POTENTIAL INVESTIGATION OF ^{99}Mo PRODUCTION VIA UO_2SO_4 LIQUID TARGETS CONTAINING ^{NAT}U IN A 5 MW RESEARCH REACTOR

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Abstract

The most routine methods of ^{99}Mo production are either by ^{98}Mo(n,γ)^{99}Mo reaction or by ^{235}U(n,f)^{99}Mo fission process; the highest specific activity of ^{99}Mo among all production methods is achieved by the second process. However, recently some research centers have directed their attention toward liquid target application instead of the previously mentioned solid ones. Therefore, this work investigates the production potential of ^{99}Mo using uranyl sulphate liquid target irradiation in a 5 MW nuclear research reactor. Irradiation of four liquid targets involving natural uranium was theoretically investigated in the TRR irradiation boxes. The most optimize uranyl sulfate concentration was determined to fulfill convection condition of the TRR irradiation box cooling. The burn up calculations using the MCNPX 2.7.0 code showed about 5 Ci of ^{99}Mo is produced after 7-days irradiation at 4 MW power. Application of an enforced cooling inside the irradiation boxes could easily increase the radioisotope production rate to >100 Ci. Low cost of such liquid targets, extraction of gaseous fission products, short-time chemically separation of the proposed products and reusability of the irradiated liquid target are the main attractions of such method for ^{99}Mo production.

Keywords: ^{99}Mo production, ^{nat}Uranyl sulfate, MCNPX, Reactor irradiation

Introduction

The medical community has been weighed down by ^{99}Mo shortages due to aging of reactors such as the NRU (National Research Universal) reactor in Canada. There are currently no US producers of ^{99}Mo, and NRU is scheduled for shutdown in 2016, which means that another ^{99}Mo shortage is needed unless a potential domestic ^{99}Mo producer fills the void [1–2]. Currently solid target plates are used for the production of ^{99}Mo. The targets are generally either miniature Al-clad fuel plates or pins containing U–Al alloy or a thin film of UO_2 coated on the inside of a stainless steel tube [3]. In these targets, separation of ^{99}Mo from the other fission products produced inside the target after irradiation is started by the target dissolution either with alkali or acid material. However, aqueous homogenous reactors inspirit such imitation by modeling a liquid target instead of the pervious mentioned solid ones.

Hence, feasibility and economically study of such liquid target irradiation in research reactor in order to ^{99}Mo and possibly other radioisotopes production was proposed in this work.

Material and methods

In this work, MCNPX 2.6.0 has been used as a powerful particle transport code with the ability to calculate steady-state reaction rates, normalization parameters, neutronic parameters, as well as fuel burn up using CINDER90 to calculate the time-dependent parameters [4–5].

A cylindrical aluminum container involved uranyl sulfate solution was modeled using the MCNPX 2.6.0 code. A 3D neutronic model was set up using the MCNPX 2.6.0 code in cold zero power situations by means of ENDF/B-VI continuous-energy cross section. The cross sections of S(α,β) was used for the fuel solution and light water. KCODE card of the computational code was used for neutronic parameter calculations. 1500000 particle histories were transported to decrease the calculation errors to less than 2%. The modeled liquid target specifications are presented in Table 1.

Table 1 Liquid target material and dimensions modeled using MCNPX 2.6.0

sample specifications	value	unit
Fuel solution ($^{Nat}UO_2SO_4$): ^{235}U, ^{238}U, O, H, S	1.2-1.3	g/cm^3
Aluminum container: Al, Fe, Cu, Mn, Si	2.70	g/cm^3
Target dimension	0.55×8	cm
Al clad thickness	0.2	cm

Four modeled targets containing about 7.5 cm^3 of the liquid solution were positioned at the irradiation boxes of the 5 MW Tehran Research Reactor (Fig.1).

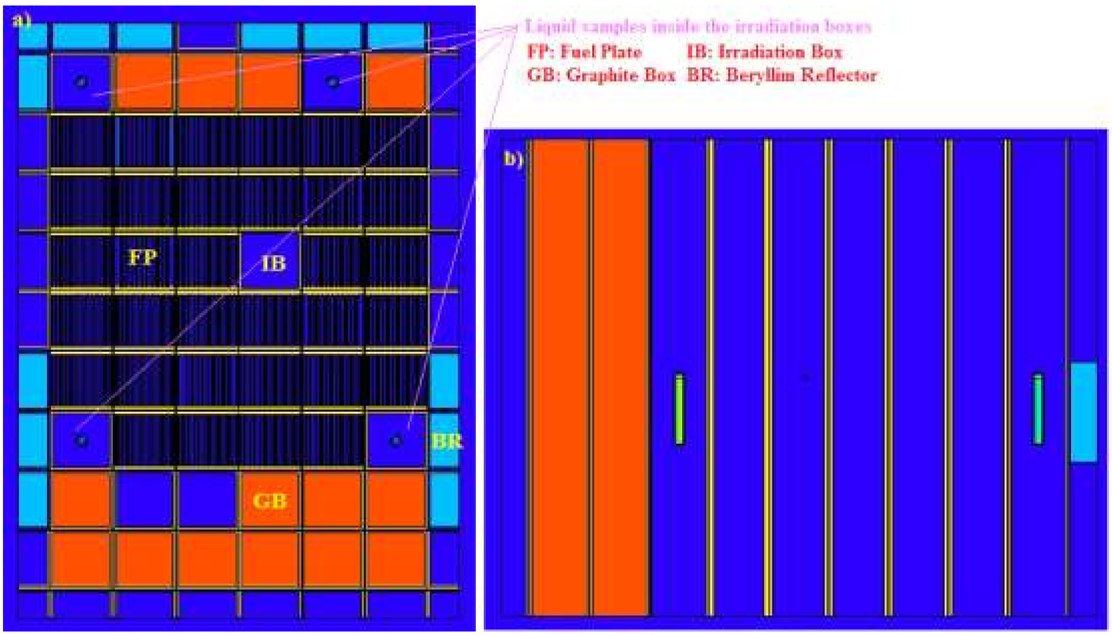

Fig. 1. Cross sectional view of TRR containing the liquid targets a) radial b) axial

Three types of sulfate solutions containing different concentrations of dissolved uranium were considered to be used as liquid solution to produce ^{99}Mo and other possible radioisotopes. Deposited power inside the modeled solution targets was calculated separately. Different concentrations of fissionable component of the liquid sample were investigated and an optimum concentration was suggested. Radial and axial deposited power was determined inside an optimum selected liquid sample using mesh tally card of the used code. ^{99}Mo, ^{131}I and ^{133}Xe production yield after 7-days irradiation of the different liquid samples comprised the optimum concentration of the fissionable isotopes were investigated. BURN card of the computational code was used to calculate fission process inside the irradiated samples. The calculations were repeated for a shell-type cylindrical geometry involves 227 cm^{3}of the liquid sample.

Results and discussion

Considering natural convection of the irradiation boxes, maximum bearable heat should be kept less than 50 W to avoid boiling of the liquid target during the 7-days irradiation. According to Table 2, a concentration of 180 g/lit will result in maximum deposited power <40 W.

Table 2 Deposited heat inside the liquid targets, the calculation errors <2%

Channel No	Uraniumconcentration: 30 g/l	Uraniumconcentration: 90 g/r	Uraniumconcentration: 180 g/l
		Depositedpower (W)	
1	8.87	17.18	32.99
2	6.25	12.28	24.11
3	10.31	19.77	38.34
4	9.89	19.27	35.41

Axial calculated deposited heat inside the cylindrical target with 7.5 cm^3 uranyl sulfate showed maximum value is <1 W/cm^3 (Fig.2a), while radial sectioning of the sample with height of 8 cm shows the value is <3 W/cm^3 (Fig.2b).

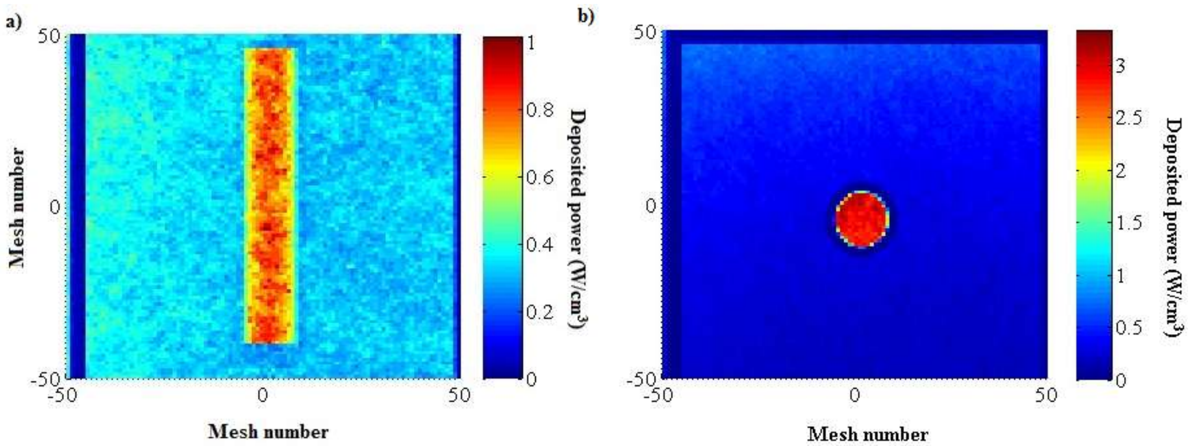

Fig. 2. Deposited heat inside the liquid target a)axial b)radial

Burn-up calculations showed about 1 Ci of ^{99}Mo is produced inside the 7.5 cm^3 liquid samples involved natural uranium (Table 3).

Table 3 Activity of different produced radioisotopes in 7.5 cm^3 targets after 7-days irradiation, Uranium concentration: 180 g/l

Channel No	^{99}Mo (Ci)	^{131}I (Ci)	^{133}Xe (Ci)
1	1.306	0.328	0.897
2	0.873	0.218	0.600
3	1.418	0.357	0.976
4	1.337	0.336	0.920

Total produced ^{99}Mo at the EOC is 4.93 Ci. About 1 Ci of ^{131}I and 3 Ci of ^{133}Xe is produced at the EOC.

By geometry optimization and an enforced cooling with 0.5 m/s flow in the irradiation boxes, 1000 W deposited power is easily removed during the sample irradiation. The calculations showed about 90 Ci of ^{99}Mo is produced inside the liquid irradiated sample at EOC.About 62 Ci of ^{131}I and 22 Ci of ^{133}Xe is produced too (Table 4).

Table 4 Activity of different produced radioisotopes in 227 cm^3 targets after 7-days irradiation, Uranium concentration: 180 g/l

Channel No	^{99}Mo (Ci)	^{131}I (Ci)	^{133}Xe (Ci)
1	23.93	6.02	16.45
2	17.48	4.39	12.00
3	26.56	6.7	18.27
4	24.57	6.19	16.89

Axial calculated deposited heat inside the cylindrical target with 227 cm^3volume showed maximum value is <2 W/cm^3 (Fig.2a), while radial sectioning of the sample with height of 14 cm shows the value is <3 W/cm^3 (Fig.2b).

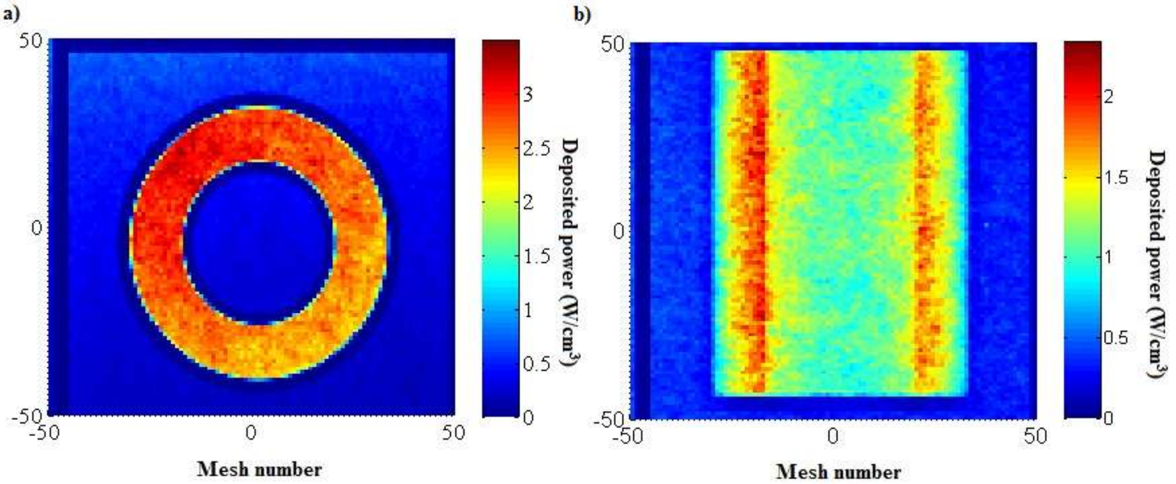

Fig. 3. Deposited heat inside the sell-type liquid target a) radial b) axial

Thermal-hydraulic calculations showed the investigated target maximum temperature is less than 70 ˚C by application of 0.13 m/s flow rate.

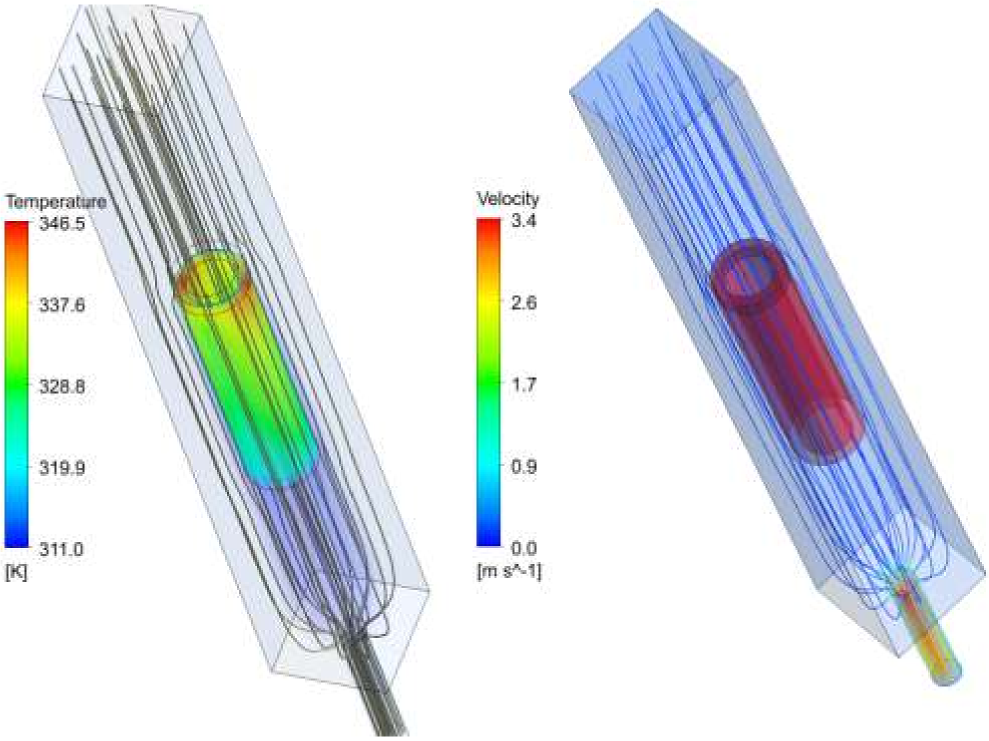

Fig. 4. Temperature profile along the liquid target length

Conclusion

The manuscript findings show that application of the liquid target with an optimized geometry opens an attractive method for simultaneously production of ^{99}Mo because of potentially production of several radioisotopes along with, minimization of radio-biologically hazardous nuclear effluent, reusable potential of the liquid target and its low cost.

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