

# NEUTRONIC AND THERMAL HYDRAULIC ANALYSES OF IRRADIATED FUEL PLATES FOR MOLYBDENUM-99 PRODUCTION

Daeseong Jo\*, Hong-Chul Kim, Kyung-Hoon Lee, Jonghark Park, Heetaek Chae

*Korea Atomic Energy Research Institute, 1045 Daedeok-daero, Dukjin-Dong, Yuseong-gu, Daejeon, 305-353, Republic of Korea*

*\*Corresponding author: djo@kaeri.re.kr*

## Abstract

Neutronic and thermal hydraulic analyses of the LEU targets irradiated in a research reactor for  $^{99}\text{Mo}$  production are performed to investigate (1) the heat production during irradiation, (2) the decay heat after irradiation, and (3) the cooling capacity under various cooling conditions. The heat production on the targets during irradiation is evaluated by MCNP. The decay heat after irradiation is estimated using ORIGEN-APR and compared against ANSI/ANS-5.1-1979. The cooling capacities of forced convection during irradiation and natural convection after irradiation are evaluated over time. The coolant and cladding wall temperatures and thermal margins i.e., minimum Onset of Nucleate Boiling (ONB) temperature margin and minimum Critical Heat Flux (CHF) ratio are evaluated with a consideration of the total power peaking factor and engineering hot channel factors. While the targets are under the water, the cladding wall temperature remains below the ONB temperature.

## 1. Introduction

Molybdenum-99 ( $^{99}\text{Mo}$ ) for nuclear medicine is produced by either the transmutation of  $^{98}\text{Mo}$  by the absorption of a neutron or the fission of uranium-235 ( $^{235}\text{U}$ ) by the absorption of a neutron. To produce a large amount of  $^{99}\text{Mo}$ , the fission of  $^{235}\text{U}$  is the most effective. Neutrons as sources are usually provided from nuclear reactors operating at powers in the range of megawatts [1]. The production rate of  $^{99}\text{Mo}$  depends on the thermal neutron fission cross section for  $^{235}\text{U}$ , thermal neutron flux on the target, mass of  $^{235}\text{U}$  in the target, and the half-life of  $^{99}\text{Mo}$ . To achieve the maximum production rate,  $^{235}\text{U}$  fission targets are usually irradiated for 5-7 days in the reactor core with a typical thermal neutron fluxes on the order of  $1 \times 10^{14}$  neutrons/cm<sup>2</sup>/s [2]. Since heat released from fission is much larger than that from activation, the fission targets require a proper design to effectively remove the heat production during and after irradiation. During the fission targets are irradiated in the reactor, heat is removed by forced convective cooling. After irradiation, the targets need to be transferred for further  $^{99}\text{Mo}$  production processes, i.e., dissolving targets and separating unnecessary fission products. During target transportation from a reactor to a hot cell, the cooling of the targets may still be required to remove the decay heat. Because the targets are transported in the water, natural convective cooling is still available. Natural convection cooling is a promising heat removal technique in research reactors [3]. To prevent cladding melting or blistering, the target plate made of an aluminum alloy should be below a blister threshold of 400 °C, and a solidus temperature of 550 °C [4,5]

In the present work, neutronic and thermal hydraulic analyses of the LEU targets irradiated in a research reactor for  $^{99}\text{Mo}$  production are performed to investigate the heat production during irradiation, the decay heat after irradiation, and the cooling capacity under various cooling conditions. The heat production on the targets during irradiation in the reactor core and decay heat after irradiation is evaluated using MCNP and ORIGEN codes, respectively [6,7]. The cooling capacity of forced convection during irradiation in the core, and the cooling capacity of natural

convection after irradiation are evaluated over time. The coolant and cladding wall temperatures and thermal margins are estimated with a total power peaking factor and engineering hot channel factors. While the targets are under the water, the cladding wall temperature remains under the ONB temperature.

## 2. Fission targets

The Fission Mo target shown in Figure 1 contains eight LEU plates. The target plates are made of Al6061 cladding and loaded 4.05 gU/cc  $U_2Si_2$  fuel meat. The plates are 1.57 mm thick, 50.0 mm wide, and 200 mm long, and placed 2.58 mm apart from each other to generate cooling channels. The thermal conductivity of the cladding is 120 W/m/K, and the thermal conductivity of the fuel meat is 54 W/m/K. The width and thickness of the coolant channel are 44.6 mm and 2.58 mm, respectively. The width and length of the fuel meat are 40.0 mm and 182.0 mm, respectively. The seven inner channels are heated from both sides, and the two outer channels are heated from one side. The rectangular target holder is loaded into different irradiation sites (IR1, IR3, IR4, IR7, IR8, IR10) where thermal neutron flux is approx.  $1 \times 10^{14}$  neutrons/cm<sup>2</sup>/s

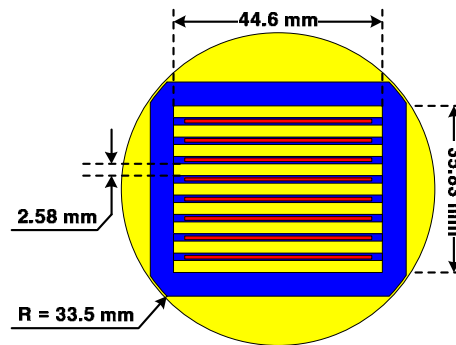


Figure 1 Fission Mo target loaded in the irradiation hole

## 3. Neutronic analysis

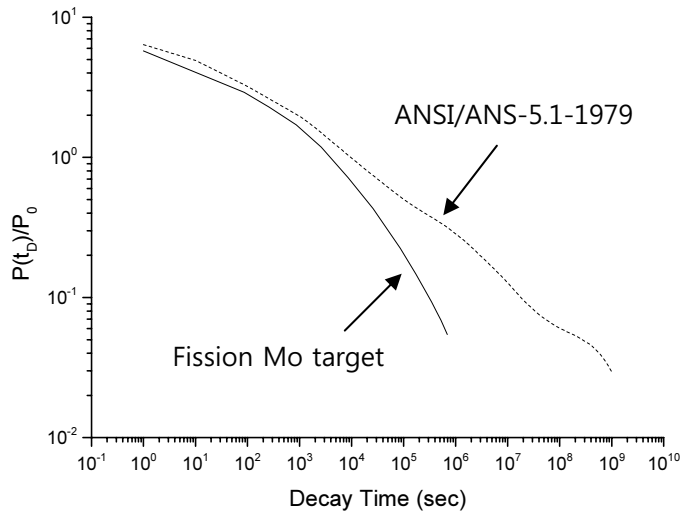
The fission Mo targets are irradiated in a research reactor with a thermal neutron flux of  $1 \times 10^{14}$  neutrons/cm<sup>2</sup>/s for 7 days to achieve the maximum production rate of the Mo<sup>99</sup>. To estimate the amount of heat released from the fuel during and after irradiation, MCNP, which is a Monte Carlo neutron-photon transport simulation code, is used. The material composition of  $U_3Si_2$  used for the analysis has 0.1409 wt% of 235U and 0.5726 wt% of 238U. The total weight fraction of U is 0.7135. Since the heat released during irradiation is dependent on the irradiation site in the core, 6 irradiation sites are tested. Table 2 lists the fission power of the Mo targets at different core statuses (i.e., BOC, MOC, and EOC) during irradiation. Plates 1 and 8 are the outmost plates, and release the more power than the inner plates. The highest power of 12.20 kW is released from the plate 8 in IR10 at MOC, and the highest power of the targets as 88.86 kW is released from IR3 at MOC. To estimate the decayed heat by ORIGEN-ARP, the uranium weight and power density per plate are required. The uranium weight per plate can be calculated as

$$M_m(U) = w(U) \times \rho_m \times W_m t_m L_m = 0.7135(4.05 \text{ gU/cm}^3)(4.0 \times 0.09 \times 18.2) \text{ cm}^3 = 18.9332 \text{ gU}$$

Figure 2 shows the decay power fraction over time. Since the targets are irradiated for 7 days, the power decays much faster than the decay power curve provided by ANSI/ANS-5.1-1979 [8]. This is because ANSI/ANS-5.1-1979 curve assumes that the fuel loaded is burnt for several months in the core.

**Table 2 Fission power of Mo targets**

Core Status	Power (kW)	IR1	IR3	IR4	IR7	IR8	IR10
BOC	Plate 1	9.73	12.67	8.87	9.95	10.65	10.83
	Plate 2	8.84	11.55	8.21	9.44	10.13	10.19
	Plate 3	8.38	10.90	7.88	9.20	9.98	9.87
	Plate 4	8.09	10.56	7.76	9.17	10.03	9.84
	Plate 5	8.07	10.43	7.70	9.32	10.14	9.98
	Plate 6	8.12	10.41	7.78	9.68	10.43	10.24
	Plate 7	8.32	10.57	8.00	10.23	11.02	10.82
	Plate 8	8.91	11.13	8.51	11.25	12.05	11.87
	Sum	68.47	88.22	64.70	78.25	84.43	83.65
MOC	Plate 1	9.85	12.64	9.13	10.38	10.74	11.19
	Plate 2	8.99	11.63	8.44	9.83	10.15	10.57
	Plate 3	8.48	11.00	8.06	9.56	10.09	10.19
	Plate 4	8.25	10.58	7.91	9.54	10.01	10.09
	Plate 5	8.22	10.50	7.86	9.61	10.20	10.24
	Plate 6	8.27	10.54	7.94	9.92	10.51	10.53
	Plate 7	8.52	10.71	8.16	10.55	11.14	11.16
	Plate 8	9.10	11.26	8.74	11.59	12.11	12.20
	Sum	69.68	88.86	66.23	80.98	84.94	86.18
EOC	Plate 1	9.46	12.13	8.80	10.00	10.19	10.73
	Plate 2	8.70	11.07	8.10	9.45	9.65	10.02
	Plate 3	8.19	10.51	7.74	9.26	9.46	9.71
	Plate 4	8.00	10.15	7.58	9.18	9.53	9.71
	Plate 5	7.91	9.99	7.54	9.35	9.65	9.80
	Plate 6	7.95	10.01	7.63	9.66	9.99	10.10
	Plate 7	8.21	10.21	7.89	10.22	10.49	10.67
	Plate 8	8.70	10.75	8.39	11.20	11.53	11.61
	Sum	67.13	84.83	63.67	78.31	80.49	82.34

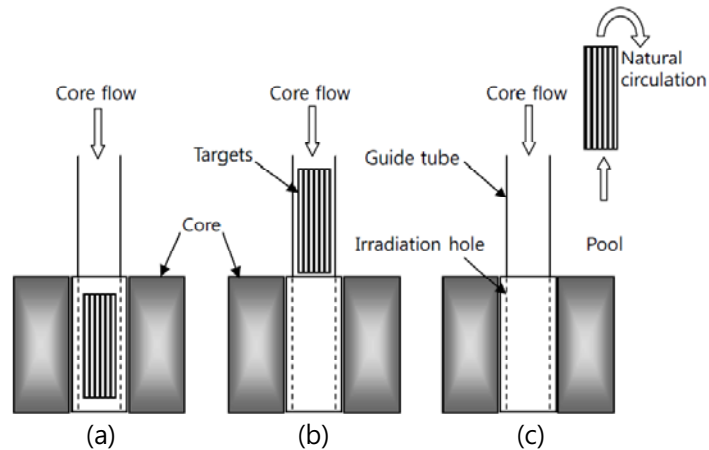


**Figure 2 Comparison of decay powers**

#### 4. Thermal hydraulic analyses

The fission Mo target requires different cooling conditions as shown in Figure 3. During irradiation, the fission Mo target is cooled by forced convection with downward core cooling. After irradiation, the target is cooled by natural circulation in a pool. Thermal margin analyses during irradiation and after irradiation are summarized in Tables 3 and 4. With the core flow velocity of 7.0 m/s, the minimum thermal margins are obtained with the power calculation at BOC. The ONB temperature

is determined by Bergles-Rohsenow correlation, and the CHF is determined by Kaminaga et al.'s correlation [9,10]. The minimum ONB temperature margin is 28.7 °C, and the minimum CHF ratio is 4.98. The thermal margin summarized in Table 4 shows that the target can be cooled by natural circulation in a pool after approx. 2.0 min from the withdrawal since the minimum ONB temperature margin becomes positive. This means that the Fission Mo target requires forced convection cooling from the withdrawal to 2.0 min.



**Figure 3 Locations of the targets: (a) during irradiation, (b) after irradiation cooled by forced convection, and (c) after irradiation cooled by natural convection**

**Table 3 Thermal margin analyses during irradiation (forced convection)**

	BOC	MOC	EOC
Channel velocity [m/s]	7.0	7.0	7.0
Avg. Heat flux [kW/m <sup>2</sup> ]	669.2	682.4	653.6
Power peaking factor, FQ	1.787	1.598	1.587
Axial peaking factor, FZ	1.185	1.037	1.048
Max. coolant temperature [°C]	42.4	42.6	42.2
Max. wall temperature [°C]	102.0	97.0	93.5
Max. fuel temperature [°C]	128.3	120.9	116.0
ONB temperature margin [°C]	28.7	33.8	37.2
Minimum DNBR [-]	4.98	5.45	5.74

**Table 4 Thermal margin analyses after irradiation (natural convection)**

Decayed time [hr]	Decay power fraction [%]	Channel flow velocity [m/s]	Max. wall temperature [°C]	Min. ONB temperature margin [°C]
0	6.280	0.091	136.4 (180.3)	-
0.01	3.490	0.068	99.7 (127.2)	-
0.024	2.912	0.062	91.4 (115.3)	-
0.03	2.820	0.061	90.1 (113.3)	1.9
0.05	2.475	0.057	85.0 (106.0)	9.3
0.072	2.285	0.054	82.1 (101.8)	13.4
0.5	1.355	0.042	67.3 (80.4)	34.7

## 5. Conclusions

Neutronic and thermal hydraulic analyses of the LEU targets irradiated in a research reactor for  $^{99}\text{Mo}$  production are performed to investigate (1) the heat production during irradiation, (2) the decay heat after irradiation, and (3) the cooling capacity under various cooling conditions. The heat production on the targets during irradiation is evaluated using MCNP. The decay heat after irradiation is estimated by ORIGEN-APR and compared against ANSI/ANS-5.1-1979. The cooling capacities of forced convection during irradiation and natural convection after irradiation are evaluated over time. The thermal margin analyses show that the minimum thermal margins resulted from BOC power distribution. The minimum ONB temperature margin is 28.7 °C, and the minimum CHF ratio is 4.98. After irradiation, the Fission Mo target is capable to be cooled by natural circulation after 2.0 min from the withdrawal. At 2.0 min from the withdrawal, the wall temperature is approx. 113 °C, which is lower than the ONB temperature of 115 °C.

## References

- [1] Ball, R.M., 1999. Characteristics of nuclear reactors used for the production of molybdenum-99. IAEA-TECDOC-1065, 5-17.
- [2] Talal, A.E., 2013. Thermal contact resistance and ambient temperature effects on the cooling of Mo99 plate targets inside the hot cell. *Annals of Nuclear Energy* 53, 476-484.
- [3] Jo, D., Park, S., Park, J., Chae, H., Lee, B., 2012. Cooling capacity of plate type research reactors during the natural convective cooling mode. *Progress in Nuclear Energy* 56, 37-42.
- [4] Olson, A.P., Kalimullah, M., A users guide to the PLTEMP/ANL V3.7 Code, RERTR program, Argonne National Laboratory, Argonne, Illinois, April 2009.
- [5] Hansen, M., Anderko, K., 1958. *Constitution of binary alloys*. McGraw-Hill, New York, USA
- [6] Gauld, I.C., Bowman, S.M., Horwedel, J.E., ORIGEN-APR: automatic rapid processing for spent fuel depletion, decay, and source term analysis. ORNL/TM-2005/39, Version 6 (Vol. I).
- [7] MCNP-A general Monte Carlo N-particle transport code, Version 5, LA-CP-03-0245, 2003.
- [8] American National Standard for decay heat power in light water reactors, ANSI/ANS-5.1-1979, 1979.
- [9] Bergles, A.E., Rohsenow, W.M., 1964. The determination of forced-convection surface-boiling heat transfer. *Trans. ASME Journal of Heat Transfer* 86, 365-372.
- [10] Kaminaga, M., Yamamoto, K., Sudo, Y., 1998. Improvement of critical heat flux correlation for research reactors using plate-type fuel. *Journal of Nuclear Science and Technology* 35, 943-951.