ANALYSIS OF Mo99 PRODUCTION IRRADIATING 20 %U TARGETS

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ABSTRACT

At present time, CNEA is producing about 800 Ci of Mo99 per week irradiating 90% enriched uranium - aluminum alloy plate targets in the RA3 reactor, a 5 Mw. MTR type one. In order to change to 20% enriched Uranium, and to increase the production to about 3000 Ci per week, some configurations were studied with rod and plate geometry with uranium (20% enriched) - aluminum targets.

The first case was the irradiation of a plate target element in the normal reactor configuration. Results showed a good efficiency, but both reactivity value and power density were too high.

An element with rods was also analized, but results showed a poor efficiency, too much aluminum involved in the process, although a low reactivity and an acceptable rod power density.

Finally, a solution consisting of plate elements with a Zircalloy clading was adopted, which has shown not only a good efficiency, but it is also acceptable from the viewpoint of safety, heat transference criteria and feasibility.

INTRODUCTION

Due to the necessity to change the enrichment of uranium used to produce Molibdenum 99, from 90% to 20%, CNEA (Comisión Nacional de Energía Atómica, Argentina) started studies to implement a method for obtaining Mo 99 from 20% enriched uranium, taking into account the reactor requirements from the point of view of reactivity and power safety and constrains about fabrication and processing of irradiated material.

The RA3 is a 5 MW pool reactor, sited in Centro Atómico Ezeiza, near Buenos Aires. It has 25 (and ocasionally, 27) MTR fuel elements, 19 plates of uranium (20% U_{235}) - aluminum . The core configuration is shown in figure 1.

At this time, the production of Mo99 is performed irradiating uranium with 90% U_{235} and aluminum plates, disposed in an irradiation device in the RA3 core, as shown in figure 2.

Figure 1: RA3 CONFIGURATION

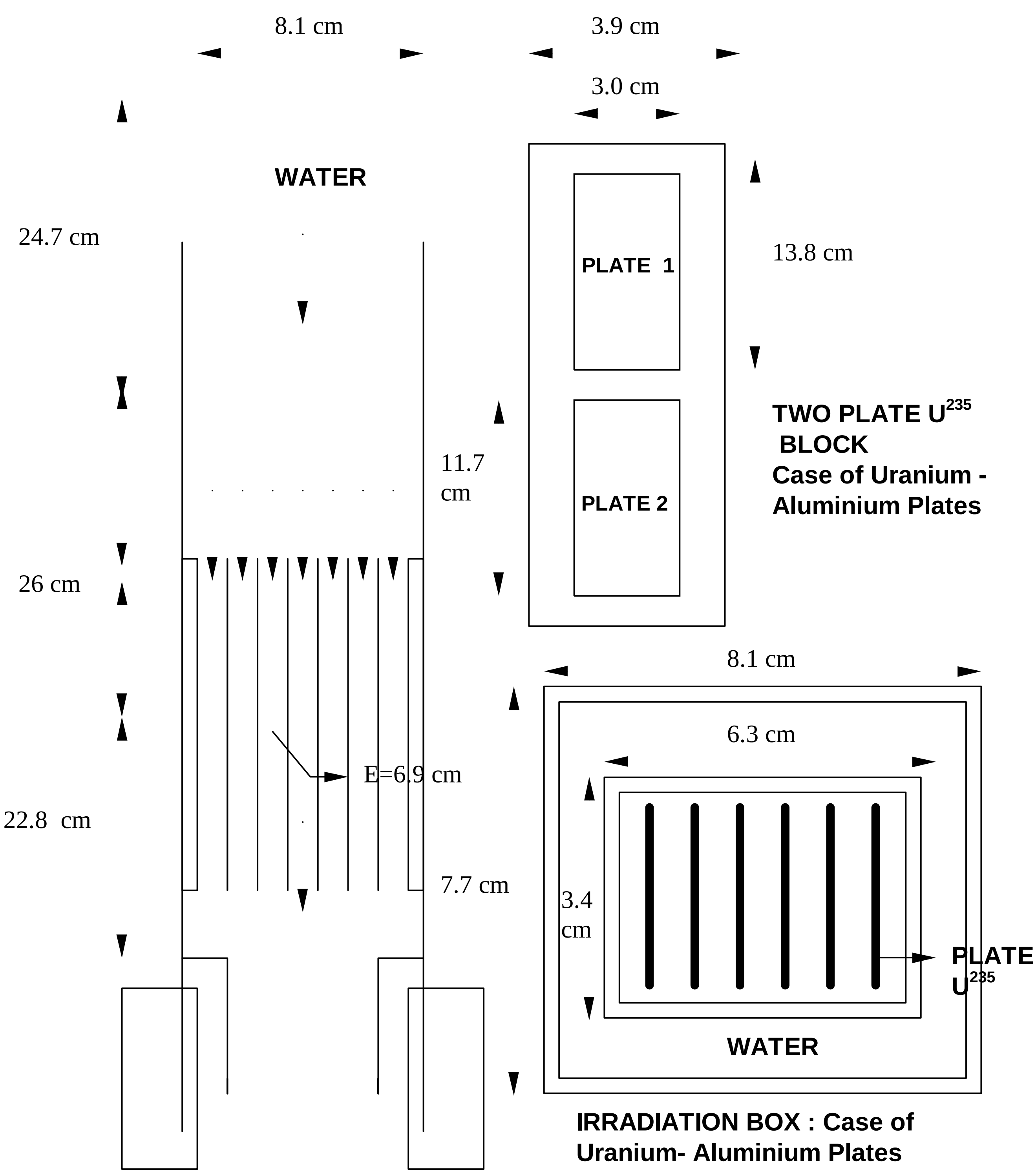
GRAPHITE

	D	E	F	G	H	I
2	IRRAD.	FE 1	FE 2	IRRAD.	FE 3	FE 4
3	FE 5	FE 6	B1	FE 7	FE 8	FE 9
4	FE 10	B4	FE 11	IRRAD.	B2	FE 12
5	FE 13	FE 14	B3	FE 15	FE 16	FE 17
6	FE 18	IRRAD.	FE 19	FE 20	FE 21	IRRAD.
7			FE 26	FE 27		

FE= Fuel Element
B= Control Fuel Element

GRAPHITE

Figure 2: IRRADIATION DEVICE



IRRADIATION BOX : Case of Uranium- Aluminium Plates

The irradiation is performed at the centre of the core (G4 position). The flux at this place is near 10^{14}n/cm2seg. Due to the physical charactheristics of this configuration, both reactivity and power of the device must be considered carefully from the point of view of reactor safety.

Even having changed the enrichment of the uranium plates, we must be able to produce a reasonable amount of Molibdenum 99, but the reactivity and power limits must be taken into account in every case.

For this reasons, several options were tested in order to reach the objective.

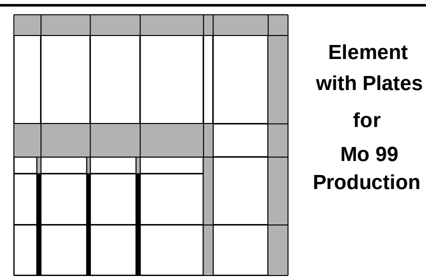
First, plates of uranium 20 % enriched were considered, keeping the same geometry of the 90% plates, but changing the uranium-aluminum densities. For the second possibility, rods were tested, and for the third one a new geometrical arrangement of Uranium - Aluminum plates with a Zircalloy clading was analized. All these analysis will be shown below.

MODELS AND CALCULATIONS

The calculations were performed using a two dimensional collision probability code, HUEMUL (Ref 1) and a three dimensional reactor difusion code, PUMA (Ref 2). Also thermohydraulic calculations were done to evaluate the behavior of the device from the point of view of the water cooling.

A] 20% Uranium-Aluminum plates

The first case analized was to consider plates made of 20% Uranium with the same geometry as the 90 % Uranium plates as shown in the figure at the right side, but with a higher density. In this case, the Uranium 235 mass per plate is 1.9 grams instead of the 1.05 grams in the 90% enrichment case.



Calculation Lattice used in HUEMUL

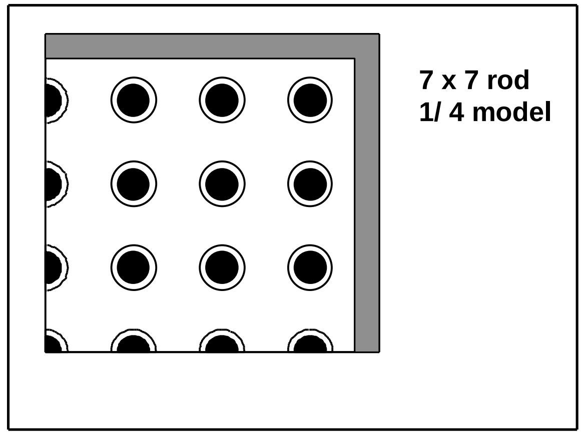
The cell calculations were performed using the HUEMUL model shown in the figure. Results obtained with PUMA and other thermohydraulic evaluations showed that the production obtained after a week of irradiation is about 3400 Ci, but the reactivity value (1842 pcm) of the rod is too high, and the power is beyond the safety limits.

For this reasons this device is not appropriate for the objective of this work.

B] Uranium Aluminum rods

The case of uranium-aluminum rods was also considered. These rodes were disposed as can be seen in the HUEMUL two dimensional model shown in the figure below. Arrangements of 7 by 7, 8 by 8,and 9 by 9 rods were analized. Reactivity values obtained for all these cases were very low. Thus, from the safety point of view these options are acceptable.

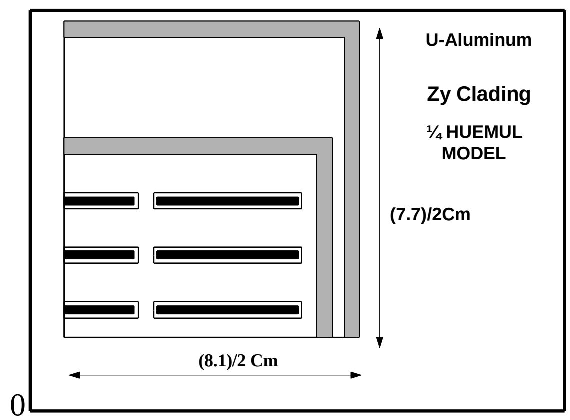
Also its efficiency is satisfactory. In an arrange of 7 by 7 rods (figure at the right side) the power and Molibdenum production (considering that the rods are about a 30% longer than the plates) are reasonable (About 2500 Ci).



However, the amount of Aluminium introduces serious problems to the chemmical process when performing the separation of Molibdenum.

C] 20% U-Al dispersion with Zircalloy clading

Another device was analized, where the Zircalloy clading used at the Embalse Nuclear Power Plant is deformed to serve as clading of Uranium - Aluminium plates as shown in the figure at the right side. Calculations with HUEMUL and PUMA were done changing the thickness of the plates. Results obtained for this case can be seen in the following table:



Uranium density = 1.74 g/cm^3

Thickness(mm)	Dr (pcm)	Activity (Ci)
0.7	828	2968
0.8	905	3252
0.9	979	3488
1.0	1049	3649
1.1	1109	3865

Both reactiviy and power values are reasonable for thicknesses of 0.7 mm and 0.8 mm. Feasibility will be analized later.

THERMOHYDRAULIC CONSIDERATIONS

A thermohydraulic analysis was performed in order to assure the irradiation plates and rods cooling.

For the rods, because of the low heat flux, no cooling problems were found.

For the 20% Uranium - Aluminium plates and the 20% Uranium - Aluminium pellets in Zircalloy flat clading configurations the maximum heat flux results in 130 w/cm^2 and the cooling velocity required was quite high.

To achieve this velocity, with the reactor in cooling conditions, it was necessary to reduce the flow area using an internal Aluminium box as the flow channel.

FEASIBILITY OF THE PROPOSED DESIGN

Hereafter we give description of the changes that have to be done in the actual facilities and techniques to get the separation of Molibdenum from the targets proposed.

- Fabrication of the assembly

a) Clading

Pressed pellets in a Zircalloy clad are used for avoiding the dissolution of the Aluminium clading and the use of Aluminium as plasticizer for the colamination.

Deformed CANDU type cladings can be used (internal radius 0.608 cm, external radius 0.656 cm). This clad is rolled in a special device designed in the PPFAE (Pilot Plant of Special Alloys) that performs the deformation of the clad with constant circunference (reference 3). In this way, clading with rectangular profile as described in the neutronic calculation can be obtained with an inner width between 0.8 and 1.2 mm. Each clad is 400 mm long.

b) Meat

Due to the use of a basic dissolution method for the separation of the Molibdenum, an aluminide alloy must be used because of its good behaviour in this medium. The obtention of aluminide powder is described reference [4], and, depending on the composition obtained, Aluminium is added to get a composition of 44 per cent in weight as described in the neutronic calculations. With this mixed powder already formed (as low as possible in free Aluminium) pellets are pressed in an hydraulic-mecanic press (Komage K-30 Tn) with a matrix of variable rectangular profile.

c) Welding and Assembling

The clads are welded after closing with a plug end cutted from a 1 mm sheet and then divided into three bars separated by two 1 mm long sheets that are pressed by a press, assembled in a rectangular 78 by 78 by 400 mm irradiation box ready to be introduced in the reactor at the irradiation place. The structure is designed in a way that the long bars can easily be disassembled.

- Irradiation

Normal procedure

- Disassembling of the bars.

The extraction of the long bars is made in a cell near the top of the reactor with a manipulator. They are cutted with a guillotine in three isolated parts previously separated by the pressed sheet. This individual 120 mm short bars are introduced into a 40 mm diameter by 140 mm height case and dropped to a moving pig that brings the case to the Separation Plant.

- Cutting and extraction of the compacts.

In the hot cell, the short bars are cutted by their ends by means of a circular saw and the pellets are extracted pushing with a 1 mm thick sheet.

- Dissolution and Separation

Normal procedure.

- Irradiation of a prototype

Due to the possibility of welding of the pellets against the clad, an irradiation experiment is suggested. Still in the case of some degree of welding of the pellets to the clad, it will be soft, due to the fact that pellets are green pellets, and the powder particles are not thightly bonded together. The particles welded will be easily broken by the extraction sheet. We estimate that the green pellets will disolve faster than the colaminate sheets.

CONCLUSIONS

Three possibilities were analized in order to obtain Molibdenum 99 in the RA3 reactor, using 20 % enriched Uranium.

The first one, consisting in the same design used in the 90% enrichment case with a greater Uranium density produces results which are inacceptable from the viewpoint of safety and heat transfer.

For the second one, arrays of 7 by 7, 8 by 8 or 9 by 9 rods, reactivity and power are within the safety limits and the Molibdenum production is appropriate, but the amount of Aluminium introduces serious problems in the chemmical process to obtain the Molibdenum.

Finally, Uranium – Aluminide pellets inside a Zircalloy flat clading have been considered as being the best solution to the problem. For this case, the activity, safety, dissolution and low Aluminium use in the dissolution requirements are accomplished. The Zircalloy flat cladings were developped and the additional tecniques and equipment needed for the practical realization of this solution seems to be reasonably feasible.

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