



Thermal Hydraulic Analyses of LEU Target Plates for Mo-99 Production

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ARTICLE INFO

ABSTRACT

Article history:

Received 18 March 2019

Received in revised form 8 May 2019

Accepted 10 June 2019

Available online 15 July 2019

Conversion from mastered High-Enriched Uranium to Low Enriched Uranium target for Molybdenum production constituted a challenge for nuclear engineering to evaluate its behavior during the three phases (irradiation, cooling and post irradiation). The present work is a contribution to thermal hydraulic analysis of Low Enriched Uranium target plate's behavior during irradiation using CFD model. Neutronic calculation, target properties and cooling parameters of Korea Atomic Energy Research Institute (KAERI) research group were used as an input in our model. The results obtained with CFD numerical model show that the irradiation conditions are below thermal margins and are approximately close to those obtained by using TMAP code. The developed CFD model will be extended to analyze the thermal hydraulic behavior of Low Enriched Uranium target plates at cooling period (natural convection) and at transfer period (in contact with air).

Keywords:

Irradiation; LEU target; Mo-99 production; Thermal-margins; CFD

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1. Introduction

Radioisotope Molybdenum-99 is produced mostly with a large amount and high specific activity around research reactors by thermal neutron induced fission of high and low enriched uranium targets using water-cooled irradiation and subsequent devices [1-4]. In accordance with the US Department of Energy's (DOE) Global Threat Reduction Initiative (GTRI) and 2009 National Research Council, the conversion from High-Enriched Uranium (HEU, ~ 93%wt U-235) to Low Enriched Uranium (LEU, ≤19.75% wt U-235) was a challenge for producers to novelty renewal of an alternative uranium target with yield and quality equivalent to those of the HEU [5-6]. Substituting LEU to HEU targets leads to irradiate five times more targets, generate five times more the volume of separation waste and generate fissile plutonium Pu-239 due to the neutron capture by the higher proportion of U-238 atoms. Consequently, this conversion may have repercussions moreover on nuclear reactor operation mode, nuclear reactor safety, existing irradiation and post irradiation facilities, production capacity, target configuration, target manufacturing, chemical processing, existing Mo-99 production facilities, and amount of solid and liquid radioactive wastes [7-10]. Therefore, LEU uranium target research and concept focused on fissile material compositions, densities and shapes (pellets, plates, pins and cylinders) underway to yield more efficiently the designs and to overcome drawbacks.

With regard to the development of target fissile materials and U-235 density, three types in monolithic and dispersion physical forms were considered in uranium alloys of (monolithic: U-Al, U-Mo, U-Be, U-Nb, U-Zr, U-ZrH_x and dispersion: U-Mo-Al), uranium ceramics of (monolithic: UO₂ and dispersion: U₃O₈-Al) and uranium intermetallic of (monolithic: U₃Si_x and dispersion: UAl_x-Al, U₃Si_x-Al) [11-13].

Monolithic fuels may offer higher physical densities and thermal conductivities, high melting point temperatures and stability. Dispersion fuels may avoid the swelling due to the fission gases and helium because of the presence of porosity, and obtain good compatibility with material cladding, achieve a uniform distribution of fissile material and enable to get a theoretical uranium density within 2.5 to 9.5 g·U/cm³. Currently new target design, called foil target, is being developed [14-17]. It consists of an LEU foil warped in thin recoil barrier material (Cu or Ni) and encased between two flat aluminum plates or cylinders. This new approach is economic; it reduces considerably radioactive wastes, and can match dispersion target yields. Dissolution technique for chemical processing after irradiation is among the main factors of choice of the appropriate chemical element to add to the LEU meat: alkaline or NaOH/NaNO₃ solutions are preferable due to high degree of uranium filtration as solid waste. Other chemical dissolutions (acid or electrolytic) investigated to dissolve other nuances of LEU material meat where the former solutions are not efficient [8] and [18].

Apart from the advantages mentioned above, these fuels present some concerns to be overcome and enhanced. Although the irradiation lasted five to seven days under thermal neutron flux of about 10¹⁴ n/cm².s, then the targets are allowed to cool for once a day and transferred to a hot cell for chemical processing.

LEU targets can lead under irradiation to an overheat due to thermal contact resistance between components at the interfaces (meat-warping, meat-cladding, warping-cladding), porosity formation, non-uniform diffusion zone, crack formation, bonding effect, swelling, thermal-mechanical deflections formation and stresses, anisotropic irradiation growth of uranium, growth of interaction layer between uranium meat and material cladding. These phenomena could lead to target failure under irradiation [19-23]. With that, one adds the difficulties of removal of LEU meat from warp or cladding materials during chemical processing.

The suitable removal of the nuclear fission heat generated in the targets during their irradiation, power decay during cooling time and transfer operation to the hot cell for processing are crucial. The purpose is to ensure that the cladding surface temperature remains below the onset nucleate boiling (ONB) margin and the critical heat flux (CHF), therefore, to ensure the safety of the reactor and hot cell, and avoid any possible failure, blistering or fusion of targets.

In this focus, the present paper gives a contribution on thermal hydraulic analysis of LEU plate type target for Molybdenum production under various operating conditions, for this purpose, a CFD numerical model using commercial CFD code ANSYS-Fluent is developed to predict the in-core targets behavior. The KAERI's LEU target properties, fuel bundle configuration, cooling parameters and neutronic calculation results are adopted as inputs in our developed model. The results obtained are compared with those obtained with TMAP code [24]. The validated model will be extended to analyze LEU cylindrical type targets.

2. Background

Annular and plate type targets for Molybdenum production are manufactured as stack of different material layers with non-perfect surface roughness (casting method, hot and cold rolling technique, co-extrusion method). Under irradiation, the presence of microscopic asperities and irregularities at interfaces lead to formation of gas cavities and solid contact points. At elevated

contact pressure, it has become a substantial thermal contact resistance and may disrupt the efficient heat conduction and lead to a hot spots formation. In addition, the temperature gradients between components (temperature drop at the interfaces) may induce stresses, due to the difference of thermal expansion coefficients, beyond the ultimate tensile strength and subsequent the fail of targets. According to the RERTR program of Argonne National LAB, related to the development of annular foil targets with 15 kW of power, thermal mechanical analysis and experimental results show that, under irradiation with a controlled surface heat transfer coefficient, meat remains bonded on inner and outer cladding in spite of compression and tensile stresses (radial and tangential) developed [14] and [25]. This design was validated too by successful irradiation of similar targets at BATAN reactor (Indonesia) and at PARR-1 reactor (Pakistan), no anomalies were reported on heat transfer of the performance of targets [22] and [26]. Similar conclusions on the performance of LEU flat plate were reported [15] and [27]. The physical integrity of three LEU superposed flat plate dispersion targets (effect of thermal contact resistance between targets and a stainless steel working table) under the effect of residual power just before chemical processing in hot cell were investigated using CFD model, the results show that the maximum temperature is below the aluminum cladding blistering temperature limit (673K) [28].

Nuclear Science and Technology Organization (ANSTO) program for developing Mo-99 production using LEU foil targets, the safety requirement and the compliance of irradiation rig and target designs with operations limits and conditions around HIFAR research reactor were verified [29]. Experimental and simulation for thermal hydraulic, using CFD models, of a two cylindrical LEU foil targets with a 15.3 kW of power and 17.4 kW mounted in a rig were done to predict streams/velocities of coolant and temperature distribution of targets. The calculation results show that the target can reach a 640.5 K of maximum temperature, the measurement, and the calculation of flow velocity of coolant values in irradiation rig of LEU foil targets are in accordance.

Recently, researchers at Korea Atomic Energy Research Institute (KAERI) developed a neutronic (using MCNP and ORIGEN-APR codes) and thermal hydraulic analysis (using TMAP code) of Mo-99 LEU plate fuel bundle inside isotope production nuclear reactor core, during cooling and the transfer to hot cell for chemical processing [24]. The results show that the LEU targets perform well during irradiation without compromising the reactor safety and the maximum cladding temperature after the decay of 24 h of time under blistering limit.

3. Thermal Hydraulic Analysis

3.1 Input Parameters and Calculation Conditions

As mentioned in the introduction of this paper, we consider the LEU targets configuration of KAERI research reactor, neutronic calculation results of the hot target and the cooling parameters [24] as applications to validate the CFD model developed. Table 1 gives material properties and dimensions of the LEU target. Table 2 gives nuclear powers generated in a hot channel at three reactor core statuses; Beginning Of Cycle (BOC), Middle Of Cycle (MOC), and End Of Cycle (EOC).

Table 1
 Materials properties and dimensions of LEU target

Item	Value
Cladding	
Material Al6061	
Density	2.70 g/cm ³
Thickness	1.58 mm
Width	50.0 mm
Thermal conductivity	120 W/m.K
Specific heat	896 J/kg.K
Meat	
Material U ₃ Si ₂ -Al	
Enrichment	19.75% U-235
Density	2.89 g.U/cm ³
Length	182.0 mm
Width	40.0 mm
Thickness	0.79 mm
Thermal conductivity	54 W/m.K
Specific heat	646 J/kg.K

During irradiation the fission heat generated by targets are removed by water-forced convection cooling with average velocity of 7.5 m/s and a pressure coolant of 1.8bar. The flow direction through targets is down warded and the inlet coolant temperature is of 308.5K. Target plates should maintain its integrity under irradiation. ONB temperature and critical heat flux were taken as thermal margins in this analysis. ONB temperature calculated using Bergles and Rohsenow correlation, Eq. (1). Sudo and Kaminaga correlation for rectangular channel used to determine critical heat flux for forced convection cooling, Eq. (2) and Dittus and Boelter correlation to calculate Nusselt number.

$$(T_{ONB} - T_{sat}) = \frac{5}{9} \left(\frac{q''}{p^{1.158}} \right)^{\frac{p^{0.0234}}{216}} \quad (1)$$

$$q_{CHF}^* = 0.005 |G^*|^{0.611} \left(1 + \frac{5.000}{|G^*|} \Delta T_{SUB,0}^* \right) \quad (2)$$

where T_{sat} [°C] is the saturation temperature, P [bar] is the pressure, q'' is the local heat flux [W/cm²], q_{CHF}^* is the dimensionless CHF, $\Delta T_{SUB,0}^*$ is the Dimensionless sub-cooling and G^* is the dimensionless mass flux.

For thermal hydraulic analysis, the highest power generated from the hot target plate at the three core statuses BOC, MOC and EOC corrected by power peaking factor and axial peaking factor are considered, see Table 2. No other conditions are applied for thermal margin evaluation as change of flow channel velocity, rise of temperature, over power.

Table 2
 Power of the hot target and correction power factors

	BOC	MOC	EOC
Power (kW)	14.17	13.59	12.69
Power peaking factor (FQ)	1.787	1.598	1.587
Axial peaking factor (FZ)	1.185	1.037	1.048

3.2 Numerical Modeling

The CFD (Computational Fluid Dynamic) method is used to simulate thermo-hydraulic behavior of LEU targets for the three reactor core statuses. This method made proof in several industrial fields and applications, its use in the nuclear domain is in the primary phases. It's a powerful tool to analyze physical phenomena at singular and localized spaces and consequently it can complete a coarse analysis generally carried out by using thermal hydraulic system codes as RELAP, TRACE, CATHARE, ATHLET or thermal hydraulic sub-channel codes as TMAP, MTR-PC. Designers in nuclear engineering, particularly in the domain of tests and irradiation of nuclear fuel, used specialized or in-house developed codes. These codes must be validated experimentally and adopted by safety authority. The current work is a contribution on LEU target thermal hydraulic calculation, in single phase flow case, using CFD ANSYS Fluent trading code [30].

The results obtained are compared by those of the TMAP/KAERI code [31]. The CFD codes solve the three transports Eq. (3), Eq. (4) and Eq. (5) (conservation of mass, momentum and energy, [30]) which govern the physical phenomena between various components of the studied domain; these equations were discrete with the finite volume method (FVM).

$$\frac{\partial \rho}{\partial t} + \nabla(\rho \vec{v}) = S_m \quad (3)$$

$$\rho \frac{D\vec{v}}{Dt} = -\nabla \vec{p} + \nabla(\vec{\tau}) + \rho g + \vec{F} \quad (4)$$

$$\nabla(\vec{v}(\rho E + p)) = \nabla(k_{eff} \nabla T - \sum_j h_j \vec{J}_j + (\vec{\tau}_{eff} \vec{v})) + S_h \quad (5)$$

where S_m : is the mass added to the continuous phase from the dispersed second phase and any user-defined sources, p : is the static pressure, $\vec{\tau}$: is the stress tensor, ρg : is the gravitational body force, F : is the external body forces, k_{eff} is the effective conductivity; the first two terms on the right-hand side of Eq. (5) are energy transfers due to conduction and viscous dissipation, respectively; and S_h is the volumetric heat sources.

The simulation domain as shown in Figure 1 represented by the LEU target surrounded on both sides by half thickness of the coolant channel, it has a dimension of 1.29mm x 44.6mm. The domain is 3D modeled, using Design Modeler integrated tool.

Meshing of the studied domain was carried out by using Meshing integrated tool. Structured meshing type is adopted for the geometry model. Several refinement of meshing were tested in order to lead to the appropriate one and to ensure minimum results uncertainties. A difference of less than 1% in two consecutive sets of results is taken as criteria of selecting the final used meshing [30,32].

For the boundary conditions as shows in Figure 1, overall the computational domain is classified into two categories, fluid and two solids domain (meat and cladding). The inlet of the LEU target is provided with velocity inlet with a turbulence intensity of 4.72 %, as calculated in Table 3. The exit of domain is set to pressure. Also, a constant heat flux is provided at the core of the meat.

The $k-\varepsilon$ standard turbulence flow model [33] is considered for the analysis of heat transfer and the flow streams of water flows between the target wall and the coolant channel inside the targets holder. The problem is treated as steady state mono-phase turbulent flow. Equations mentioned in Table 3 were used to determine the initial conditions and the boundary conditions for $k-\varepsilon$ turbulent flow model.

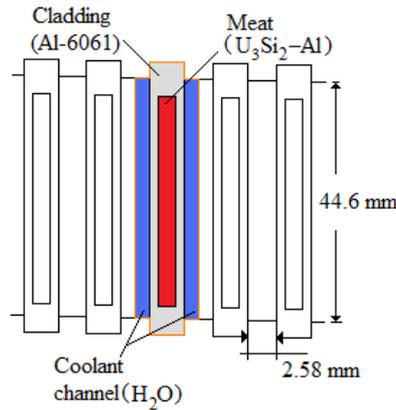


Fig. 1. Domain of simulation

Table 3
 Turbulence parameters [30]

Variable	Equation	Numerical value
Turbulent intensity (I)	$I = 0.16 * Re_{D_h}^{-\frac{1}{8}}$	4.72 %
Turbulent kinetic energy (k)	$k = \frac{3}{2} (V_m * I)^2$	0.164 m ² /s ²
Dissipation rate (ε)	$\epsilon = C_\mu^{3/4} \frac{k^{3/2}}{l'}$	60.31 m ² /s ³
Volumetric source (S _v)	$S_v = \frac{P * FQ * FZ}{L * l * e}$	S _v (BOC)=4.1E+09 (W/m ³) S _v (MOC)=3.2E+09 (W/m ³) S _v (EOC)=3.08E+09 (W/m ³)

where Re : is the Reynolds number ; D_h : is the hydraulic diameter ; V_m : is average coolant velocity ; l' : is turbulent length scale ; C_μ : is turbulence empirical constant equal 0.09 ; P : is a target power ; FQ : is power peaking factor ; FZ : is the axial peaking factor ; L : length of the fuel meat ; l : width of the fuel meat ; e : thickness of the fuel meat.

Table 4 lists the inputs parameters and correlated information for the numerical model.

Table 4
 Numerical model inputs

Input	Value
Solver	
Time	Steady
Type	Pressure Based
Velocity Formulation	Absolute
Gravity	-9.81 m/s ² (Z-direction)
Models	
Energy	Active
Multiphase	Off
Viscous	Standard k-ε
Near Wall Treatment	Scalable Wall Functions
Solution Methods	
Scheme	Coupled
Spatial Discretization	
Gradient	Least Square Cell Based
Pressure	Second Order
Momentum	Second Order Upwind
Turbulent Kinetic Energy	Second Order Upwind
Turbulent Dissipation Rate	Second Order Upwind
Pseudo Transient	Active

4. Results and Discussions

According to initial conditions and hypothesis, thermal hydraulic analysis during irradiation of the highest power LEU target of targets bundle was done at three statuses of KAERI reactor core. Used Defined Function (UDF) [34-35] elaborated in order to accurate the power volumetric source profile and materials properties (water coolant, aluminum cladding and LEU meat) with respect to the temperature variation.

As illustration of the results obtained, Figure 2 shows three contours of temperature distribution in LEU target plate and coolant channel at the Beginning of the Cycle of reactor core (BOC). Maximum temperature of 428.2K located at the center of the meat of target plate where maximum nuclear power pronounced and it decreases until a temperature of 355K at the entry and exit edges.

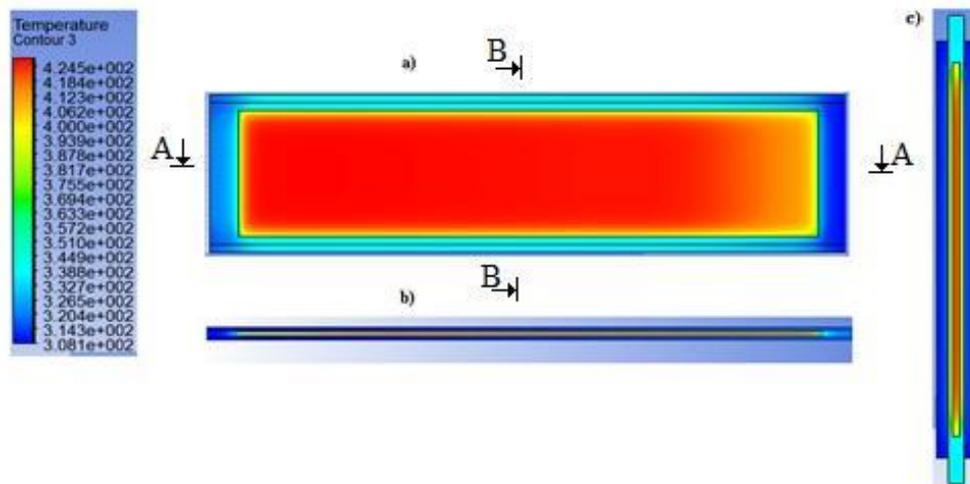


Fig. 2. Contours of temperature distribution of LEU target plate and coolant channel at BOC status a) Contour of temperature of target and coolant channel in direction of length and width, b) contour of temperature of target and coolant channel in direction A-A and c) contour of temperature of target and coolant channel in direction B-B

Figure 3 shows the radial distribution of LEU target plate and coolant channel temperatures (in direction of target plate thickness) at the entry edge, middle and exit edge positions in the case of BOC reactor core status. A 288.5K of temperature difference takes place between two edges of LEU target meat. The LEU center meat and outer cladding reaches maximum temperatures of 428.2K and 385.9K respectively, the maximum cladding temperature obtained is below the saturation temperature of the coolant which is of 390.5K at coolant pressure of 1.8bar and is in agreement with that obtained by Daeseong Jo [24]. The temperature difference between inlet and outlet water coolant is about 285.5 K. Furthermore, it is observed that the exit edge region temperature is higher than the center and entry edge regions as mentioned in Figure 4.

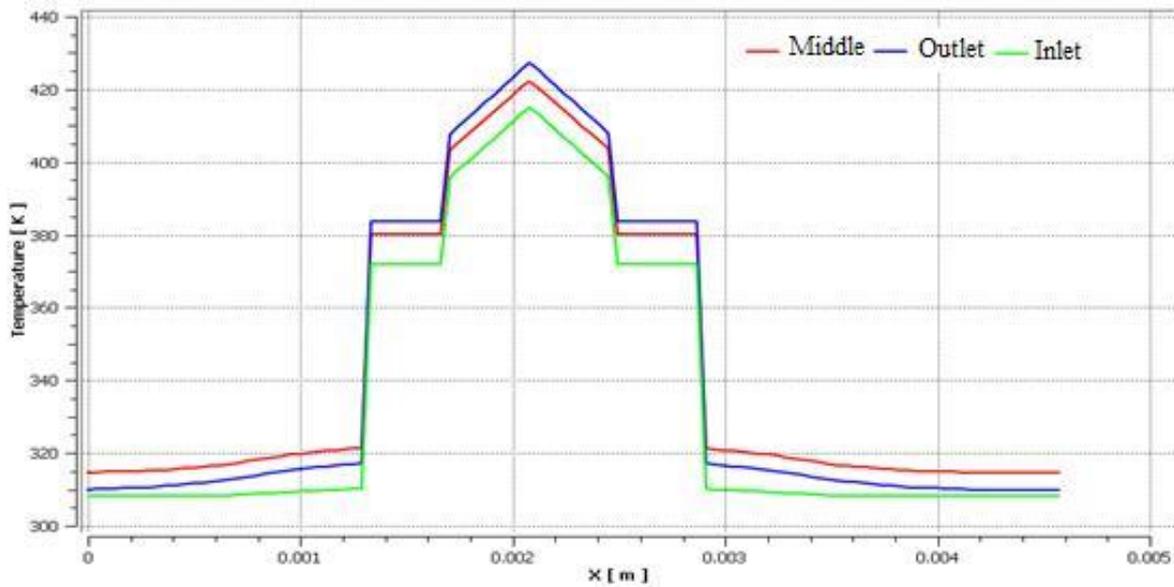


Fig. 3. Radial distribution of LEU target plate and coolant channel temperatures at BOC status

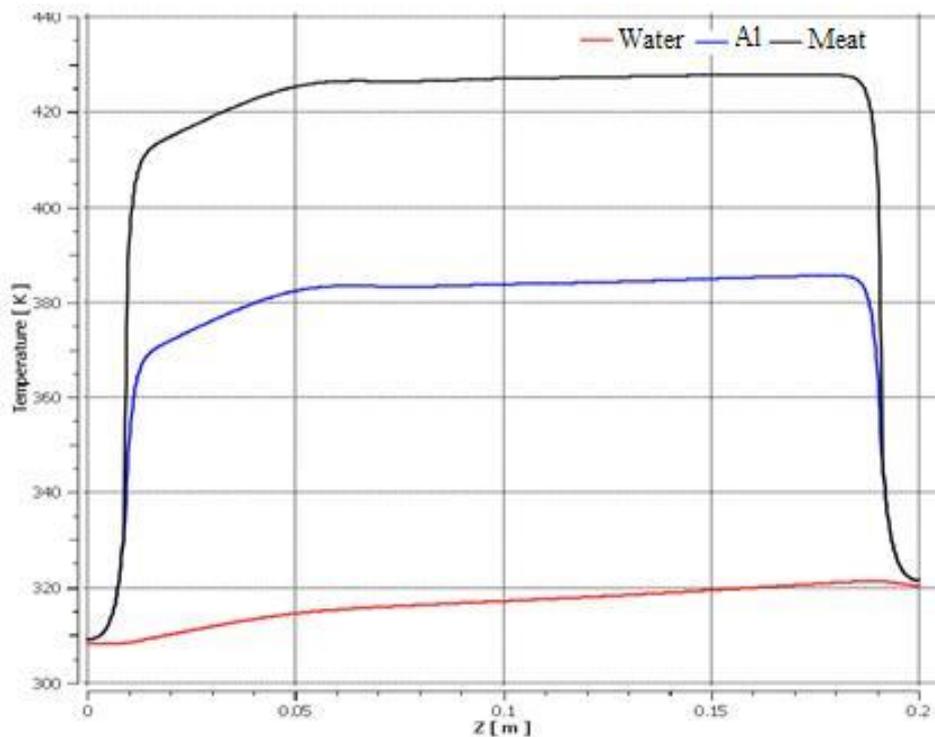


Fig. 4. Axial distribution of target plate temperature and coolant channel at BOC status

Maximum temperatures obtained of coolant, cladding and LEU meat at the three reactor core statuses (BOC, MOC and EOC) are summarized in Table 5. These results are compared with those obtained by using TMAP code [24]. It was noticed that it does not have an important discrepancy between CFD and TMAP results, CFD values are slightly larger than TMAP one's at BOC status. A maximum Absolute Relative Error (AER) of 9.72% of meat temperature at BOC status and minimum AER of 0.66% of meat temperature at EOC status are obtained. This difference between the two results can be due the calculation parameters, conditions and hypotheses.

Table 5
Coolant channel maximum temperatures

Temperature (K)	BOC	AER	MOC	AER	EOC	AER
	TMAP/CFD	%	TMAP/CFD	%	TMAP/CFD	%
Maximum coolant temperature	315.9/321.8	13.92	315.9/318.4	5.90	315.4/317.9	5.97
Maximum cladding temperature	385.8/385.9	0.09	376.5/366.9	9.32	366.5/363.2	3.55
Maximum meat temperature	414.5/428.2	9.72	401.5/399.6	1.48	394.8/394	0.66

Table 6 summarized thermal margins, CHF ratio (Minimal Departure from Nucleate Boiling, MDNBR), for the BOC, MOC and EOC statuses during irradiation. The obtained results using CFD calculation show that the MDNBR at the three statuses are less conservative to that obtained using TMAP code, a maximum AER is of 18.47% for BOC status and minimal AER is of 12.894 % for EOC status.

Table 6
Thermal margins results between two calculation codes

	MDNBR		
	(BOC)	(MOC)	(EOC)
TMAP	4.98	5.45	5.74
CFD	4.06	4.63	5.0
AER (%)	18.47	15.05	12.89

5. Conclusion

CFD model was established to analyze thermal hydraulic behavior of LEU target plate during irradiation at three statuses of KAERI reactor core. The results obtained were compared with those obtained by TMAP code. It was verified that target plate performed well under irradiation and irradiation conditions are below thermal margins. Acceptable differences in results between the two calculations, CFD and TMAP, were observed. This may be due to the calculation parameters and conditions. The CFD model will be extended to analyze the thermal hydraulic behavior at cooling period and at transfer period.

Acknowledgements

Authors present deep acknowledgements for Daeseong Jo *et al.*, from Korean Atomic Energy Research Institute (KAERI) for sharing information and guidance to conduct this work.

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