Assessment of Candidates for Target Window Material in Accelerator-Driven Molybdenum-99 Production

Nuclear Engineering Division

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ASSESSMENT OF CANDIDATES FOR TARGET WINDOW MATERIAL IN ACCELERATOR-DRIVEN MOLYBDENUM-99 PRODUCTION

ABSTRACT

NorthStar Medical Technologies is pursuing production of an important medical isotope, Mo-99, through a photo-nuclear reaction of a Mo-100 target using a high-power electron accelerator. The current target utilizes an Inconel 718 window. The purpose of this study was to evaluate other candidate materials for the target window, which separates the high-pressure helium gas inside the target from the vacuum inside the accelerator beamline and is subjected to significant stress. Our initial analysis assessed the properties (density, thermal conductivity, maximum stress, minimum window thickness, maximum temperature, and figure of merit) for a range of materials, from which the three most promising were chosen: Inconel 718, 250 maraging steel, and standard-grade beryllium. These materials were subjected to further analysis to determine the effects of thermal and mechanical strain versus beam power at varying thicknesses. Both beryllium and the maraging steel were calculated to withstand more than twice as high beam power than Inconel 718.

1 INTRODUCTION

NorthStar Medical Technologies is pursuing production of an important medical isotope, Mo-99, through a photo-nuclear reaction on Mo-100 using a high-power electron accelerator. To make this pathway for Mo-99 production commercially feasible, one has to utilize an enriched Mo-100 target. The cost of the Mo-100 is in the range of \$1000 for kilogram quantities. The high cost of the target material and the desire to maximize production drive a requirement to minimize the size of the target and maximize the power deposition on the target. High power deposition in the target requires unconventional cooling. Argonne National Laboratory (Argonne) and Los Alamos National Laboratory (LANL) have demonstrated gaseous helium cooling of the metal molybdenum target at high power densities. This method of cooling requires high flow and high pressure of the helium gas.

The target window separates the high-pressure helium gas inside the target from the vacuum inside the accelerator beamline; this pressure differential introduces significant stress on the window. Also, the high-current electron beam transmitting through the window deposits significant power into the window material. The window is cooled by gaseous helium only on one side, which makes the window the most stressed component of the target and makes design and the choice of material for the window critical for operation of the target. The current target designed by LANL and tested at Argonne utilizes an Inconel 718 window. The purpose of this study was to evaluate other candidate materials for the target window.

Note that the work presented here is only for comparison of materials and their ability to handle power from the electron beam. The window geometries and analysis results in this report are not for optimized designs and are only used as a means for comparing the materials. Presented here are (1) the many materials considered and three selected for full evaluation, (2) other factors considered (such as beam shape and window shape) in developing our analysis model, (3) the reasoning behind our analysis model (such as the chosen geometry and boundary conditions), (4) the basis for setting temperature and stress limits, and model calculations for final selection of window target material.

2 MATERIALS CONSIDERED

As presented in Table 1, a comprehensive list of metals and metal alloys was initially considered for potential window-material candidates based on their high specific strength (ratio of yield strength to density). This initial list included high temperature alloys, referred to as “superalloys,” that are based mainly on nickel or cobalt, such as Inconel 718 (the material selected by LANL), Hastelloy X, Inconel 706, Waspaloy, Rene 41, and Haynes Alloy 25. Steel alloys included 316 stainless steel (SS), Grade 250 maraging steel, and AerMet 100. Other materials included titanium alloy, standard-grade beryllium, and magnesium alloy.

Although aluminum alloys were included for comparison, they were not considered viable candidates due to their poor performance at elevated temperatures. Magnesium was removed from the list of potential candidates for the same reason, once reliable temperature-dependent mechanical property data had been acquired for the material.

TABLE 1 Comprehensive List of Initial Candidates for Window Material

Material	Density (kg/m ³)	Thermal Conductivity (W/m-°C)	Maximum Stress (MPa)	Minimum Window Thickness (mm)	Maximum Temperature (°C)	Figure of Merit (FOM)
Inconel 718	8,221	17.3	456	1.15	403	1
Hastelloy X	8,221	26.0				Disqualified ^a
Inconel 706	8,055	22.5	75	2.87	1,280	2.45
Waspaloy	8,193	17.3	357	1.30	481	1.13
Rene 41	8,249	17.3	507	1.09	388	0.96
L-605 Haynes Alloy 25	9,134	19.0				Disqualified ^a
316 SS	7,806	22.5				Disqualified ^a
250 Maraging Steel	7,916	29.4	706	0.93	269	0.78
AerMet 100	7,889	31.2	793	0.87	249	0.73
2024-T81 Aluminum	2,768	173.1				Disqualified ^a
6061-T6 Aluminum	2,713	173.1				Disqualified ^a
Titanium Alloy AMS 4910	4,484	13.9	175	1.88	497	0.90
Beryllium Standard Grade	1,855	138.5	147	1.96	131	0.39
Magnesium Alloy	1,800	77.0				Disqualified ^a

^a Indicates that there is no window thickness value that satisfies the allowable stress requirement for the baseline beam power.

We have also considered, but not included in the table, several materials that are known to have high specific strength but lack sufficient material property data (chromium, molybdenum, vanadium, and niobium). Materials with high melting points, tungsten and tantalum, were also considered but were not included in the table because of insufficient property data and their high density. Non-metallic materials were not examined in this study due to their weak mechanical properties and brittle behavior, which cause them to be an unreliable material for use as a pressure boundary.

Based on the calculated results from Table 1 (in particular, the FOM value), as well as the above-noted reasons, the three materials in Table 2 were selected for further evaluation.

TABLE 2 Final Three Materials Selected for Evaluation

Material	Density (kg/m ³)	Yield Strength (MPa)	Specific Strength (m ² /s ²)	FOM
Inconel 718	8220	1000	121,600	1.00
250 Maraging Steel	7916	1640	207,300	0.78
Beryllium	1855	345	185,900	0.39

3 CALCULATIONS USED

For all materials in Table 1, calculations were performed to develop a FOM that quantitatively evaluated each listed material for its potential usefulness for beam window applications. The FOM is defined as:

$$FOM = \frac{\rho t}{\rho_I t_I} \quad (1)$$

where:

ρ = density of material to be evaluated

t = minimum acceptable thickness of material to be evaluated

ρ_I = density of Inconel 718

t_I = minimum acceptable thickness of Inconel 718

FOM = figure of merit

Note: Because Inconel 718 is currently used as a beam window material, we used its properties as a basis to normalize the FOM. Since Inconel 718 has a $FOM = 1$, a material with $FOM < 1$ is potentially a better window material choice.

This definition implicitly assumes that the beam attenuation is proportion to the material density and the distance the beam travels through the material. Therefore, the FOM is inversely related to the beam attenuation and, in turn, the heat generation in the beam window. Further, it is assumed that minimum beam attenuation and heat generation are highly desirable for beam window materials (i.e., materials with low FOMs are desirable).

The window geometry is assumed to be a disk with a 25-mm diameter and an adjacent flow channel depth of 0.71 mm (i.e., depth in the axial direction of the beam). The window was assumed to be cooled by helium flowing through the 0.71-mm deep channel at an absolute pressure of 31 bar and ambient temperature.

The minimum acceptable window thickness is based on equating the maximum calculated stress in the window to half the yield stress of the material at the maximum operating temperature. Both the maximum calculated stress and temperature occur at the center of the window. Also, the half-yield stress is considered to be an acceptable allowable stress.

Because temperature is a function of thickness for a given beam power, and in turn, the allowable stress is a function of temperature, an iterative (or graphical) procedure is required to determine the required minimum thickness for use in the FOM calculation. A typical graphical

method using Inconel 718 is demonstrated on the plot of stress versus window thickness in Figure 1.

FIGURE 1 Stress Plot Used to Determine Minimum Acceptable Window Thickness

The calculated stress curve in Figure 1 was developed by using the classical stress correlation for a flat round disk with free edges to determine the maximum stress at the operating pressure differential of 31 bar:

$$\sigma_{max} = \frac{3}{8} \frac{a^2 p (\gamma + 3)}{t^2} \quad (2)$$

where:

a = radius of disk

p = uniform differential pressure loading across disk

γ = Poisson's ratio for the material

t = uniform thickness of the disk

σ_{max} = maximum stress in the disk under pressure loading (occurs at the center of the disk)

Determining the allowable stress curve in Figure 1 required the maximum window temperature as a function of thickness. Therefore, we performed a 1-D heat transfer analysis

considering an infinite plate with internal heat generation, a convective boundary on the helium side, and an insulated boundary on the vacuum side. Radiant heat transfer was assumed negligible. The 1-D heat transfer correlation used was:

$$T_{max} = qt \left(\frac{t}{2k} + \frac{1}{h} \right) + T_{amb} \quad (3)$$

where:

q = uniform internal heat generation in the plate

t = thickness of the plate

k = thermal conductivity of the plate (assumed to be constant at all temperatures)

h = thermal convective coefficient at one side of the plate (see Eq. 6)

T_{amb} = ambient temperature of the helium

T_{max} = maximum temperature of the plate (occurs at insulated side of plate)

The heat-generation density in the window was determined by assuming a baseline beam power of 30 kW. An analysis using the Monte Carlo N-Particle code (MCNP) was then performed for an Inconel 718 window with a thickness of 0.38 mm at its center. The Inconel 718 heat generation was used as a basis for determining the heat generation density in the other materials by assuming that beam attenuation by the materials was proportional to their densities; hence:

$$q_m = q_I \frac{\rho_m}{\rho_I} \quad (4)$$

where:

q_I = internal heat generation density in Inconel 718

ρ_m = density of material to be evaluated

ρ_I = density of Inconel 718

q_m = internal heat generation density of the material to be evaluated

The thermal convective coefficient in Eq. 3 was calculated by assuming forced flow between two infinite plates spaced 0.71 mm apart. The average helium flow was assigned an assumed typical velocity in the target of 130 m/s at a pressure of 30 bar. The Nusselt number correlation for this flow condition is:

$$Nu = 0.023 Re^{0.8} Pr^{0.33} \quad (5)$$

where:

Re = Reynolds number for helium flow (fully turbulent for the above flow conditions)

Pr = Prandtl number for helium at the flow conditions

Nu = Nusselt number

The corresponding convective coefficient is given by:

$$h = \frac{Nu k_f}{d} \quad (6)$$

where:

Nu = defined above

k_f = thermal conductivity helium at the flow conditions

d = hydraulic diameter (2 x distance between the infinite plates)

h = thermal convective coefficient used for the 1-D thermal conduction equation

The result of this 1-D heat transfer analysis for the Inconel 718 case is shown in Figure 2. This plot of window thickness versus temperature, combined with the graph of yield stress

FIGURE 2 Maximum Window Temperature Using 1-D Thermal Heat Transfer Correlation

versus temperature in Figure 3, was used to develop the allowable stress curve. We obtained temperatures at four window thicknesses from Figure 2, then using Figure 3, derived the yield stress at those temperatures. Hence, the window thickness versus half-yield stress could be plotted as the allowable stress versus window thickness, as shown in Figure 1.

FIGURE 3 Effect of Temperature on Yield Strength of Inconel 718 (from Ref. 2)

Lastly, the minimum and maximum window thicknesses were found at the intercepts of the calculated stress curve and the allowable stress curve. The plot can be interpreted as any window thickness between the two intercepts (i.e., minimum and maximum as indicated on the plot) that meets the allowable stress requirements, or any window thickness where the calculated stress curve is below the allowable stress curve. Also, if no intercept exists for a particular material (i.e., the allowable stress curve is always below the calculated stress curve), it does not meet the allowable stress requirements regardless of window thickness for the fixed baseline beam power in this study (and is noted as “disqualified” in the table).

In Table 1, the first column lists the proposed material to be evaluated. The density and thermal conductivity are then given for each material. The density is assumed constant at ambient temperature. The thermal conductivity values are assumed at the maximum temperature. Note that Poisson's ratio used in Equation 2, not shown in the table, is assumed at the maximum temperature. The maximum stress and the corresponding minimum window thickness are obtained from plots similar to that shown in Figure 1. These values are determined at the intercept of the allowable stress and calculated stress curves on this graph. The maximum temperature for the minimum window thickness is determined from Figure 2. The FOM for each material is listed in the last column and is calculated using the corresponding density, minimum thickness, and Equation 1.

4 OTHER FACTORS CONSIDERED

Window geometries with non-uniform thickness were studied. Because the shape of the electron beam can be modified, the possibility exists of finding an optimum combination of beam shape and window-thickness profile. For example, instead of a Gaussian beam profile, the electron beam can be scanned to provide a uniform or even toroid profile.

When searching for an optimal thickness profile for a disk, it is important to think in terms of cylindrical coordinates and avoid direct comparison to, for example, a simple 1D structural beam model. One example is determining an optimum location in the radial direction for a stiffening ring to increase the overall stiffness of the window. A numerical solution was used to examine this particular scenario (Figure 4). The location of the ring, r , is allowed to vary as a fraction of the radius of the disk R . All other parameters are held constant.

FIGURE 4 A quarter symmetric model of a disk with a stiffening ring feature. The edge of the disk is pinned. All parameters are held constant except for the radial location of the ring.

Calculations with an initial structural model without the stiffening feature were run to obtain a baseline value for deflection in the disk. The deflection results for r/R from 0.15 to 0.85 were compared to the baseline results and are summarized in Figure 4. The greatest effect on minimizing stress and deflection occurs at approximately $r/R = 0.6$.

5 SELECTION OF MATERIAL FOR MAXIMUM BEAM POWER

Having narrowed the choice of material candidates, we developed a modeling method for a parametric study. The modeling method uses volumetric heat generation data calculated with the Monte Carlo N-Particle (MCNP) code, considers temperature and stress limits for the particular materials, and includes the effects of thermal and mechanical strain. Calculated results were then compared to values of stress relative to the material versus beam power at varying thicknesses.

5.1 STRESS AND TEMPERATURE LIMITS

For each material candidate, limits had to be set for both maximum stress and temperature. A value of allowable stress for the three candidate materials was not readily available from an established and well recognized code, such as the ASME Boiler & Pressure Vessel Code. Therefore, 50% of material yield strength was chosen as a limit on stress for the purpose of comparing the materials with this model. Since the yield strength is temperature dependent, the limit stress was calculated from the maximum temperature found in the analysis results. This limit was then applied over the entire window. Although this approach can lead to increased conservatism, it was done for simplicity in developing a parametric model. Additionally, Inconel and maraging steel are precipitation hardening materials and their mechanical properties degrade with prolonged exposure to temperatures over 700 °C and 400 °C, respectively.

5.2 MATERIAL PROPERTIES

All three material models relied on temperature-dependent properties obtained from the 2003 edition of Department of Defense's *Handbook for Metallic Materials and Elements for Aerospace Vehicle Structures* [2]. These properties are summarized in Figures 5-10.

FIGURE 5 Thermal properties versus temperature for Inconel 718 [2]

FIGURE 6 Yield strength and modulus of elasticity versus temperature for Inconel 718 [2]

FIGURE 7 Thermal properties versus temperature for grade 250 maraging steel [2]

FIGURE 8 Yield strength and modulus of elasticity versus temperature for grade 250 maraging steel [2]

FIGURE 9 Thermal properties versus temperature for standard grade beryllium [2]

FIGURE 10 Yield strength and modulus of elasticity versus temperature for standard grade beryllium [2]

5.3 MODEL FOR TEMPERATURE AND THERMAL STRESS

The first analysis model calculates the temperature distribution from the electron beam and the resulting stress induced by variation of temperature within the material of the window.

Thermal boundary conditions included volumetric heat generation and a convective boundary applied to the bottom of the disk. Heat generation rates were determined based on calculations from the MCNP code that were scaled linearly for varying beam power (Figure 11). A heat transfer coefficient of 2795 Btu/hr-ft²-°F (15870 W/m²-K) with bulk temperature of 70 °F (~ 21 °C) was applied to the bottom of the disk (Figure 12).

A common concern with modeling stress from thermal strain is over constraining the model. Fixed displacement constraints in a finite element model are infinitely rigid and can lead to unrealistically high stresses. As an alternative, the model can be constrained such that it is free to expand with temperature (i.e., a uniform change in temperature will result in zero strain). The stress then arises from temperature variation within the material only. Figure 13 shows the use of symmetry constraints and a single-point constraint fixed in the y-direction. This provides the idealized strain-free condition for uniform changes in temperature. This approach is considered valid as a means for comparing the material candidates.

FIGURE 11 (Top Row) Volumetric heat generation (kW/cm³ per kW of beam power) calculated by MCNP code with the data displayed for a cross section of the window. (Bottom Row) Corresponding heat generation rates (W/cm³) after importing the data into the ¼ symmetric model used in finite element model.

FIGURE 12 Thermal stress model based on hexagonal elements with a minimum of four elements through the thickness used in simulations (Ref. 2). As shown on the right, the bottom surface of the window model has a convective boundary applied with a heat transfer coefficient of 2795 Btu/hr-ft²-°F (15870 W/m²-K) and bulk temperature of 70 °F (~ 21 °C).

FIGURE 13 Thermal stress calculation based on symmetry constraints (highlighted in red) and a point at the center fixed in the y-axis direction. This minimal constraint setup allows the model to deform based on variation in temperature only, as if the window were merely resting on a table.

5.4 MODEL FOR MECHANICAL STRESS DUE TO PRESSURE

The main purpose of the window is to provide a pressure boundary between the target compartment and the evacuated beam tube. The other side of the window, opposite the vacuum, holds helium at 30 bar. A separate model was used to calculate the effects of pressure loading on the window using temperature-dependent material properties but without the effects of thermal strain, as the mechanical boundary conditions are necessarily different.

The mechanical stress model uses the same mesh and quarter symmetry created in the thermal model. A pressure loading of 30 bar is applied along the bottom surface of the window model, which is the same surface as the convective boundary. Taking advantage of the symmetry conditions, the only remaining degree of freedom for this model is in the y-axis direction. The surface at the outer radius is constrained by being fixed along the y-direction (Figure 14). Adding any additional constraints at the outer radius adds rigidity, which provides stiffness to the window, and is not realistic.

FIGURE 14 Pressure load of 30 bar applied at the bottom surface of the window model. The edge of the window is fixed in the direction of the Y-axis.

5.5 MODEL OUTPUT RESULTS AND DISCUSSION

Figures 15 and 16 show the calculated temperature contours and stress resulting from thermal strain varying within the material, respectively. The temperature plots are from the results of the maximum beam power for the respective material. The thermal stress is highest in tension around the edge of the window as the hot center tries to expand against the cooler material surrounding it. Areas of high stress at the center of the windows are in compression.

FIGURE 15 Temperature (°C) for three materials calculated with thermal model. The convective boundary is at the bottom of the window. The electron beam direction is indicated by the black arrow. The convective boundary is on the opposite side of the beam.

FIGURE 16 Stress intensity (MPa) for three materials calculated with thermal model. The model is constrained such that a uniform change in temperature would result in zero stress. Thus, in this model, thermal stress is only a result of non-uniform distribution of temperature within the material. The electron beam direction is indicated by the black arrow. The convective boundary is on the opposite side of the beam.

Figures 17 and 18 show the results of pressure loading and the combination of the effects of mechanical and thermal loading.

FIGURE 17 Stress due to pressure loading. Plotted as stress intensity in MPa. The electron beam direction is indicated by the black arrow. The convective boundary is on the opposite side of the beam.

FIGURE 18 Total stress combining results from thermal and pressure loading. Plotted as stress intensity in MPa. The electron beam direction is indicated by the black arrow. The convective boundary is on the opposite side of the beam. Note that the maximum value of stress intensity for the steel window occurs at the center of the window on the convective side.

After running the analyses for the three materials at different thicknesses and beam powers, the results were incorporated into plots. Each of the plots shows calculated stress, which is a combination of thermal and mechanical stress, and the limit stress as a function of material thickness. The variation of limit stress is directly coupled to the temperature of the material. The plots for Inconel 718, grade 250 maraging steel, and standard grade beryllium are presented in Figure 19, Figure 20 and Figure 21, respectively. The maximum beam power for the maraging steel window is primarily constrained by the temperature dependence of material yield strength. However, Inconel 718 and standard grade beryllium reach their respective limits due to increasing thermal stress.

FIGURE 19 Calculated maximum stress and limit stress versus material thickness for Inconel 718 at beam powers of 15, 17.5, and 20 kW. The calculated stress is a combination of both stress due to thermal variation within the material, and stress due to the pressure differential. Limit stress is calculated based on the maximum temperature in the window.

FIGURE 20 The calculated maximum stress and limit stress versus material thickness for 250 grade Maraging Steel at beam powers of 30, 35, and 39 kW. The calculated stress is a combination of both stress due to thermal variation within the material, as well as stress due to the pressure differential. Limit stress is calculated based on the maximum temperature in the window. The curves for 35 kW and 39 kW of beam power stop at thickness values of 1.15 mm and 1.04 mm, respectively. This is the point where the maximum temperature of the material reaches its limit of 400 °C for that beam power.

FIGURE 21 Calculated maximum stress and limit stress versus material thickness for standard grade beryllium at beam powers of 35, 37.5, and 40 kW. The calculated stress is a combination of stress due to thermal variation within the material, and stress due to the pressure differential. Limit stress is calculated based on the maximum temperature in the window. This plot is similar to FIGURE 1, where the strength of the material begins to diminish rapidly with increasing temperature.

The above results for the three candidate materials clearly indicate that the maraging steel and beryllium perform better than Inconel in their ability to handle the most power from the electron beam while withstanding the difference pressure between the helium coolant and the vacuum of the beam tube. The materials and their calculated maximum power ratings are summarized in Table 3. The maraging steel and beryllium outperform the Inconel by more than a factor of two. Although the maraging steel shows a slightly larger limit on maximum beam power, multiple factors make both beryllium and the maraging steel worth considering as window materials. As an example, beryllium has an advantage with respect to activation.

TABLE 3 Summary of the results of the parametric study.

Material	Maximum Beam Power (kW)
Inconel 718	18
Beryllium	40
250 Maraging Steel	39

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