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USING A BERYLLIUM IRRADIATION ELEMENT

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ABSTRACT

The IEA-R1 reactor has undergone the modernization to increase its operating power to 5 MW, in order to allow a more efficient production of the ^{99}Mo radioisotope. An irradiation element made of Be was acquired for the reactor, and studies were initiated for determining its performance when compared to other irradiation elements currently in use in the reactor.

The results show some advantages of the Be irradiation element for producing ^{99}Mo: the epithermal neutron flux in the Be irradiation element approximately 15 % greater than those in the water and graphite irradiation elements; the negative reactivity introduced in the reactor by the Be irradiation element is substantially smaller than the those introduced by the other elements: -1636 pcm for the Be element, -2568 pcm for graphite element and –2977 pcm for the water element.

It is concluded that the production of the ^{99}Mo radioisotope with the Be irradiation element can be increased by 15 % in the IEA-R1m reactor. It also requires less fuel for the reactor operation due to the smaller negative reactivity introduced in the reactor core.

INTRODUCTION

IPEN has been assisting approximately one million patients a year with its radioisotope production for the diagnosis and therapy of several illnesses. One of the proposed uses for the IPEN's research reactor is the production of ^{99}Mo through the irradiation of molybdenum oxide. Today, the weekly demand of that radioisotope, imported from Canada, is about 6290 GBq (170 Ci). IPEN produces ^{99m}Tc generators starting from the ^{99}Mo and distributes to medical clinics and hospitals in the country.

This work is part of a research program for obtaining radioisotopes not available in the country and sought by the medical community. In this program the IEA-R1m reactor underwent a reform aiming at attending that purpose in which its power increased from 2 to 5 MW, and has been acquired a Beryllium Irradiation Element (EIBe) for producing ^{99}Mo.

The method for ^{99}Mo production chosen in IPEN is the neutron capture in the ^{98}Mo when irradiated in the reactor under the form of natural molybdenum oxide (MoO_3) [1]. The EIBe, occupying the space of a fuel element in the reactor support plate, consists of a Be cube with two holes of 3,3 cm, aligned in diagonal, which are denominated irradiation channels.

This work seeks to study this irradiation element by comparing it with the other irradiation elements currently in the IEA-R1m reactor, namely the Water Irradiation Element (EIA) and the Graphite Irradiation Element (EIGRA). The paper starts presenting the basic ideas and methods which were used for comparing the three irradiation elements. The measurements required by the analysis, namely, reaction rates and reactivities, are then presented, followed by the results and the conclusions.

ANALYSIS OF THE BERYLLIUM IRRADIATION ELEMENT

The ideal characteristics of an irradiation element are 1) presenting a great number of irradiation positions; 2) presenting irradiation positions with a reasonable volume in order to accommodate different types of materials in different forms and sizes; 3) providing high neutron fluxes in the irradiation positions; 4) presenting neutron fluxes with small gradients in the irradiation positions; and 5) introducing small negative reactivity into the core in order to minimize excess reactivity decrease. The best irradiation element would have to attend the characteristics mentioned above. For the IEA-R1m reactor the first two characteristics are fixed since the irradiation elements shall occupy a fuel element position in the reactor support plate, and, practically, provide the same space for irradiation.

For considering the third and fourth characteristics, the approach taken in this work was to perform a series of irradiation experiments in the IEA-R1m reactor. Neutron reaction rates were measured with activation foils of Au, Mo, and In along the irradiation elements and inside the standard irradiation capsule used in the reactor [2,3,4]. It was used Cd covers for the foils in order to determine the epithermal neutron reaction rates that are more appropriate for Mo isotope production. For the fifth characteristic, the determination of the reactivity perturbation caused by the irradiation elements in the core was done through inference against the required motion of the reactor control the control rods in order to maintain criticality. With control rod calibration curves for each of the core configurations used in the experiments, it was determined the core excess reactivity and, consequently, the impact that each of the irradiation elements cause when placed in certain positions of the core.

The radioisotope of interest, ^{98}Mo, has neutron-gamma cross-section with resonance in the epithermal energy range between 10 eV and 10 KeV. The beryllium is a highly dense material with a low absorption cross-section and large scattering cross-section which makes it an excellent neutron moderator. Since it is not as light as H, the neutron spectrum in a beryllium media is somewhat harder than that found in a water media. Moreover, the Be has the interesting (n,2n) reaction which produces neutrons and increases the reactor effective multiplication factor.

The beryllium irradiation element, EIBe, has the dimensions of the standard fuel element, 60x8x8 cm^3, and the two irradiation channels can each accommodate up to 8 capsules in the vertical position. The other irradiation elements also occupy a similar volume of in the core. In the EIA and in EIBe, the 16 cylindrical irradiation capsules are positioned in the vertical direction, and in the EIGRA, the same capsules are positioned in the horizontal direction and amounts to 24.

Table 1 outlines the experiments performed the reactor. In the experiments labeled A were measured reaction rates along the irradiation elements, and in the experiments labeled R were measured reaction rates inside the irradiation capsules. It was considered two irradiation locations: position 45 nearer to the core reflector and position 65 nearer to the center of the core. For the measurements labeled A the activation foils or wires were mounted inside the capsules on an aluminum support; the capsules which were not sealed, were filled with water. For the experiments labeled R the procedure was the same except that three foils were placed inside the irradiation capsule and one outside the capsule. The capsule was filled with MoO_3 powder in order to emulate more closely the actual irradiation environment for the production of ^{99}Mo, and positioned from top to bottom in the sixth shelf of the irradiation elements. The irradiation time was 60 minutes with the IEA-R1m reactor operating at 20 kW. The counting system was based on a NaI(Tl) detector, model 905 EG&G Ortec, and a multichannel analyzer, model Seiko 7800 EG&G Ortec.

Table 1. Experiments performed for analyzing the irradiation elements for production of ^{99}Mo in IEA-R1m reactor.

	LOCATION	EXPERIMENT DESCRIPTION	OBJECTIVE
A1	EIBe in position 45	Irradiation of 8 Au foils in channel A Irradiation of 8 Au foils covered with Cd in channel A	Distribution of thermal and epithermal reaction rates along the EIBe channel A
A2	EIBe in position 45	Irradiation of 8 Au foils in channel B Irradiation of 8 Au foils covered with Cd in channel B	Distribution of thermal and epithermal reaction rates along the EIBe channel B
A3	EIBe in position 45	Irradiation of 8 In foils covered with Cd in channel A Irradiation of 8 In foils covered with Cd in channel B	Distribution of fast neutron energy reaction rates along the EIBe channels A and B
R1	EIBe in position 45	Irradiation of Al foils doped with Au inside a standard irradiation capsule filled with MoO_3	Distribution of reaction rate inside the standard irradiation capsule filled with MoO_3
A4	EIBe in position 65	Irradiation of 8 Au foils in channel A Irradiation of 8 Au foils covered with Cd in channel A	Distribution of thermal and epithermal reaction rates along the EIBe channel A
A5	EIBe in position 65	Irradiation of Mo wires in channel A Irradiation of Mo wires in channel B	Distribution of Mo reaction rates along the EIBe
A6	EIBe in position 65	Irradiation of Mo wires covered with Cd in channel A Irradiation of Mo wires covered with Cd in channel B	Distribution of Mo reaction rates along the EIBe
R2	EIBe in position 65	Irradiation of Al foils doped with Au inside a standard irradiation capsule filled with MoO_3	Distribution of reaction rate inside the standard irradiation capsule filled with MoO_3
A7	EIBe in position 65	Irradiation of 8 Au foils in channel A Irradiation of 8 Au foils covered with Cd in channel A	Distribution of thermal and epithermal reaction rates along the EIBe channel A
A8	EIA in position 65	Irradiation of 8 Au foils in channel A Irradiation of 8 Au foils covered with Cd in channel A	Distribution of thermal and epithermal reaction rates along the EIA channel A
A9	EIGRA in position 65	Irradiation of 8 Au foils in channel A Irradiation of 8 Au foils covered with Cd in channel A	Distribution of thermal and epithermal reaction rates along the EIGRA channel A
R3	EIBe in position 65	Irradiation of Mo wires inside a standard capsule filled with MoO_3 – positions 5, 6 e 7	Distribution of reaction rate inside the standard irradiation capsule filled with MoO_3 in the EIBe
R4	EIA in position 65	Irradiation of Mo wires inside a standard capsule filled with MoO_3 – positions 5, 6 e 7	Distribution of reaction rate inside the standard irradiation capsule filled with MoO_3 in the EIA
R5	EIGRA in position 65	Irradiation of Mo wires inside a standard capsule filled with MoO_3 – positions 5, 6 e 7	Distribution of reaction rate inside the standard irradiation capsule filled with MoO_3 in the EIGRA

OBTAINED RESULTS

Some of the results obtained in the experiments are presented in this section. Figure 1 shows the reaction rates from the In foils covered with Cd in channels A and B of the EIBe, with channel B closer of the center of the core. The counting presents excellent statistics and in some cases they were repeated up to 4 times as it can be observed in the figures, and is presented per unit of time and foil mass.

There are differences between the two channels from the irradiation elements depending on where they are located in the core. The channel closer to the center of the core present higher neutron fluxes. It is noticed that there is a difference of reaction rate levels between the two channels of about 35 %. Also, positions 4, 5, 6 and 7, referring to the numbered shelves in the irradiation elements, are those of higher reaction rates and, consequently, they may provide greater production of ^{99}Mo. In channel B, position 3, can be produced an equivalent activity to that in channel A, position 4.

The axial neutron flux gradient for all irradiation elements are very similar. The difference for the reaction rates from positions 4 and 5 (of greater neutron flux) is about 16 %.

Figure 2 presents the reaction rates in the Au foils covered with Cd in the EIBe irradiation element, located in position 65 of the reactor support plate. The reaction rate in the position 65, closer to the core center, is about 17 % superior than that in the position 45, closer to the reflector, suggesting a similar increase in the production of ^{99}Mo activity. Figure 3 presents the reaction rates in the Au foils covered with Cd in the EIGRA irradiation element. The measurements were made in the position 65 of the reactor support plate.

Regarding the reaction rate distribution, it is seen that all the three irradiation elements present similar ones along the element. This means that the three irradiation elements present similar volumes for the production of ^{99}Mo. The larger or smaller radioisotope production is dependent on the intensity of the epithermal neutron flux in each irradiation element. Reaction rates of foils, covered with Cd, are proportional to the epithermal neutron flux in the reactor location since the thermal neutron flux is absorbed by the Cd cover.

Comparing Figures 2 and 3 and other results not shown here, it is seen that the epithermal neutron flux in the EIBe be 15 % superior than that in the EIGRA, and approximately 13 % superior than that in the EIA. The thermal neutron flux in the EIGRA is 18 % superior than that in EIBe and 4 % superior than that in EIA. These results show that the Be irradiation element is superior, when compared to the of water and graphite irradiation elements, for the production of radioisotopes that require epithermal neutrons, which is the case of the ^{99}Mo, but by a small margin. For radioisotopes that require thermal neutrons the graphite element is the superior one.

Figure 4 shows the reaction rate distribution inside of an irradiation capsule located in the Be irradiation element. There is a decrease in the reaction rates from outside to inside the capsule, followed by an almost constant value up to its center. This indicates that the activity production inside the capsule is rather uniform.

The negative reactivity introduced in the reactor by the three irradiation elements were also measured. The decrease in the reactor excess reactivity caused by the Be irradiation element is substantially smaller than those caused by the other elements: -1636 pcm for the EIBe, -2568 for the EIGRA and –2977 pcm for the EIA. Consequently, the Be irradiation element requires less fuel in order to maintain the necessary reactor excess reactivity.

CONCLUSIONS

The obtained results show some advantages for the EIBe in relation to the other irradiation elements for the production of ^{99}Mo in the IEA-R1m reactor. It is pointed out an epithermal neutron flux 15 % superior in the EIBe than that in the EIGRA and approximately 13 % superior than that in the EIA. This means that EIBe allows a production of ^{99}Mo about 15 % superior than what is possible with the irradiation elements currently available in IEA-R1m reactor.

The flux gradient and irradiation volumes for the three irradiation elements are similar, showing that the core neutron flux distribution depends, basically, on the control rods positions.

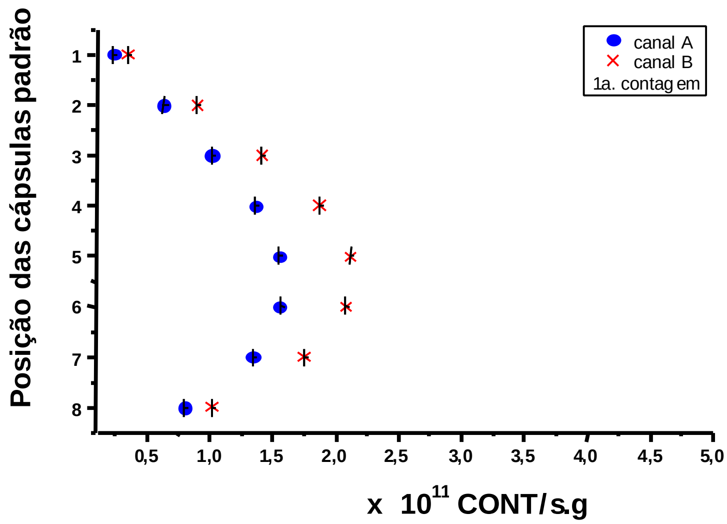

Figure 1. Reaction rates in the In foils covered with Cd in the EIBe, channels A and B, and core position 45.

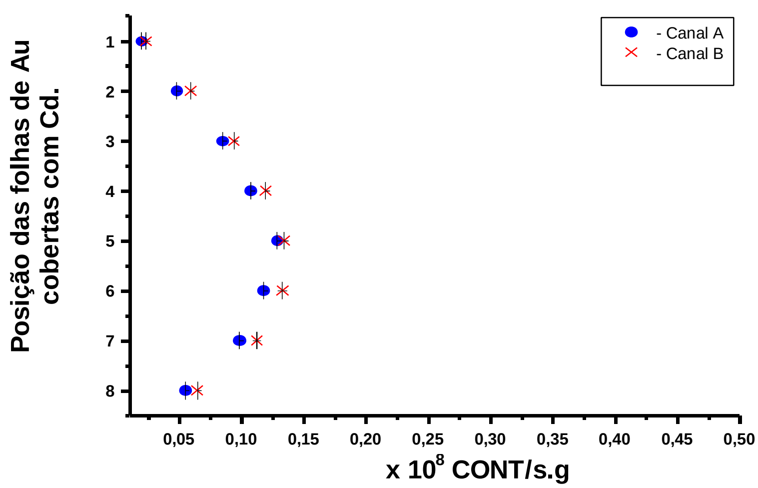

Figure 2. Reaction rates in the Au foils covered with Cd in the EIBe, channels A and B, and core position 65.

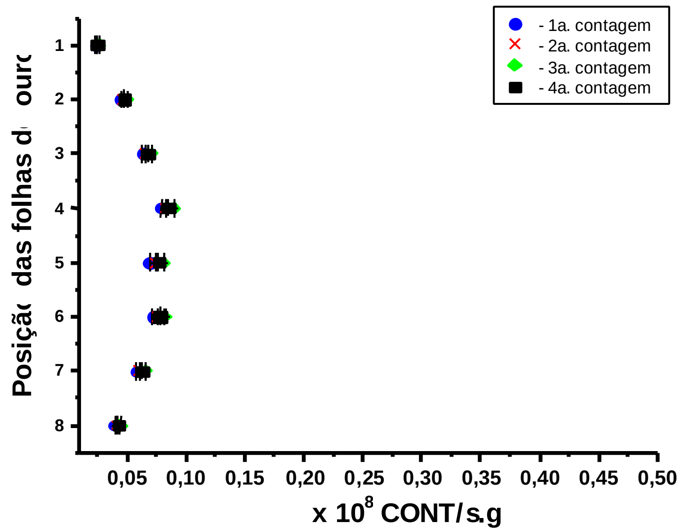

Figure 3. Reaction rates in the Au foils covered with Cd in the EIGRA, channels A and B, and core position 65.

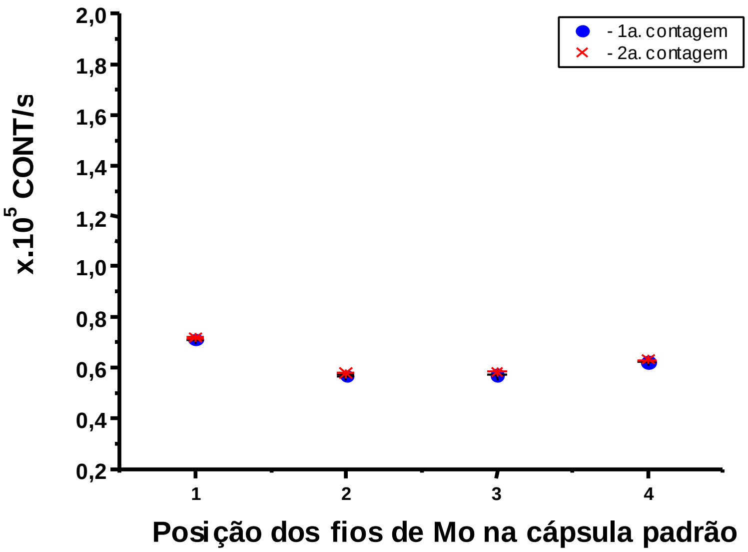

Figure 4. Reaction rates in the Mo wires inside the irradiation capsule filled with MoO_3 in the EIBe, channel A, and core position 65.

The flux gradient and irradiation volumes for the three irradiation elements are similar, showing that the core neutron flux distribution depends, basically, on the control rods positions. Irradiation positions 4, 5, 6 and 7 are those of higher neutron flux regardless the location of the irradiation element in the core; consequently, these are the positions which may provide the higher production of ^{99}Mo. The difference of neutron flux level between channels A and B is about 35 % for the EIBe located in position 45, indicating an important dependency on where the irradiation element is placed in the core. The difference between positions 4 and 5 (of maximum neutron flux) is about 16 % for any channel or irradiation element location.

In terms of reactivity the Be irradiation element presents the smallest negative insertion in the core, demanding less fuel elements in order to keep the same core excess reactivity.

It is concluded that the production of the ^{99}Mo radioisotope with the Be irradiation element can be increased by 15 % in the IEA-R1m reactor. It also requires less fuel for the reactor operation due to the smaller negative reactivity introduced in the reactor core.

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