Thermal Contact Resistance and Ambient Temperature Effects on the Cooling of Mo$^{99}$ Plate Targets inside the Hot Cell

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ABSTRACT

The Cooling of Low Enriched Uranium (LEU) irradiated fuel plate targets that used for Molybdenum -99 productions, after their extraction from the nuclear reactor core require a good degree of interest. This is to satisfy two important criteria’s, the first keeping the radiation dose limits within the safety limits to protect the worker and the second to keep the integrity of the plate itself for easy handling and manoeuvring at the different stages. The cooling processes of the plate targets are passing through different modes, by forced convection cooling using water coolant inside the core during irradiation, by natural convection cooling using water coolant inside the reactor main tank during the decay period and finally by air convective and radiation inside the hot cell during their loading. In this study, a steady state mathematical model was developed to study the effect of thermal contact resistance and the ambient air temperature inside the hot cell on plate targets cooling by free convection during an abnormal condition of lose of electrical power supply, which means no ventilation system, is available. In this simulation, it assumed that three targets were unloaded on the table inside the cell in the form of pile, one target on the top of the other, as a purposed human error. The maximum pile temperature variation with target power ratio (target power in hot cell relative to target maximum power during irradiation) was analyzed and calculated. Also the probable temperature variation against different power ratios for one free vertical plate target hold by the Tele-manipulator, exposed to air free convection and thermal radiation on its both sides, during the loading process was estimated in this study. This study was clarified that, the contact resistance and the ambient temperature have an essential effects on the targets plate cooling inside the hot cell as will be clarified in that work.

Key words: Mo$^{99}$, Plate targets, hot cell

INTRODUCTION

The target used for Mo-99 production is a material containing uranium- 235 that is designed to be irradiated in a nuclear reactor. The target is designed to satisfy several requirements: First, it must be properly sized to fit into the irradiation position inside the reactor. Second, it must contain a sufficient amount of U-235 to produce the required amount of Mo-99 when it is irradiated. Third, it must have good heat transfer properties to prevent over-heating (which could result in target failure) during irradiation. Fourth, the target must provide a barrier to the release of radioactive products, especially fission gases, during and after irradiation. Fifth, the target materials must be compatible with the chemical processing steps that will be used to recover and purify Mo-99 after the target is irradiated as indicated in Committee on Medical Isotope Production without Highly Enriched Uranium (2009). To meet these criteria, targets are fabricated in a wide variety of shapes and compositions to meet the needs of individual Mo-99 producers. Targets may be shaped as plates (Figure 1), pins, or cylinders. Target compositions include uranium metal, uranium oxides, and alloys of uranium, nearly always with aluminium. Metallic targets are typically encapsulated in
aluminium or stainless steel to protect the chemically reactive uranium metal or alloy and to contain the fission products produced during irradiation. This encapsulation is referred to as the target cladding. Sometimes an intermediate barrier material such as aluminium or nickel is used to separate the cladding from the U-235 target material.

**FIG. 1** CNEA’s high-density LEU-aluminium dispersion targets. These targets have been used since 2002 to produce Mo-99 in Argentina. The target is approximately 15 cm in length. SOURCE: Courtesy of Pablo Cristini, CNEA, Argentina, Committee on Medical Isotope Production without Highly Enriched Uranium (2009)

### 1.2. Irradiation of Targets in a Nuclear Reactor

Mo-99 is produced in the uranium-bearing targets by irradiating them with thermal neutrons. Some of the U-235 nuclei absorb these neutrons, which can cause them to fission. The fission of the U-235 nucleus produces two but sometimes three lower-mass nuclei referred to as fission fragments. Approximately 6 percent of these fission fragments are Mo-99 atoms. Nuclear reactors provide an efficient source of thermal neutrons for Mo-99 production. This is why all major Mo-99 producers irradiate their targets in nuclear reactors. The amount of Mo-99 produced in a target is a function of irradiation time, the thermal neutron fission cross section for U-235, the thermal neutron flux on the target, the mass of U-235 in the target, and the half-life of Mo-99. For typical reactor thermal neutron fluxes on the order of $10^{14}$ neutrons per square centimetre per second, irradiation times of about 5 to 7 days are required to achieve near-maximum Mo-99 production in the targets, Committee on Medical Isotope Production without Highly Enriched Uranium (2009). Beyond these irradiation times, the amount of Mo-99 produced in the targets approximately balances the amount of Mo-99 being lost to radioactive decay, so further irradiation is not productive. Even at maximum production, only about 3 percent of the U-235 in the target is typically consumed. The remaining U-235 along with the other fission products and target materials are treated as waste. Technicium-99m (99mTc) has a 6 hour half-life and emits a gamma ray when it de-excites. Attached to various chemicals, it can be followed by its gamma emissions through the body and thereby can be used to examine the functioning of various organs. Its short half-life and lack of beta radiation minimizes unnecessary radiation doses. It is derived from molybdenum-99 (99Mo), which has a half-life of 2.7 days and decays into 99mTc. 99Mo is adsorbed onto the surface of a bed of small alumina particles in “generators” from which the 99mTc decay product is drawn off in solution.
1.3. Contact resistance

Contact resistance is primarily caused by the imperfect contact between two surfaces due to the presence of microscopic asperities of engineering surfaces. The real area of contact for such surfaces is only a small fraction (1-2%) of the nominal contact area even at very high pressure, Bowden and Tabor (1950). In spite of that, most of the heat through the interface flows through the actual contact spots, as the thermal conductivity of these contact spots is much higher than that of the surrounding gap. That reason causes constriction of heat lines through the bulk solid material in the vicinity of the contact interface, which leads to constriction resistance at all of the contacting asperities on a surface. A finite element analysis was carried out in order to investigate the role of thermal contact resistance on heat management within a simple central processing unit (CPU)/heat sink assembly. A special attention is paid in assessing the effect of surface roughness characteristics, mechanical and thermal properties of the contacting bodies, applied contact pressures and the use of thermal interface materials on the maximum temperature experienced by the CPU. Two classes of thermal interface materials are shown in Figure 2: (a) phase-change materials and (b) acrylic- or silicone-based tapes are considered. The results clearly reveal that plastic deformation of micro-contacts (promoted by high contact pressures and lower micro-hardness levels) and the use of thermal interface materials which eliminate (high thermal resistance) micro-gaps can significantly lower the overall CPU/heat sink thermal contact resistance and facilitate heat management. It is also shown that the retention of asperity micro-contacts and good wetting of the mating surfaces by the thermal interface material are critical for achieving an effective removal of the heat generated by the CPU, Grujicic et al. (2005).

![Fig. 2. A schematic of two contacting bodies: (a) in direct contact and (b) separated by a thermal interface material. The arrows denote the heat flow across the interface](image)

The general theory adopted for the prediction of thermal contact resistance was modified by using the guarded hot plate method for thermal conductivity measurements. A good estimation of thermal resistance with a variety of interface materials was predicted. The effects of pressure, material hardness, surface roughness and thermal properties of interface material on the thermal resistance between two smooth steel surfaces were studied, Wolf and Schneider (1998). A predictive model for estimating thermal contact resistance between two nominally flat rough surfaces was developed and experimentally validated. In that study, the actual area of contact for each contact spot was calculated, also the effects of constriction resistance and gas gap conductance between the contacting surfaces were calculated by Vishal Singhal et.al. (2005). Generally the thermal contact conductance or resistance is of interest in many fields including internal combustion and superconductors.
2. Thermal model

A steady state, one dimensional axial heat flow model is built to simulate the in pile of three irradiated Mo-99 plate targets rested on a table inside the hot cell as a result of an operator error during the handling and transporting process of the targets, as shown in Figure 3. The table is simulated as a four diagonal strips each with length $L_d$ and cross sectional area $W_d \times t$, while the table leg length is $L_{leg}$ and leg cross sectional area is $A_{leg}$.

The in pile simulation of the three irradiated plate targets is carried out for different power ratios (plate decay power to plate nominal power during irradiation) to calculate the maximum probable temperature inside each plate to ensure the limit of blistering of the Aluminium cladding ($< 400 \, ^\circ C$) for safe handling inside the hot cell without releases of radioactive nuclides or to avoid deformation during the transportation to the process plant. The thermal resistances for heat transfer through each plate includes the thermal resistance of the clad, the thermal resistance of the fuel meat and the contact resistance between each two contact plates or between the bottom plate surface and the stainless steel table. A convective and thermal radiation boundary condition was considered for the upper plate so; the simulation was carried out at two ambient temperatures to evaluate the ambient temperature effects. During the normal procedures of handling of the irradiated plate targets inside the hot cell, holding the plate by the tele-manipulator tool free of contact with the other plates or with the table, only expose to convective and thermal radiation boundary condition on both faces, the plate target simulation during that situation is carried out also in this study.

The upper plate steady state energy balance equation;

$$Q_{gen} \frac{(T_m - T_{m2})}{\left(\frac{2\delta_{al}}{k_{al} \cdot A} + \frac{\delta_m}{k_m \cdot A} + \frac{1}{TCC_{al-al}}\right)} - Q_{rad} - Q_{conv} = 0.0 \tag{1}$$

The intermediate plate energy balance equation;

$$Q_{gen} \frac{(T_{m2} - T_{m1})}{\left(\frac{2\delta_{al}}{k_{al} \cdot A} + \frac{\delta_m}{k_m \cdot A} + \frac{1}{TCC_{al-al}}\right)} - \left[\frac{2\delta_{al}}{k_{al} \cdot A} + \frac{\delta_m}{k_m \cdot A} + \frac{1}{TCC_{al-al}}\right] = 0.0 \tag{2}$$

The third plate energy balance equation;

$$Q_{gen} \frac{(T_{m3} - T_{m2})}{\left(\frac{2\delta_{al}}{k_{al} \cdot A} + \frac{\delta_m}{k_m \cdot A} + \frac{1}{TCC_{al-al}}\right)} - \left[\frac{2\delta_{al}}{k_{al} \cdot A} + \frac{\delta_m}{k_m \cdot A} + \frac{1}{TCC_{al-al}}\right] = 0.0 \tag{3}$$

The upper plate boundary condition;

$$Q_{rad} - Q_{conv} = 0.0 \tag{4}$$
The energy balance equation for a free vertical plate;

\[ R Q_{\text{gen}} - 2 Q_{\text{rad}} - 2 Q_{\text{conv}} = 0 \] (5)

Heat transfer by free convection;
The Rayleigh number is defined as follows

\[ R_d = \frac{2}{T_d + T_{\text{inv}}} \alpha (T_d - T_{\text{inv}}) \left( \frac{A}{P} \right)^3 \] (6)

For a horizontal plate, Nusselt number can be calculated as follows [6];

\[ \text{Nu} = \begin{cases} 
0.54 \text{Ra}^{0.25} & \text{if } 1 \cdot 10^4 \leq \text{Ra} \leq 1 \cdot 10^7 \\
0.15 \text{Ra}^{0.33333333} & \text{if } 1 \cdot 10^7 \leq \text{Ra} \leq 1 \cdot 10^{11} \\
"error" & \text{otherwise} 
\end{cases} \] (7)

For a vertical plate, Nusselt number can be calculated as follows [6];

\[ \text{Nu} = \frac{0.825 + 0.387 \text{Ra}^{1/6}}{1 + (0.492/(\Pr t)^{1/6})^{27}} \] (8)

\[ h = \frac{\text{Nu} \cdot k}{L} \] (9)

\[ Q_{\text{conv}} = h \cdot A (T_d - T_{\text{inv}}) \] (10)

Heat transfer by thermal radiation;

\[ Q_{\text{rad}} = \varepsilon \sigma A \left( T_d^4 - T_{\text{inv}}^4 \right) \] (11)

The conductance between two Aluminium plates (TCC_{al-al}) or between Aluminium and stainless steel (TCC_{al-ss}) were adopted according to pressure and surface roughness as shown in references [5] and [6].

RESULT AND DISCUSSIONS

The previous system of equations through 1 to 5 was numerically calculated by using the MATHCAD software to determine the maximum temperature in each plate from the table surface. Two leading parameters were supposed in that simulation, the first parameter was the maximum power that could be generated inside the irradiated Mo-99 plate target during the reactor full power operation \(Q_{\text{gen}} = 10\text{ Kwatt for thermal neutron flux of } 10^{14} \text{ nv}\), while the second parameter was the power ratio parameter \((R= \text{ decay power/nominal power})\). Different power ratios \((0.16, 0.18, 0.20 \text{ and } 0.22)\) were tested during the simulation to ensure closeness from the cladding blistering limit \((\leq 400^\circ\text{C})\) at the purposed case study \((Q_{\text{gen}} = 10\text{ Kwatt})\). For a different maximum generated powers \((Q_{\text{gen}})\) inside the Mo-99 plate target, there were another different power ratios should be tested to attain the cladding blistering limit. Figure 4 shows the in pile simulation of three plates at a power ratio of 0.16% with and without contact resistance consideration for two different environmental temperatures \((25^\circ\text{C and } 50^\circ\text{C})\) to simulate the purposed winter or summer conditions inside the hot cell. In this

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simulation, no ventilation system is available inside the hot cell, so a free convection and thermal radiation boundary conditions were considered.

For no contact resistance consideration, the temperature distribution of the in pile three plates is approximately flat with approximately constant value of 298.2 °C at an ambient temperature of 25 °C. This flat constant value was raised to 321 °C when the ambient temperature was increased to 50 °C. For contact resistance consideration, the plate in contact with the table recorded a minimum temperature of 299.8 °C, a gradual increasing is temperature with distance was observed while moving upward. The maximum temperature was attained at the intermediate plate meat with a value of 306.406 °C, hence, a gradual decreasing in temperature with distance was observed but with little slope till reaching the upper plate surface the recorded temperature value was 304.866 °C. A similar trend but with higher values was noticed if the contact resistance was taken into consideration at higher ambient hot cell temperature of 50 °C. The plate temperature in contact with the table recorded 322.613 °C while a maximum temperature of 329.1 °C was recorded at the intermediate plate meat and 327.437 °C at the upper plate surface. For this simulation, at a power ratio of 0.16%, all the recorded temperature were below the cladding blistering limit (400 °C). Different simulation steps were performed at different higher power ratios as shown in Fig. 5a and Fig. 5b, looking for the power ratio at which the limit of blistering is reached. The results showed the same trend as in Fig. 4 but with higher values because of the increase in power ratio.
For many simulations, the maximum in pile temperature was recorded against the power ratio as shown in Fig. 6. This maximum temperature was occurred at the intermediate plate. For an ambient temperature of 25°C, the contact resistance effect made this maximum temperature very close to the blistering limit at a power ratio of 0.22%. For an ambient temperature of 50°C, without the contact resistance effect the blistering limit was exceeded at a power ratio lies between 0.21% and 22% while with contact resistance consideration this blistering limit is occurred between 0.2% and 0.21%.
Figure 7 shows the average percentage of the amount of heat transfer by different mechanisms through the simulations at different power ratios. The amount of heat transfer by conduction though the table legs showed the highest rate of heat transfer, about 60%. While the heat transfer by free convection averagely recorded 30% and the remainder 10% for the heat transfer by thermal radiation during this simulation.

Solution of equation (5) for one plate under convective and thermal radiation boundary conditions on both sides inside the hot cell gave a good estimation for the maximum expected surface temperature of the plate. This simulation was carried out at two different ambient temperatures also as shown in Fig.8. The plate surface temperature increases with the increase in power ratio. The increase in ambient temperature increases the expected plate surface temperature. The difference in plate surface temperature for the two ambient temperatures (25 °C, 50 °C) was approximately constant at a value of 22.1 °C. In comparing a one holding plate (not in contact with the table) maximum surface temperature and the three in-pile plates (laid on table) maximum temperature for the purposed power ratio, it was shown a great difference in temperature can lead to blistering limit for the in-pile plates.
CONCLUSION

Generally, transferring the irradiated Mo-99 plate targets from the water to the ambient air inside the hot cell expose them to a poorer cooling due to the change in medium thermal properties. Keeping the Mo-99 plate target in a vertical position during the handling is better than rest it horizontally on a table. The hot cell operator not allowed putting the plates in the in-pile form because the contact resistance between plates and the high ambient temperature could raise the plate temperature to the blistering temperature limit. The ventilation system inside the hot cells plays as a strong factor for limiting the plate temperature rise, because it supplies the cell with forced – cooled air, that make the mode of plate cooling is a forced convection instead of free convection and prevent the hot cell temperature rise.

Nomenclature

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<tr>
<th>Symbol</th>
<th>Description</th>
<th>Unit</th>
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<tbody>
<tr>
<td>A</td>
<td>Plate surface area</td>
<td>m²</td>
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<tr>
<td>A_{leg}</td>
<td>Table leg cross section Area</td>
<td>m²</td>
</tr>
<tr>
<td>h</td>
<td>Heat transfer coefficient</td>
<td>W/m².°C</td>
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<tr>
<td>k</td>
<td>Thermal conductivity</td>
<td>W/m.°C</td>
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<tr>
<td>L_{leg}</td>
<td>Leg length</td>
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</tr>
<tr>
<td>L_d</td>
<td>Table diagonal strip length</td>
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</tr>
<tr>
<td>Nu</td>
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<tr>
<td>P</td>
<td>perimeter</td>
<td>m</td>
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<td>Q_{gen}</td>
<td>Heat generation per plate</td>
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<td>Convective heat flow</td>
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<td>Irradiative heat flow</td>
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<td>R</td>
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<tr>
<td>Ra</td>
<td>Rayleigh number</td>
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<tr>
<td>T</td>
<td>Temperature</td>
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<tr>
<td>TCC</td>
<td>Conductance</td>
<td>W/K</td>
</tr>
<tr>
<td>t</td>
<td>Table thickness</td>
<td>m</td>
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<tr>
<td>W_t</td>
<td>Table diagonal strip width</td>
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Greek symbols

<table>
<thead>
<tr>
<th>Symbol</th>
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<tr>
<td>(\delta_{al})</td>
<td>aluminium cladding thickness</td>
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$\delta_m$ fuel meat thickness
$\alpha$ Thermal diffusivity $m^2/s$
$\nu$ Kinematic viscosity $m^2/s$
$\varepsilon$ emissivity factor
$\sigma$ Stefan-Boltzmann constant $W/m^2.K^4$

Subscripts
al aluminium cladding
al-al aluminium to aluminium contact
al-ss aluminium to stainless steel contact
ss stainless steel
m Meat of fuel
m1 Upper plate meat
m2 Intermediate plate meat
m3 lower plate meat
S1 Upper plate surface
inv environment

REFERENCES