Neutronic analysis for the fission Mo-99 production by irradiation of a LEU target at RECH-1 reactor

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Abstract. For the purpose of developing the capability to produce fission ^{99}Mo, the Chilean Nuclear Energy Commission is participating in the IAEA Coordinated Research Project: "Developing Techniques for Small Scale Indigenous Mo-99 Production using LEU Fission or Neutron Activation". Fission ^{99}Mo will be produced irradiating, at RECH-1 reactor, a target made of a LEU metallic uranium foil held between two concentric aluminum tubes. KAERI will provide the LEU foil.

Neutronic calculations were performed to estimate the fission products activity for a 13 grams LEU foil annular target, which will be irradiated at the level power of 5 MW during 48 hours.

1.- Introduction

The RECH-1 research reactor is a pool type reactor with a nominal thermal power of 5 MW. This reactor is operated by the Chilean Nuclear Energy Commission (CCHEN) at La Reina Nuclear Center. The RECH-1 is a light water-moderated, water-cooled and beryllium-reflected reactor and it employs a flat plate MTR-type fuel with low enriched uranium. Six blade-plates control absorbers pass through the core in three groups of two.

The present core configuration of the RECH-1 reactor, Nº 62 (Fig. 1), has 32 LEU fuel elements containing U_3Si_2-Al. These LEU fuel elements were built by the Chilean Fuel Fabrication Plant with a uranium density of 3.4 g/cm^3 [1]. The technical specifications of these fuel elements were developed by the Chilean Manufacturer based on the original HEU assembly and approved by the reactor operator.

2.- Description of the preliminary target irradiation system

The preliminary target irradiation system is formed by a LEU (19.75% of ^{235}U) metallic uranium foil of 13 grams of 50 mm x 100 mm and 130 microns of thickness wrapped in a thin nickel fission product-recoil barrier of 15 microns thickness. KAERI will provide the LEU foil.

The metallic uranium foil with its nickel coating surrounds an aluminum tube of 152 mm in length, 27.99 mm outer diameter and 26.42 mm inner diameter. This inner tube has an undercut to position the foil. This set, as well, is surrounded by an aluminum tube of 28.22 mm inner diameter, 30.15 mm outer diameter and 152 mm in length.

Outer and inner cylinders are swaged to give good thermal contact. By means of a tool specially designed, the inner tube is become deformed to produce a good contact between the different materials that constitute the target. The design of the target irradiation system was done to maximize the target heat dissipation by the coolant flow inside the target. The Fig. 2 shows the preliminary target irradiation system [2].

	1	2	3	4	5	6	7	8	9	10
G	Bk	Be	Be	Be	Be	Be	Be	Be	Be	Bk
F	Bk	Al	LR60 4.183	LR61 5.143	LR56 6.691	LR57 6.791	LR62 5.153	LR63 4.198	Al	LREX1
E	Be	LR53 5.359	LR51 7.632	LR45 15.936	LR01L 41.110	LR02L 41.882	LR46 16.543	LR50 7.880	LR55 5.447	Be
D	Al	Be	LR47 9.567	LR41 20.744	CI	CI	LR42 21.833	LR48 10.310	Be	Al
C	Be	LR52 5.352	LR49 7.177	LR44 15.703	LR03L 37.931	LR04L 37.837	LR43 16.755	LR82 7.710	LR54 5.644	Be
B	Be	Be	LR66 3.974	LR67 4.610	LR58 5.863	LR59 6.145	LR64 4.721	LR65 4.399	Be	Be
A	R 11/8”	Al	Be	Be	CI	CI	Be	Be	TI	Be
H	Al	Bk	R 1 ½”	CI	PIBM	PIBM	CI	Pb	Pb	Al Fn

Fig. 1.- Present core configuration, Nº 62, of the RECH-1 reactor with the average burnup, in percent, at beginning of cycle (BOC) for each fuel element

3.- Neutronic and activity calculations

The neutronic calculations were performed using WIMS-D [3], [4], [5] and CITATION [6] codes, neutronic programs used routinely in the fuel management of this reactor. Different cell models were needed to generate appropriate cross sections for the various reactor regions in structures of three and five energy groups. The WIMS-D code was used to generate the multigroup nuclear constants library for different regions as a function of burnup. The main transport calculation was performed in 20 and 36 energy groups, condensing to 5 and 3 groups respectively for the diffusion theory calculations in two and three dimensions (Table 1).

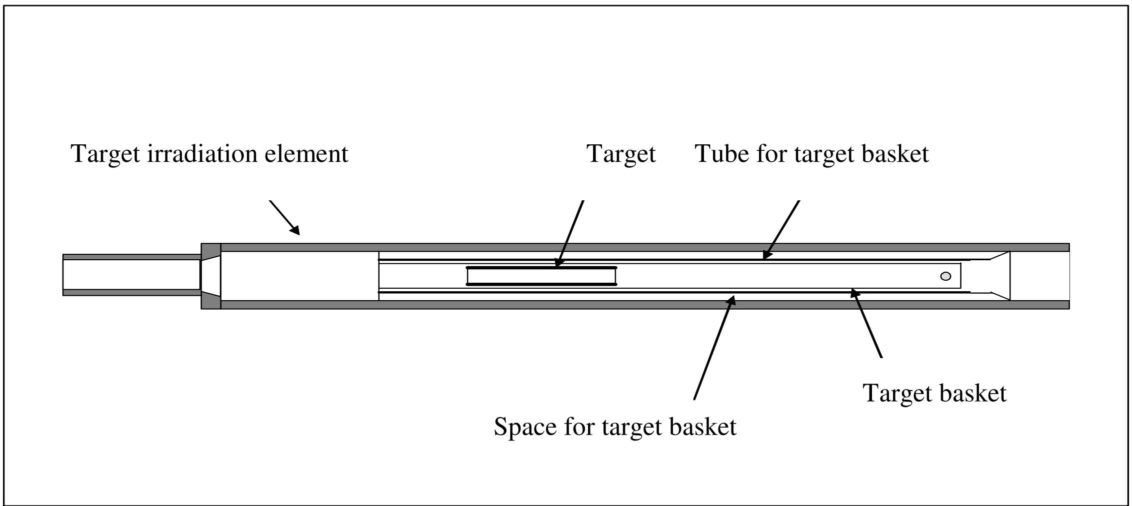

Fig. 2.- Preliminary target irradiation system

In order to calculate the cross sections for fuel element, a unit cell formed by half thickness of meat, the cladding, half thickness of the coolant channel and a homogeneous extra zone was used. The extra region includes the aluminum in the plates beyond the width of the meat, the aluminum side plates, the water beyond the width of the meat and the water channels surrounding the fuel element. In a diffusion calculation, a fuel element will be represented by three zones, the central zone is a homogenization of the meat, cladding and the cooling and the lateral zones are identical to each other and are formed by a homogenous mixture of aluminum and water. The nuclear constants for beryllium elements, aluminum elements, blanking elements, absorber tailplates and the water as a reflector have been obtained using different macrocell models [7].

The neutronic calculations for the core configuration have been performed using the diffusion code CITATION in two dimensions, with a five-group structure and bucklings imposed for the axial dimension. Calculations in three dimensions were carried out with a three-group structure.

Table 1.- Broad group energy structures used in the diffusion theory calculations

Group	Upper energy (5-group set for 2D calculations)	Upper energy (3-group set for 3D calculations)
1	10 MeV	10 MeV
2	0.821 MeV	0.821 MeV
3	5,530 eV	0.625 eV
4	2.1 eV	—
5	0.625 eV	—

The theoretical neutronic calculations related to fission Mo-99 production are based on the core configuration N° 62, but excluding the experimental fuel element LR-EX-01, [8], in order to simplify these calculations. This fuel element was replaced by a blanking element. The neutronic calculations were performed for a 13 grams LEU foil target of tubular shape, supposing that the target would be introduced in the positions D2 or D5 of the reactor grid (Fig. 1). The results of the neutronic analysis are given for the target irradiation system formed by the element to irradiate targets, the target basket and the target itself (Fig. 2).

The results show that, when the foil is irradiated in the D5 position of the reactor grid with a thermal neutron flux of 8.062×10^{13} n/cm^{2}s, it generates about 7.44 kW, (Table N° 2), representing a heat flow of 144 W/cm^2, with an increase of reactivity of the reactor core of 317 pcm, that is 0.41 \$. In these conditions, the maximum temperature in the target is obtained on the inner surface and its value is 111 ºC, with a margin of 12 °C respect to the onset of nucleate boiling. This maximum temperature is located approximately 10 cm from the top end of the target; that is, 6.9 cm from the top edge of the uranium foil [9].

The previous value of temperature considers a uniform distribution of the heat generated in the plate; therefore the maximum plate temperature that would be obtained considering a not uniform heat distribution would be greater than the one shown here. Taking into account the maximum wall temperature obtained, the non-uniform heat distribution and the limits and conditions authorized, it is necessary to improve the coolant conditions.

	1	2	3	4	5	6	7	8	9	10
G	Bk	Be	Be	Be	Be	Be	Be	Be	Be	Bk
F	Bk	Al	LR60 4.183	LR61 5.143	LR56 6.691	LR57 6.791	LR62 5.153	LR63 4.198	Al	Bk
E	Be	LR53 5.359	LR51 7.632	LR45 15.936	LR01L 41.110	LR02L 41.882	LR46 16.543	LR50 7.880	LR55 5.447	Be
D	Be	Target	LR47 9.567	LR41 20.744	CI	CI	LR42 21.833	LR48 10.310	Be	Al
C	Be	LR52 5.352	LR49 7.177	LR44 15.703	LR03L 37.931	LR04L 37.837	LR43 16.755	LR82 7.710	LR54 5.644	Be
B	Be	Be	LR66 3.974	LR67 4.610	LR58 5.863	LR59 6.145	LR64 4.721	LR65 4.399	Be	Be
A	R 11/8"	Al	Be	Be	CI	CI	Be	Be	TI	Be
H	Al	Bk	R 1 ½"	CI	PIBM	PIBM	CI	Pb	Pb	Al Fn

Fig. 3.- Core configuration with the LEU foil target in the D2 position of the reactor grid

	1	2	3	4	5	6	7	8	9	10
G	Bk	Be	Be	Be	Be	Be	Be	Be	Be	Bk
F	Bk	Al	LR60 4.183	LR61 5.143	LR56 6.691	LR57 6.791	LR62 5.153	LR63 4.198	Al	Bk
E	Be	LR53 5.359	LR51 7.632	LR45 15.936	LR01L 41.110	LR02L 41.882	LR46 16.543	LR50 7.880	LR55 5.447	Be
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A	R 11/8"	Al	Be	Be	CI	CI	Be	Be	TI	Be
H	Al	Bk	R 1 ½"	CI	PIBM	PIBM	CI	Pb	Pb	Al Fn

Fig. 4.- Core configuration with the LEU foil target in the D5 position of the reactor grid

The results of the 3D neutronic calculations are presented in the Table 2. The reactivity increments, when the target irradiation system is introduced in the positions D2 or D5, are acceptable from the point of view of safety for RECH-1 reactor. Therefore, the position D5 would be the best one, nevertheless is necessary to review the thermal-hydraulic analysis and the target irradiation system design or to consider a position of lower thermal neutron flux, like for example D2. Neutronic calculations, but in two dimensions, were carried out for the same target system [10].

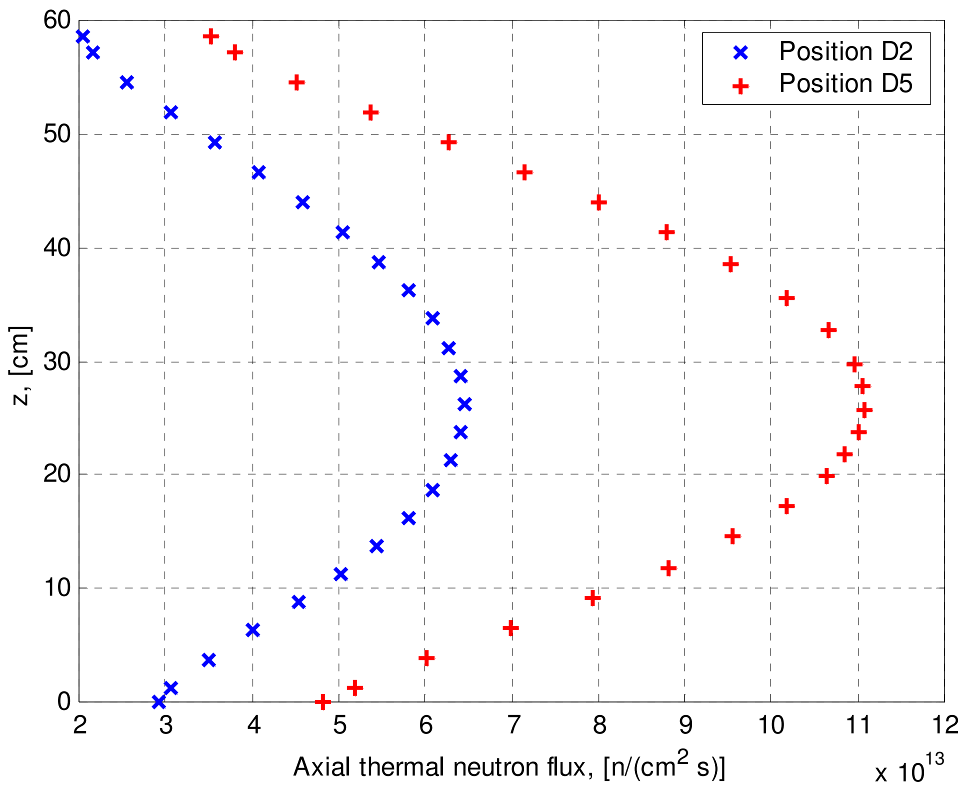

Fig. 5.- Axial thermal neutron flux distributions in the positions D2 and D5

In the Fig. 5, the axial thermal neutron flux distributions in the positions D2 and D5 are shown, when the control plates are withdrawn 70%. The LEU foil has been located where the axial thermal neutron flux is maximum, value which is approximately 26.7 cm from the bottom of the active length of the fuel element.

Table 2.- Effective multiplication factor, reactivity, reactivity variation, average thermal neutron flux and average power generated by the foil in the positions D2 or D5 in the reactor grid

k_{eff}	ρ , pcm	Δρ , pcm	Φ_{th} , n cm^{-2} s^{-1}	P , kW	Position
1.031625	3,066	305	5.37 E+13	4.96	D2
1.037912	3,653	317	8.06 E+13	7.44	D5

The fission product activities were calculated using ORIGEN-S code, SCALE-4.4a system module [11], considering an irradiation time of 2 days for a target containing 13g of 19.75% enriched uranium, at constant thermal neutron flux, 5.37×10^{13} n cm^{-2} s^{-1}, position D2, and 8.06×10^{13} n cm^{-2} s^{-1}, position D5.

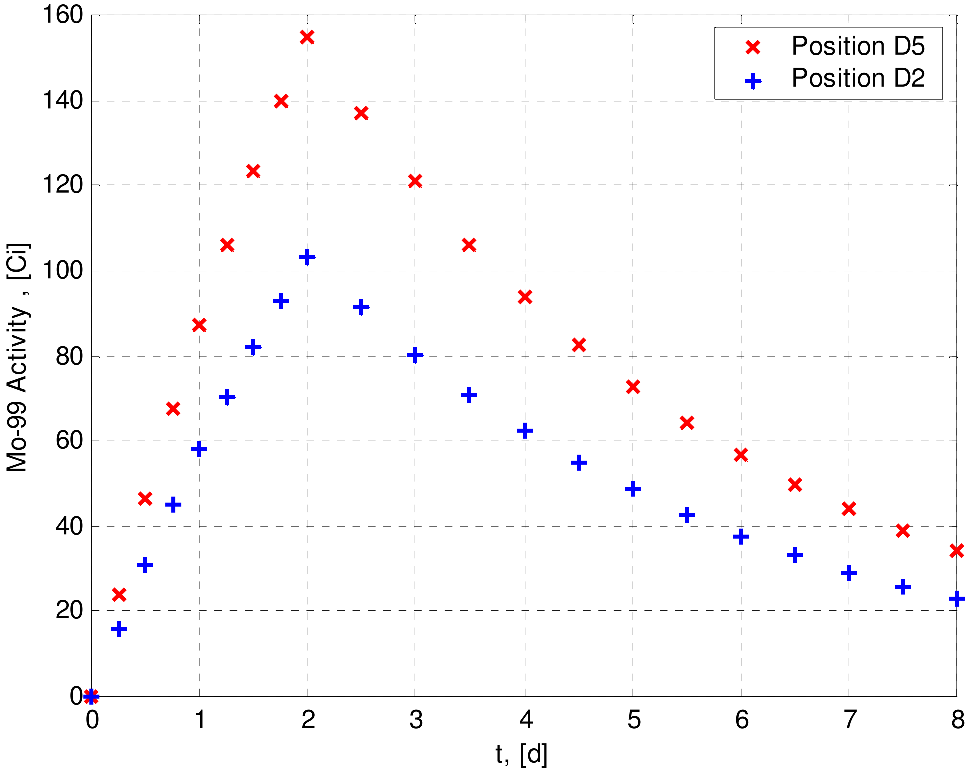

Fig 6.- ^{99}Mo activity for 13 grams of 19.75% enriched uranium and irradiation time of 2 days

The irradiation time has a large impact on specific activity. All the molybdenum isotopes from ^{95}Mo to ^{100}Mo are produced from fission and they continue to build up, making the ^{99}Mo Ci content per gram of molybdenum material decrease with increased irradiation time. A determination of the specific activity of the ^{99}Mo must consider the fission products which are stable isotopes of molybdenum (^{97}Mo, ^{98}Mo and ^{100}Mo) [12].

The results of the ORIGEN-S calculations are presented in the Tables 3 and 4, where A_{sp} is the specific activity of ^{99}Mo in Ci per mg of Mo. At end-of-irradiation with a thermal neutron flux of 8.06×10^{13} n/cm^{2}s, considering 48 hours of irradiation, the activity of ^{99}Mo is 155 Ci, that is 11.92 Ci ^{99}Mo per gram of 19.75% enriched uranium irradiated, the specific activity of ^{99}Mo is 116.24 Ci per milligram of molybdenum (^{97}Mo, ^{98}Mo, ^{99}Mo and ^{100}Mo), the total activity of the actinides, ^{237}U, ^{239}U, ^{239}Np and ^{240}Np, is 21.69 Ci/g U and the total activity of the fission products is 30,800 Ci. Considering that the beginning of the production of generators is 24 hours after the end of irradiation, the initial activity of ^{99}Mo is 121 Ci, and the total activity of fission products is 1,720 Ci.

Table 3.- Activity of ^{99}Mo and specific activity of ^{99}Mo, Ci per mg of Mo, for different irradiation times with a thermal neutron flux of 5.37×10^{13} n/cm^{2}s

t_i, h	**6.0**	**12.0**	**18.0**	**24.0**	**30.0**	**36.0**	**42.0**	**48.0**
^{99}Mo, Ci	15.94	30.92	45.03	58.09	70.57	82.09	93.13	103.00
A_{sp}, Ci/mg Mo	154.71	147.24	140.46	134.44	129.44	124.44	120.32	115.97

At the end of the irradiation, the LEU foil target will be allowed to decay in the reactor pool for a period of 6 hours minimum. Prior to reactor start up, the irradiated target will be removed from the reactor pool and transported to the hot cell for disassembly. Allowing the target to decay in the reactor pool for 6 hours will reduce the end-of-irradiation total fission product activity in 87%**.**

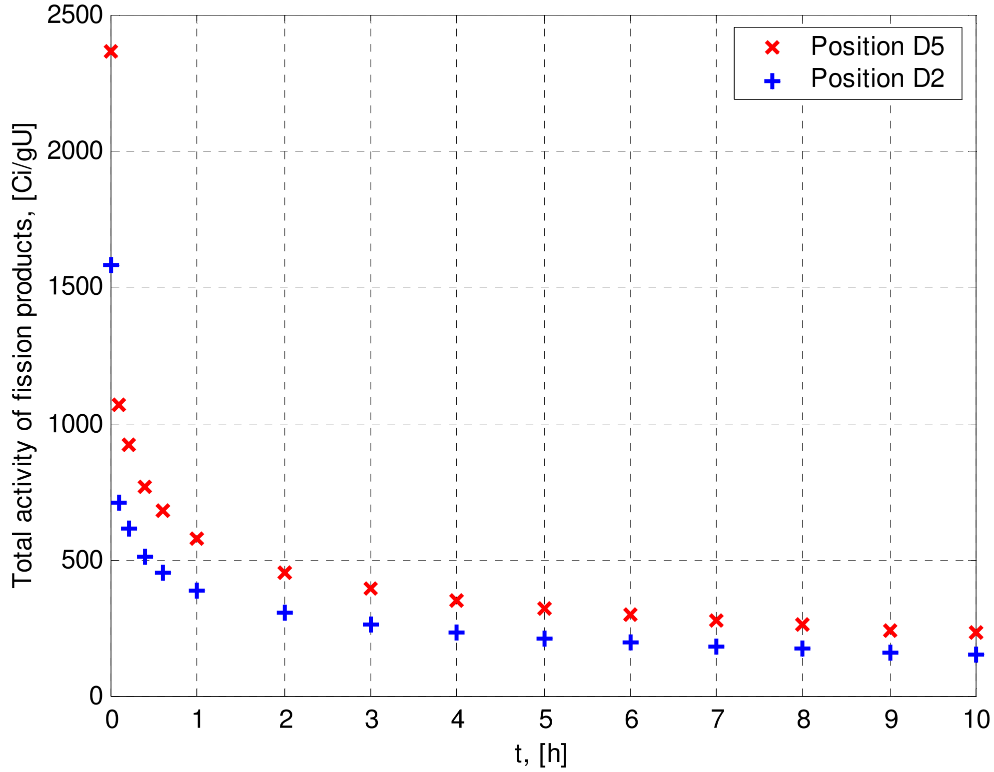

Fig. 7.- Total activity of fission products, A_{Tfp}, in Ci/gU after 48 hours of irradiation and different decay times

Preliminary activity calculations done with ORIGEN-S, show that the irradiation of 13 grams LEU foil target with a thermal neutron flux of 6.6×10^{13} n/cm^{2}s during 48 hours produces an activity of 127 Ci of ^{99}Mo and a total fission products activity of 25,200 Ci at end of irradiation. The initial theoretical activity of ^{99}Mo, 24 hours after the end of irradiation, is 98.8 Ci and the total activity of the fission products is 1,410 Ci.

Table 4.- Activity of ^{99}Mo and specific activity of ^{99}Mo, Ci per mg of Mo, for different irradiation times with a thermal neutron flux of 8.06×10^{13} n/cm^{2}s

t_i, h	6.0	12.0	18.0	24.0	30.0	36.0	42.0	48.0
^{99}Mo, Ci	23.91	46.42	67.69	87.37	106.09	123.37	139.70	155.00
A_{sp}, Ci/mg Mo	154.66	147.04	140.82	134.79	129.49	124.62	120.22	116.24

Similar 2D neutronic calculations were performed for a target containing 8g of uranium 19.75% enriched, supposing that the target would be introduced in the D5 position of the reactor grid. In this case, the reactivity increases in 198 pcm, the mean neutron thermal flux is 6.63×10^{13} n cm^{-2} s^{-1} and the power generated in the target is 4.39 kW. The fission product activities were calculated using ORIGEN-S code, considering an irradiation at constant thermal neutron flux, 6.63×10^{13} n cm^{-2} s^{-1}, and an irradiation time of 72 hours. At end of irradiation, the activity of ^{99}Mo is 105 Ci, that is 13.08 Ci ^{99}Mo per gram of 19.75% enriched uranium irradiated, the specific activity of ^{99}Mo is 103.00 Ci per milligram of molybdenum, the total activity of the actinides is 18.75 Ci/g U and the total activity of the fission products is 1,963 Ci/g U [13].

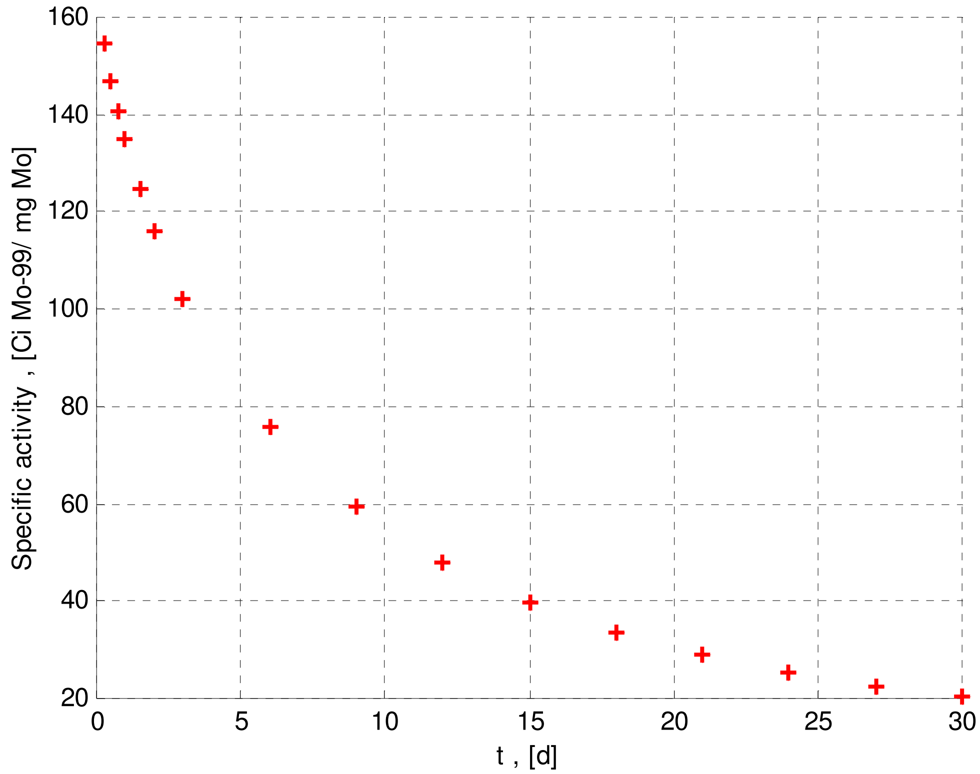

Fig. 8.- Specific activity of ^{99}Mo per mg of Mo (^{97}Mo, ^{98}Mo, ^{99}Mo and ^{100}Mo) for different irradiation times, considering a target of 13 grams of 19.75% enriched uranium

4.- Conclusions

The neutronic calculations were performed for the present core configuration of RECH-1 reactor, with some modifications, using WIMS-D and CITATION codes, neutronic programs used routinely in the fuel management of this reactor. The neutronic calculations were performed supposing that a target, containing 13g of uranium 19.75% enriched, would be introduced in the D2 or D5 positions of the reactor grid.

The results of the 3D neutronic calculations show that, from the point of view of safety for RECH-1 reactor, the target irradiation system could be introduced in the position D5. Nevertheless is necessary to review the thermal-hydraulic analysis and the target irradiation system design or to consider a position of lower thermal neutron flux, like for example D2.

The fission product activities have been calculated using ORIGEN-S, SCALE-4.4a system module, considering an irradiation at constant thermal neutron flux, 5.37×10^{13} n cm^{-2} s^{-1} and 8.06×10^{13} n cm^{-2} s^{-1}, for a target containing 13g of uranium 19.75% enriched, taking into account different irradiation and decay times.

The specific activity decreases with increased irradiation time, therefore a determination of the specific activity of the ^{99}Mo must consider the fission products which are stable isotopes of molybdenum, ^{97}Mo, ^{98}Mo and ^{100}Mo.

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