elements in the figures represent the dimensions between centers of two fuel assemblies. The control assemblies, manufactured by IPEN, has the same dimensions as the standard fuel assembly, but with 12 fuel plates (figure 3). The second and second to last plates do not exist, giving way to the control rods. The first, third, third to last, and last plates are made of aluminum, leading from guides to the control bars. The control bars, consisting of an Ag-In-Cd alloy (80-15-5 wt%) are of the fork type with a thickness of 0.31 cm, width of 66 cm and active length of 65.1 cm. Figure 4 shows an axial view of the fuel element. The average length (or height) of the fuel plate is 62.5 cm, with 60 cm active length and 1.25 cm aluminum at the top and bottom of the plate.



Figura 4: Figure 4: Axial View of the Fuel assembly [5].

The control plate alloy composition by weight is 80% Ag, 15% In and 5% Cd. Silver is composed of two isotopes, Ag-107 and Ag-109 with abundances of 51.83% and 48.17% atoms respectively. Indium is also composed of two isotopes, In-113 and In-115 with abundances of 4.28% and 95.72% respectively.

The fuel plate has a density of 3.0 gU / cm³ consisting of an alloy of U₃Si₂-Al, with 19.9% enrichment of U-235, with the mass of each plate being equal to 15.58 g of U- 235, aluminum mass of each plate equal to 46.8 g, 0.076 cm thick, 6.26 cm wide and 60 cm high.

2.2. Parametric study

In the parametric study performed in this project were made some model simplifications in order to reduce the number of variables present in the comparative study. In the standard configuration to be used in the study all fuel assemblies were considered "fresh", ie without any burning as if they were newly manufactured. Thus, the comparative study was conducted for a whole new nucleus. Modeling has also been simplified and structures like plugs and

miniplates presented in figure 1. The simplified configuration that will be the reference of the study.



Figure 5: IEA-R1 standard configuration – E01

Then a variation of the E01 configuration was studied, where the graphite reflectors were exchanged for beryllium, in a first approximation only in the reactor limits (E02) and in a second approximation in the whole reactor (E03). These settings are show in the below figures 6 and 7:



In the next parametric study 4 fuel assemblies were removed in 2 different arrangements E04 and E05 which are seen in figures 8 and 9 below:



Finally a combination of fuel assemblies removal and reflector material change was performe d to give the E04A, E04B, E05A and E05B configurations which can be seen in the figures 10, 11, 12 and 13 below:

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Figure 10: Configuration E04A





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Figure 11: Configuration E04B



The calculations are based on the SERPENT code which is a 3-dimensional Monte

Carlo code with static and burnup neutronic capabilities [9][10] and the thermal hydraulic calculations are based on the COBRA code [11].

3. METHODS AND DATA

2.1. Parametric studies results

The main objective of this work was to determine a new configuration for the IEA-R1 reactor in order to ensure the increase of neutron flux magnitude in the beryllium element irradiators in relation to a reference configuration called E01. Thus, eight different configurations were taken, where two of them (E02 and E03) were changed only the reflectors surrounding the reactor core, two others where only four fuel assemblies were removed in different arrangements (E04 and E05), with their replacement with beryllium reflectors and four where reflector changes were combined with the removal of fuel assemblies (E04A, E04B, E05A and E05B). The full description of these settings can be seen in section 2.2.

In figures 14, 15, 16 and 17 the neutron thermal flux in the four radiators in each of the investigated configurations is shown. The thermal flux was taken to E < 0.625 e.V. over the 10 positions used for material irradiation and its results can be seen below. Table 1 shows the

average gain on each detector from the reference setting. The computational modeling was performed with SERPENT code that uses the Monte Carlo method in its calculations with 8 million stories. Since flow uncertainty is around 0.05%, this uncertainty will be omitted from the graphs as they do not become visible.



Figure 14: Thermal flux distribution at beryllium element irradiator (EIF)



Figure 16: Thermal flux distribution at beryllium element irradiator (EIBRA1)



Figure 15: Thermal flux distribution at beryllium element irradiator (EIBe)



Figure 17: Thermal flux distribution at beryllium element irradiator (EIBRA2)

| beryllium element | | | F | Reactor co | nfiguratio | 'n | | |
|-------------------|--------|-------|-------|------------|------------|-------|-------|-------|
| intuition | E02 | E03 | E04 | E05 | E04A | E04B | E05A | E05B |
| EIF | 6.76 | -1.03 | 10.71 | 13.25 | 12.60 | 11.27 | 12.69 | 6.91 |
| EIBe | -3.62 | -4.54 | 13.43 | 18.88 | 10.64 | 9.65 | 15.13 | 18.88 |
| EIBRA1 | -24.43 | 35.62 | 13.62 | 13.87 | 29.56 | 49.96 | 30.02 | 52.27 |
| EIBRA2 | -14.97 | 2.38 | 11.82 | 13.68 | 17.60 | 14.82 | 18.71 | 10.88 |

Table 1: Average flux gain compared to default configuration (E01)

Figures 14, 15, 16 and 17 show that simple replace of the reactor reflectors materials (E02) causes a displacement of the spatial distribution of the thermal neutron flux in the nucleus leading to an average lose radioisotope production capacity in the beryllium element irradiators, since the flux is smaller.

The removal of fuel assemblies from the core (E04 and E05), in turn, causes a considerable increase in the thermal neutron flux in the beryllium element irradiators. This is because power is proportional to the neutron flux in the fuel. With core compaction, the neutron flux should rise in the remaining fuel assemblies to compensate for fuel removal. In this case, the highest gain setting is E05, where the flow of thermal neutrons increases by an average of 15%. The reason why we chose to only remove four fuel assemblies from the twenty in the core is to avoid getting too close to the fuel thermal limit during operation.

The combination of the strategy of changing the beryllium graphite reflector with the removal of fuel assemblies E04A, E04B, E05A and E05B proved to be very advantageous, especially for the E04B and E05B configurations. In this study a total gain in neutron beryllium element irradiators was observed for the E04B configuration of approximately 21.42% and for the E05B configuration of approximately 22.24%.

2.2. Safety parameters

Since in this study, the most adequate configuration was the E05B, the next step is to verify the neutronic characteristics of this configuration so that the safety of the reactor is maintained. For this, the moderator and fuel temperature reactivity coefficients, the reactivity defects during the beginning of the operation, the kinetic parameters: delayed neutron fraction (β_{eff}) and mean generation time of the prompt neutrons (Λ) and temperature in the hottest fuel channel were verified. These data are presented in tables 2, 3, 4 and 5 and figure 18. All data were calculated using the SERPENT code, except for the hot channel temperature that was calculated with COBRA code. All parameters were estimated at cycle start (BOC).

| Effect | Reactivity (pcm) | | | |
|-------------------------------|-------------------|--------------------|--|--|
| | E01 Configuration | E05B Configuration | | |
| Temperature defect | -53 | -43 | | |
| Power defect | -96 | -64 | | |
| Xenon balance | -2397 | -2567 | | |
| Power defect Xenon balance | -96 -2397 | -64 -2567 | | |

Table 2: Reactivity defects during reactor start-up (BOC).

* The uncertainties of the calculations are negligible.

The reactivity defects showed little variation from the default setting, which does not greatly affect operation. There was a slight increase in the negative reactivity inserted by xenon in the order of 7%, which may cause a reduction in the margin of maneuver and an additional reduction of each reactor cycle, since there would already be a reduction caused by the smaller amount of fissile material and There will be a new one due to the reduction of the effective multiplication factor during operation thanks to xenon.

| Table 5. Temperature reactivity coefficients (BOC). | | | | |
|-----------------------------------------------------|-----------|---------------------------------|-------|-------------------|
| Temperature (°C) | | Reactivity coefficient (pcm/°C) | | |
| Moderator | Fuel | Configuration | | |
| | | E01 | E05B | |
| 20 - 40 | 80 | -7.07 | -3.70 | Moderator |
| 40 - 60 | 80 | -1.78 | -1.59 | |
| 40 - 80 | 80 | -8.04 | -7.80 | |
| 80 | 20 - 50 | -0.95 | -1.33 | |
| 80 | 50 - 100 | -1.36 | -2.73 | Fuel (Doppler) |
| 80 | 100 - 200 | -2.02 | -1.02 | (Boppier) |

Table 3: Temperature reactivity coefficients (BOC).

* The uncertainties of the calculations are negligible.

The temperature reactivity coefficients showed a slight reduction with respect to the moderator temperature variation but remain within safe values for reactor operation.

| | 1 | | | |
|-------------------|---------------|---------|--|--|
| | Configuration | | | |
| | E01 | E05B | | |
| β_{eff} | 0.00726 | 0.00717 | | |
| $\Lambda (\mu s)$ | 46 | 57 | | |

 Table 4: kinetic parameters (BOC)

* The uncertainties of the calculations are negligible

The kinetic parameters of not show a large variation with respect to the reference configuration (E01) for the proposed new configuration (E05B), thus ensuring a similar behavior during operation.

Table 5: Effective multiplication factor (k_{eff}) in cold zero power reactor with control banks inserted .

| Configuration | $\mathbf{k}_{\mathrm{eff}}$ |
|---------------|-----------------------------|
| E01 | 0,98504 (3) |
| E05B | 0,94329 (3) |

As the amount of fuel has been reduced the control bars become more effective and the safety of shutdown is ensured. The high value of k_{eff} with control bars inserted is due to the fact that in this study all fuel assemblies were taken as fresh fuel compounds, which in practice does not occur during operation. Since on average only 2 new fuel assembly enter each new IEA-R1 reactor operating cycle.



Figure 18: Hot channel temperature distribution as a function of plate height

The temperature in the hottest reactor channel indicates complete safety during operation in this new configuration as the temperature peak is well below the fuel jacket melting limit.

2.3. Fuel consumption and transuranic production

Once the thermal and neutron characteristics inherent to the reactor safety were verified, a last investigation was made regarding the consumption of U-235 and Plutonium production (Pu-239 and Pu-241). The data can be seen in figures 19, 20 and 21 and are given in kg per ton of fuel material.



Figure 19: Fuel consumption (U-235) as a function of time



Figure 20: Transuranic production (Pu-239) as a function of time

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Figure 21: Transuranic production (Pu-241) as a function of time

Figure 19 indicates a higher consumption of U-235 per ton of fuel, which is expected due to increased power density in the fuel assemblies. Figures 20 and 21 indicate an increase in fissile plutonium produced as a result of increased neutron flux in fuel assemblies.

2.4. Thermal neutron flux in the new beryllium element irradiators

Figure 22 shows the thermal neutron flux in the new beryllium radiator elements of configuration E5B (figure 10), numbered 1, 5, 16, and 20. Where the radiator element in position 5 will be called IR1, position 1 of IR2, position 20 of IR3 and position 20 of IR4.



Figure 22: Thermal neutron flux in the new beryllium element irradiators

Figure 22 shows that thermal flux has the magnitude of the order of the other detectors and is

suitable for use in sample irradiation, thus increasing the irradiation capacity of the IEA-R1 reactor.

4. CONCLUSIONS

The parametric study demonstrated a significant increase in neutron flux (~ 22%) for the configuration chosen in the parametric study as the best option (E05B). This setting was selected due to the higher average flow increment. In addition to the gain in flow radioisotope production, this configuration also provides 4 more beryllium irradiating elements, similar to the EIBRA1 and EIBRA 2, which will result in a larger volume of sample irradiations per time interval.

The E05B configuration has been shown to meet the necessary neutron and thermal requirements for safe operation, as well as a considerable increase in control bar effectiveness, which contributes to reactor safety. However, as can be seen from the results analysis, there was a significant increase in fissile material consumption (~ 20%) and a slight increase in reactivity defect due to xenon (~ 7%). These factors will cause higher fuel demand and shorter operating cycles. There was also an increase in fissile plutonium production (~ + 40% Pu-241 and ~ + 14% Pu-239).

In a future work will study the operating cost of this new configuration and the irradiation gains, in an analysis to discuss the economic viability of this new configuration (E05B). More detailed thermo-hydraulic studies should also be performed to ensure concept safety and accident analysis.

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