PROGRESS IN TECHNOLOGY DEVELOPMENT FOR ^{99}Mo ISOTOPE PRODUCTION

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ABSTRACT

To obtain new revenue streams for the nuclear research reactor and desiring to serve the nation's needs by providing radioisotopes to the Romanian market, the Institute for Nuclear Research at Pitesti, Romania decided to implement the technology for ^{99}Mo isotope production using LEU metal foil target and the modified Cintichem process. To accomplish this task, Romania is participating in IAEA Co-ordinated Research Project (T.1.20.18) "Developing techniques for small scale indigenous Molybdenum 99 production using Low Enriched Uranium fission or neutron activation".

The paper will present activities and the results obtained in the first year by the Institute for Nuclear Research. Activities were focused on: performing calculation for modeling of the target and irradiation device; designing target, irradiation device, and adequate equipment or tools for processing; fabrication of certain devices; design modifications of an existing radiochemical cell in order to accommodate entire process; developing a cooperation and partnering with other institutions to align with other researches that are proceeding for dispensing the product.

1. Introduction

Nuclear medicine is relying on ^{99m}Tc and its parent ^{99}Mo whose major source of production is from fission of ^{235}U. The Uranium and other adequate materials are used to prepare suitable targets. These are irradiated in research or test reactors and after that are dissolved for ^{99}Mo recovering from the solution.

To reduce nuclear proliferation concern, in the frame of US RERTR program, a lot of work was done to identify changes of target design and chemical processing determined by LEU utilization.

In the frame of CRP "Developing techniques for small scale indigenous Molybdenum 99 production using Low Enriched Uranium fission or neutron activation" IAEA sustains collaboration and facilitates the transfer of knowledge in the field of the so called Cintichem modified process which is now a freeware technology for target fabrication and chemical processing. Romania decided to participate to this CRP together with other interested countries in developing technology for fission Molybdenum production and was invited to make part of this CRP.

The main reasons taken into consideration to sustain such a decision were:

- the ability to manufacture target, using imported enriched Uranium metalic foils;
- the ability of the existing TRIGA 14 MW reactor to irradiate target;
- the ability of the existing hot cell facility to assure handling, disassembling and chemical processing of the target, taking into consideration some modifications;
- the ability of Institute for Nuclear Research to handle, treat and condition radioactive waste;

- the existence in Romania, of the Center for Radioisotopes Production able to assure the dispensing of the product.

Target material and size

Modified Cintichem target is not based on UO_2 , but is made of a piece of uranium foil covered with nickel foil, held and compressed between two aluminum cylinders having welded ends. The annulus type LEU target assembly consists of:

-two aluminum cylinders having 18 cm length and diameters of 27.99 mm and 29.77 mm;
-one piece of uranium foil 7 cm x 5 cm x 0.014 cm;
-coating Ni foil of 0.0015 cm thickness.

Target irradiation

The target will be mounted into the irradiation device and loaded in TRIGA reactor irradiation location. The maximum un-perturbed neutron thermal flux in LEU core is $2.5x10^{14}$ n/cm^2 .s

Currently, TRIGA 14 MW reactor is not in operation but in process of modernization of certain reactor systems. Commissioned in 1979 at the Pitesti Institute for Nuclear Researches, is a tank type reactor, initially fuelled with high enriched uranium elements and beryllium reflector elements in a rectangular lattice. Partial neutron moderation and core cooling are provided through forced convection of de-mineralized water. HEU-LEU conversion was terminated on May 2006. All reactor systems are in operational status with the exception of the cooling towers of the secondary circuit whose upgrading will be terminated at the end of this year. The upgrading of the other reactor systems (ventilation system, radiological surveillance system, reactor console) will be done next year, so that to have a minimum impact on reactor operation.

Target transfer to the Hot Cell Facility

In the vicinity of the reactor building there is the Hot Cell building containing two big cells (receiving cell and examination cell) and two small cell (radiochemical cell and metallographic cell). A water filled channel provides a communication way between reactor tank and Hot Cell, facilitating underwater transfer of irradiated samples from core region directly into the receiving hot cell. Here, irradiated target will be disassembled for chemical processing.

Isotope extraction at the Hot Cell Facility

Uranium foil recovered from the target will be dissolved in a stainless steel dissolver using nitric acid. A fission gas recovery system, provided with cryogenic traps will permit to recover radioactive iodine and xenon. The amount and the specific activity of ^{99}Mo obtained, depend on the target loading, target power and irradiation time.

Radiochemical cell modification

Initially, chemical processing will be developed in examination cell, but routine production activities will be moved in radiochemical cell. It can accept only a maximum gamma activity (1 MeV energy) of 1000 Ci. More shielding, relocation of a pneumatic transfer system and processing equipment installation, unique to isotope production, have to be considered in order to produce ^{99}Mo.

Radioactive waste treatment

The waste will be generated as solids, liquids and gases and will include material in the low and intermediate levels radioactive categories. Initial treatment of waste will be required at the production site prior to transfer to local facility final treatment. The treatment and elimination of gaseous waste is solved by an efficient off gas system. Xenon and Iodine isotopes are trapped on special materials for decay before elimination in atmosphere.

Volume reduction of solid waste is carried out through cutting and subsequently, they are conditioned in special containers.

Liquids containing enriched Uranium will be stored till a decision concerning recycling for Uranium recovery will be taken. Other waste solutions, containing fission products and actinides will be mixed and adjusted to neutral pH and immobilized with cement.

2. Calculations

2.1 Neutronic modeling of the target and irradiation device for small scale fission Molybdenum production.

TRIGA neutron calculation system is based on the following codes and libraries;

-Transport code cell –WIMS; it is an ANL version permitting to obtain microscopic cross section of the nuclides for material, collapsed on 7 energy groups for different burn up steps. Its 69 groups library was supplemented with other useful nuclides from ENDF/B-V.

-3-D diffusion code supplemented with a burn up loop – DFA; this code was developed in our Institute from 3 DDT diffusion code being used for neutron flux distribution calculation and for fuel burn up calculation of the reactor core. It is also used to calculate the power and reactivity induced by an experiment in TRIGA core. Microscopic cross sections of the nuclides are prepared with the WIMS code.

TRIGA fuel assembly is made up of 25 rods arranged in a square net. TRIGA core, Beryllium reflectors and water are modeled by using 73x63x27 meshes. Each fuel rod is described by an x-y mesh (1,633 cm). On z direction there are 13 meshes. Neutron flux and power are calculated in 13 points for each of the 725 fuel rods.

Our current core configuration contains 100% LEU fuel. Based on the WIMS - DFA aforementioned codes and using the irradiation device model from the fig. 1 and fig. 2 have been possible to calculate linear power and reactivity change associated with Molybdenum target inside of the irradiation device introduced in the G7 core location (fig.3). This is the most powerful irradiation position in our reactor. Cylindrical symmetry of the irradiation device determines a correct WIMS modeling and therefore averaged cross section obtained are reliable. The main results obtained for a reactor power of 10 MW, 5 cm as Uranium foil height, 7 cm length, 0.014 cm thickness and uranium mass of 9.29 grams are:

P_{target}=7200 W/target; $\phi_{thermal}$= $1.6x10^{14}$n/cm^2.s (E<1.125 eV)

Positive reactivity induced by irradiation device and target is 0.43$ and this is determined mainly by Aluminum water removal and much less by Uranium foil.

Fig.1 Cross section through the irradiation device containing the target

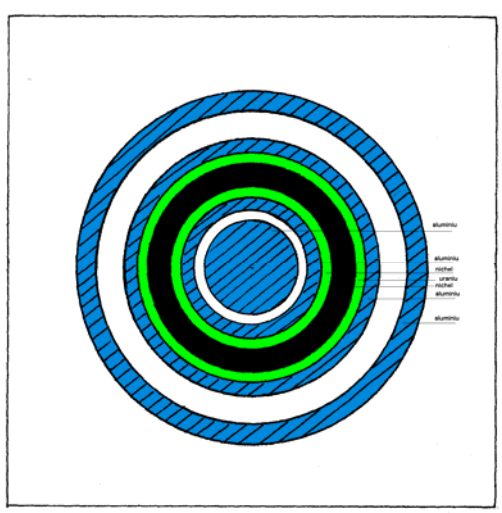


Fig.2 Vertical section through the irradiation device and target (not to scale)
(values in parenthesis are corresponding values obtained after using a sizing draw plug)

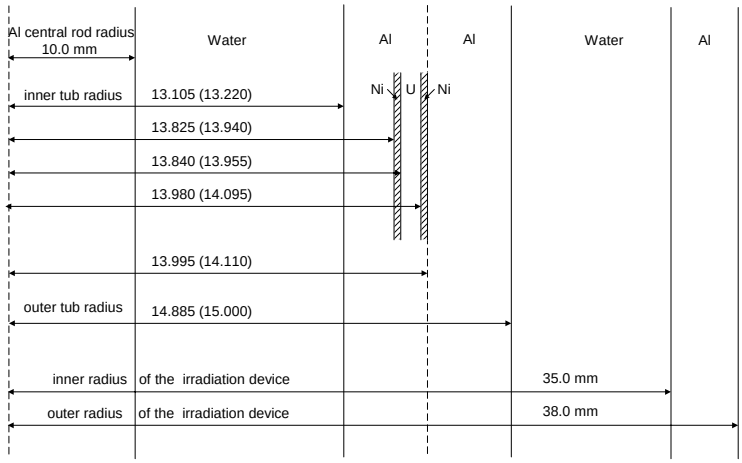
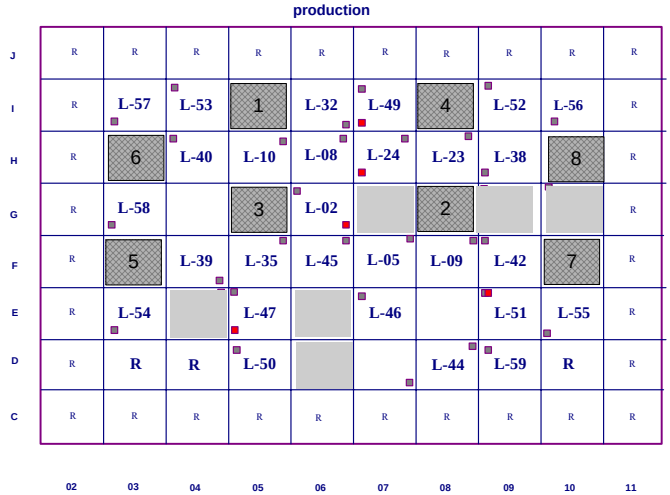


FIG: 3 TRIGA core configuration; different irradiation location of U-target for Mo-99 production



1 = Control rod L-xx = LEU fuel = Irradiation location

2.2 Thermal hydraulic assessment of the target and irradiation device.

Heat transfer computations are based on the following assumptions:

1. The full power developed by the Uranium target is 9.0 KW;
2. Uranium foil covered with Ni foil is tight pressed between the two aluminum cylinders and is considered with no gaps;
3. Heat generated in metallic uranium foil is propagating through conduction in nickel foils and aluminum tubes and through forced convection to cooling agent circulating in inner aluminum tube and around outer aluminum tube;

4. The flow through irradiation device containing target is equivalent with that through a TRIGA fuel bundle.

RELAP5 is the transient analysis code used for thermal hydraulic design of the fission molybdenum target.

The light water reactor (LWR) transient analysis code, RELAP5, was developed at the Idaho National Engineering Laboratory (INEL). Code uses include analyses required to support licensing audit calculations, evaluation of accident mitigation strategies, and experiment planning analysis. The RELAP5/MOD3 code is based on a non homogeneous and non equilibrium model for the two phase system that is solved by a fast, partially implicit numerical scheme to permit economical calculation of system transients.

A cross section of irradiation device containing the target situated in the middle of the square (side length = 88.9 mm) aluminum shroud is presented in fig.1. The water column delimited by the aluminum shroud is divided in three channels. The exterior channel limited by shroud wall and outer tube of irradiation device, medium channel limited by outer tube of irradiation device and outer tube of the target and interior channel limited by inner tube of the target and central rod of the irradiation device. Heat generated by Uranium foil is mainly evacuated in the medium channel and interior channel.

Outside diameter of irradiation device is 76 mm and central rod is 20 mm diameter. The hydraulic layout for RELAP5 code of irradiation device is presented in the fig. 4. RELAP5 geometric layout for irradiation device is presented in the Fig. 2.
Taking into consideration power source localization its adiacent volume is divided into 9 subvolumes acording to fig. 4. There are, also, a lateral cooling channel limited by shroud walls and external tube of irradiation device (105) These volumes are conected at the top and bottom parts with superior and inferior plenum of the cooling channel delimited by the shroud walls. The above model is equivalent with the cooling channel of the TRIGA fuel assembley and is coupling with two time depending volumes- a superior one (108) representing reactor pool and the inferior one (109) representing inlet duct of the primary circuit.

With this data and considering an output power generated during irradiation P_{target} = 9 kW, it is obtained a temperature distribution into irradiation target presented in the fig. 5. Maximum temperature in the Uraniu foil is aprox. 125 °C. The temperatures of outer and inner surfaces of the target are included in interval (117-121) °C. The thermal parameters data from the RELAP code output are presented in the Table 1.

Table 1 Mo 99 heat transfer characteristics for 9 kW

side	bdry.vol.	surface	heat-trf.	heat-flux	critical	CHF	heat-trf.	int.-heat	vol.ave.
	number	temp.	Conv.	Conv.	heat-flux	mul	coef.conv	source	temp.
		(K)	(Watt)	(Watt/m2)	(Watt/m2)		(Watt/m2-K)	(Watt)	(K)
left	101-	393.6	3775	9.09E+05	1.52E+07	1.68	17389	9000	394
right	104-	390.9	5225	1.11E+06	2.50E+07	2.28	14585		394

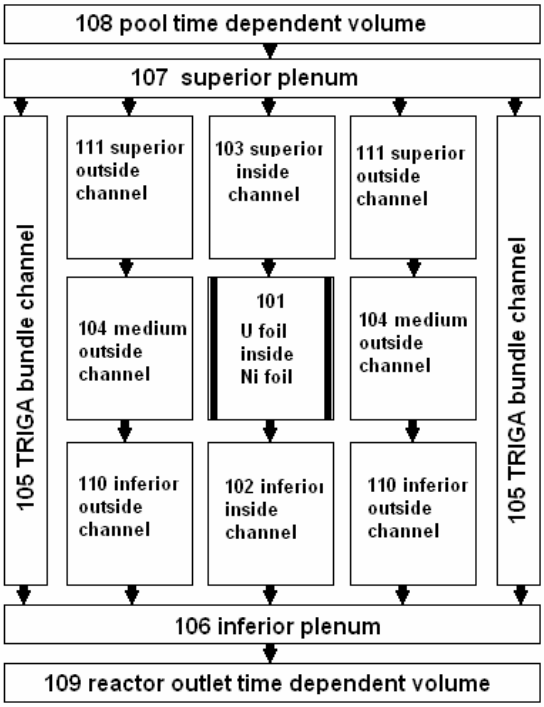


Fig. 4 RELAP hydraulic layout for irradiation device

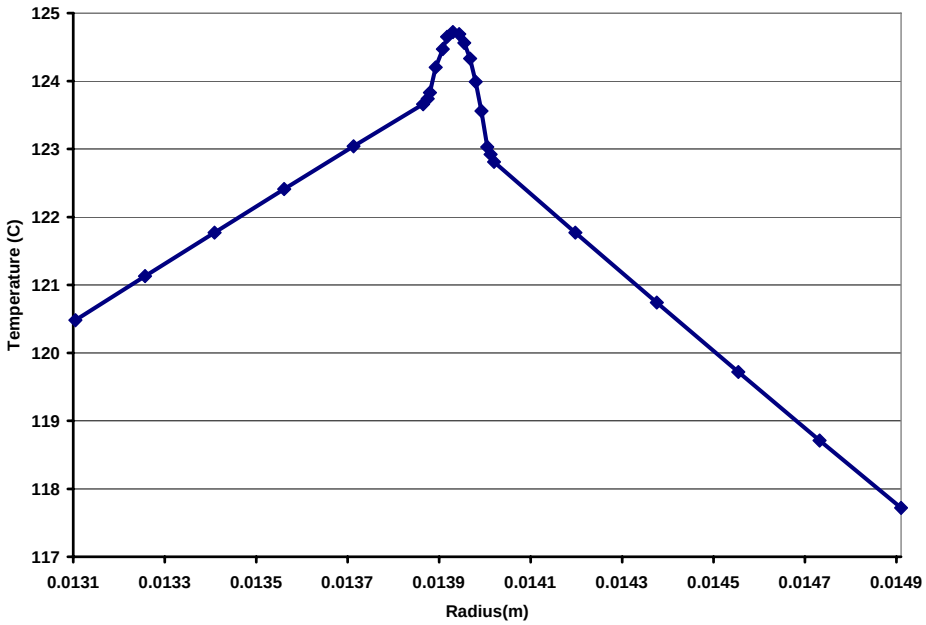


Fig. 5 Temperature distribution inside irradiation target (P_{target} = 9 kW)

2.3 Activities calculations

Calculation of fission products activities was done with ORIGEN –S code from the SCALE 5 system considering an irradiation at constant neutron flux ($\Phi=1.16*10^{13}$ n/cm^2.s) so that target power to be 7.2 kW.

ORIGEN –S code utilizes a Wescott neutron spectrum description in irradiation location. Therefore for calculation of THERM, RES and FAST it is used TRITON code from the SCALE 5 system. To carry out this task it is needed to work out a 2- D geometric super cell and adequate material description of the core. The result of the 2-D model is presented in the fig.6. The values obtained for the spectral index are: THERM =0.750, RES = 1.077 FAST = 0.597.

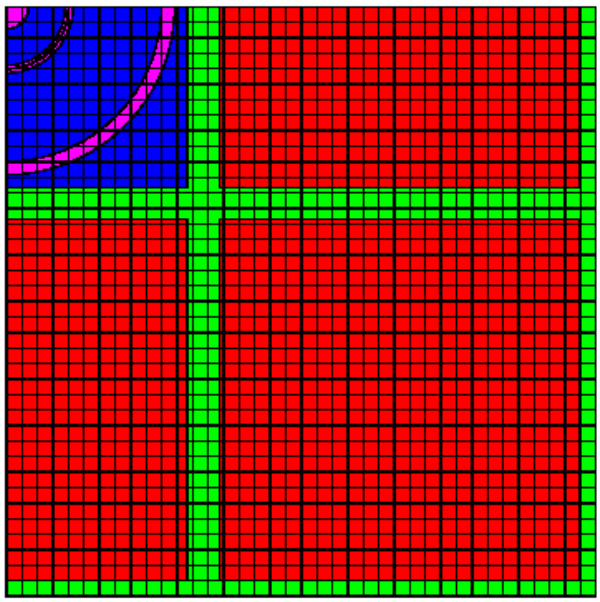


Fig. 6. 2- D geometric super cell description for TRITON code
(red: homogenized TRIGA fuel, purple: Al, blue: water, green: shrouds interface)

Maximum irradiation time was considered 10 days of a target containing 9.29g of Uranium, 20% enriched. Also, has been calculated level of activities of the target irradiated for 5 days after different cooling time (^{99}Mo and total activities). The results are given in table 2 and table 3.

Table 2 Molybdenum activity after different irradiation time (target containing 9.29 g of Uranium, 20% enriched)

Irrad. Time (days)	1	2	3	4	5	6	7	8	9	10
^{99}Mo (Ci)	84.74	150.3	200.7	239.6	269.4	292.1	309.5	322.5	332.3	339.5

Table 3 Activities after 5 days of irradiation and different cooling time (target containing 9.29 g of Uranium, 20% enriched).

Cooling time (days)	0	0.33	0.67	1	2	3	4	5	6	7
^{99}Mo (Ci)	269.4	247	227.7	209.4	162.7	126.4	98.2	76.3	59.3	46.1
Total (Ci)	30620	4420	3222	2580	1662	1263	1033	878.4	764.4	675.8

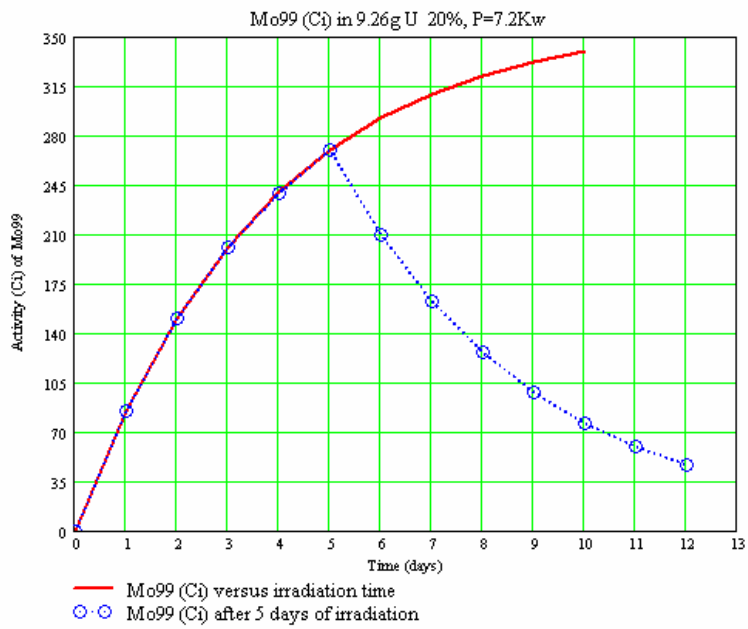


Fig.7 Activity evolution in the target (red color) during irradiation from 1to 10 days and after 5 days of irradiation followed by cooling (blue color)

2.4 Accident analysis

It is supposed a sudden loss of the main coolant loop flow. In 10 seconds flow rate vanishes and the reactor is shut down by scram (this type of accident is considered in TRIGA reactor safety analyses). In such an event temperature evolution in the fission Molybdenum target is presented in fig.8. The data in this analyses correspond to a target power of 9000W when the maximum Uranium foil temperature is 125 °C. Initially, foil temperature decreaseto 48°C in two seconds from the reactor scram. In the following 5 seconds the temperature increases to 57 °C due to the inversion of the flow rate during natural convection loop formation in reactor pool. In continuation the temperature decreases constantly without any danger of temperature increasing in the target.

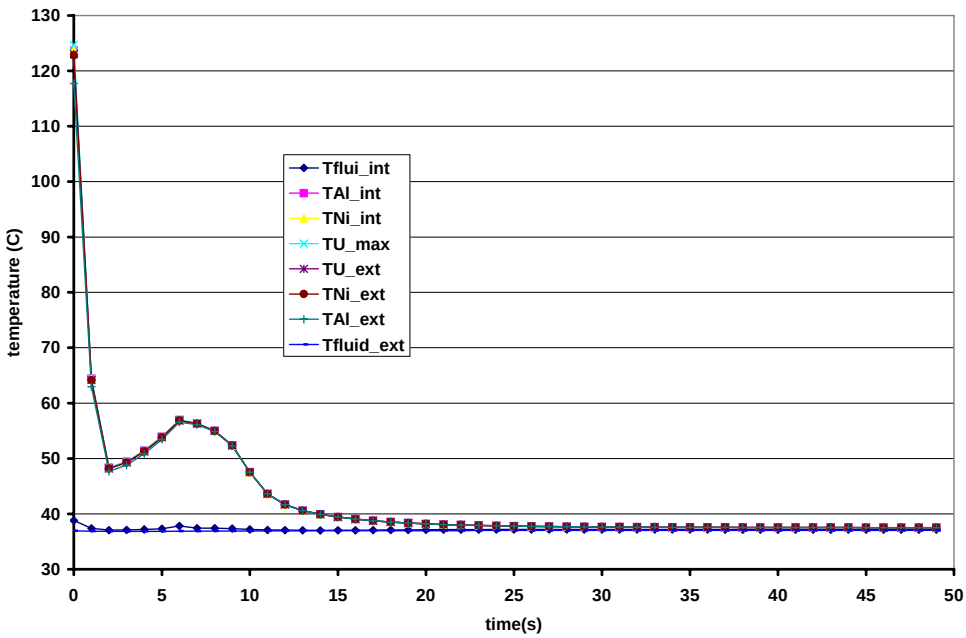


Fig.8 Temperature evolution in the target during loss of the main coolant loop flow

Insertion reactivity accident is another case taken into consideration. Assumption is made that the reactor is operated at 14 MW and a puls is produced in 0.1 second which increases the power by a factor of 5. The results obtained in this case are presented in fig.9. The Uranium foil temperature increases in 0.12 seconds from 125 °C to 188 °C and immediately decreases in marked manner.
The target do not suffer any damage.

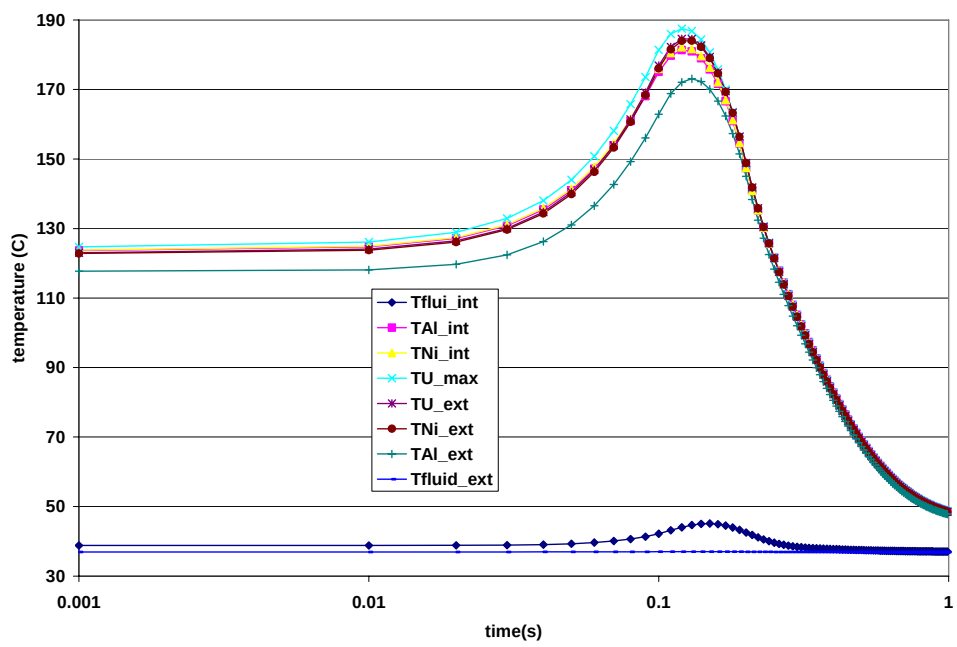


Fig.9 Temperature evolution in the target during reactivity insertion

3. Conclusions

We took into consideration a fission Molybdenum target conceived by ANL-USA maintaining the geometry (dimensions).

Neutronic calculation based on WIMS-DFA codes provided the following data (reactor power -10 MW, irradiation location –central one G7):

-Uranium foil geometry: cylinder (m = 9.29 grams, R= 14 mm, H= 50 mm)

- P_{target}=7200W/target; $\phi_{thermal}$= $1.6x10^{14}$n/cm^2.s (E<1.125 eV)
- Positive reactivity induced by irradiation device and target: 0.43$

Thermal hydraulic assessment was done with RELAP5 MOD 3.3 a transient analysis code used for designing.

The irradiation device containing the target is situated in the middle of the square (side length = 88.9 mm) aluminum shroud which is identical with the TRIGA fuel shroud. The water column delimited by the aluminum shroud is divided in three channels by the irradiation device. Outside diameter of irradiation device is 76 mm and central rod is 20 mm diameter.

With this data and considering an output power generated during irradiation P_{target} = 9 kW, it is obtained a maximum temperature in the Uraniu foil of aprox. 125 °C. The temperatures of outer and inner surfaces of the target are comprised in interval (117 °C-121 °C).

Calculation of fission products activities was done with ORIGEN –S code from the SCALE 5 system considering an irradiation at constant neutron flux (Φ=$1.16*10^{13}$ n/cm^2.s) so that target power to be 7.2 kW. For a uranium mass of 9.29 grams after 5 days of irradiation the ^{99}Mo activity is 270 Ci and total activity is 30620 Ci. After one day of cooling these activities change to 210 Ci and 2580 Ci, respectively.

For accident analyses we have taken into consideration a target power of 9 kW and two cases:

- loss of the main coolant loop flow;
- insertion reactivity.

In the first case maximum temperature (125°C for 9 kW) in the Uranium foil is not exceeded.

In the second case maximum temperature is increased for short time from 125°C to 188°C and immediately decreases in marked manner.

The target does not suffer any damage.

4.References

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