

ENGINEERING DESIGN OF GALLIUM-NICKEL TARGET IN NIOBIUM CAPSULE, WITH A MAJOR FOCUS ON DETERMINING THE THERMAL PROPERTIES OF GALLIUM-NICKEL THROUGH THERMAL TESTING AND FEA, FOR IRRADIATION AT BLIP*

S.K. Nayak[†], S. Bellavia, H. Chelminski, C. S. Cutler, D. Kim, D. Medvedev,
Brookhaven National Laboratory, Upton, NY, USA

Abstract

The Brookhaven Linac Isotope Producer (BLIP) produces several radioisotopes using a variable energy and current proton beam. The targets irradiated at BLIP are cooled by water and required to be isolated in a target capsule. During the design stage, thermal analysis of the target and cladding is carried out to determine the maximum beam power a target can handle during irradiation without destruction.

In this work we designed a capsule for Gallium-Nickel (Ga 80%, Ni 20%) alloy target material and irradiated the target at the BLIP to produce the radioisotope $Ge-68$. Since no literature data is available on $Ga_{4}Ni$'s thermal conductivity (K) and specific heat (C), measurements were carried out using thermal testing in conjunction with Finite Element Analysis (FEA). Steady-state one dimensional heat conduction method was used to determine the thermal conductivity. Transient method was used to calculate the specific heat. The test setup with same methodologies can be used to assess other targets in the future. Here, we will detail these studies and discuss the improved design and fabrication of this target.

INTRODUCTION

The BLIP produces a variety of radioactive isotopes by striking proton beams on different target materials. The target material, sealed in a container further referred as capsule, when subjected to proton beam absorb proton energy most of it in a form of heat. To limit the temperature rise of the capsule, target irradiation is carried out by keeping the capsule cool by continuously flowing water along its faces [1]. After a detailed thermal analysis of the capsule, suitable beam power is determined for irradiation so that structural integrity of the capsule is maintained, and irradiated target material is not exposed to water. Any structural failure of capsule will lead to the release of radioactivity to the water tank, loss of radioisotope and may affect operation time.

BLIP traditionally irradiated pure Ga metal targets to produce $Ge-68$ radioisotope, but targets' survivability in the beam was poor [2] and $Ge-68$ yields were inconsistent. We designed a capsule for $Ga_{4}Ni$ alloy target material (ACI Alloys, Inc., CA, USA) and irradiated at BLIP to produce the radioisotope $Ge-68$. Precise knowledge of thermal conductivity of the target material is key in the steady-state

*This paper is authored by BSA operated under contract number DE-SC0012704. This research is supported by the U.S. DOE Isotope Program, managed by the Office of Science for Nuclear Physics.

[†]snayak@bnl.gov

thermal analysis which yields optimum irradiation parameters. Since no literature data is available on $Ga_{4}Ni$'s thermal conductivity value, thermal testing was carried out to calculate the same. Using the same test and FEA, its specific heat was also estimated. Specific heat will be required if there is a need in the future to run transient thermal analysis.

DESIGN

Gallium attacks most common metals such as Aluminum and Stainless Steel (SS) by diffusing into the metal lattice and making them brittle [3]. Niobium which is more resistant is one of the best choices of metal for this application and was used to make the capsule [2, 3]. It has good resistance to Ga attack up to 400 °C. In the initial design, the $Ga_{4}Ni$ target was enclosed by a ring and two thin (0.012") windows (Fig. 1, upper drawing). The overall dimension of the capsule is 2.750" diameter and 0.220" thick. The windows were laser welded to the ring in an environment of helium gas. This design is typical to most isotope production at BLIP due to simplicity of capsule manufacturing and welding.

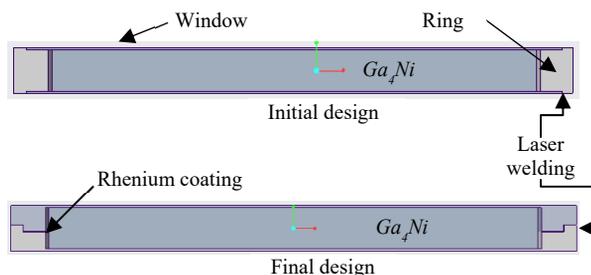


Figure 1: $Ga_{4}Ni$ target in Niobium capsule.

During test irradiation (about 33 MeV proton energy at 160 μA , i.e 5294 W of beam power), the welding joint between the window and the ring failed at a very small section. Due to this failure, the irradiated $Ga_{4}Ni$ got exposed to flowing cooling water along the capsule face meant for cooling and washed away. The reason for the failure could be one or more of the following; at high temperature Ga reacted with Niobium, welding joint failed at high temperature.

In the next generation design, the capsule was made of two halves (Fig. 1, lower drawing) where the thin window was a part of the ring rather than welded to the ring. This design has been used for most of the targets irradiated at Isotope Production Facility at Los Alamos National Laboratory (Los Alamos, NM, USA) [4]. The welding joint at the rim produces a high penetration stronger welding joint than in the initial design. The mating area of two halves provide

a much longer path between the target material and the weld, thus reducing the chance of target material reaching welding joint. In addition, the inner surface of capsule was Rhenium coated (Plasma Processes, AL, USA), which provides good resistance to *Ga* attack up to 775 °C [3] and protects the Niobium. The machining of such a thin (0.012”) window in each half of the capsule was very difficult as it has a tendency to gall, tear and weld to the face of the cutting tool. After a few trials, high speed steel tools with a good amount of water-soluble cutting oil for lubrication were used for the milling operation. Rhenium coating was challenging to control the coating thickness as the windows were not flat, but rather bowed out due to machining stress. After coating, a minor thickness adjustment to *Ga*/*Ni* puck was done such that the two halves of capsule fit well for welding.

EXPERIMENTAL SET UP FOR THERMAL CONDUCTIVITY AND SPECIFIC HEAT MEASUREMENTS

Methods

There are a variety of measurement techniques available to determine the thermal conductivity (*K*) and specific heat (*C*), most were ruled out by considering its measurement range [5]. Here, traditional SS method was used to derive the *K* and subsequently using *K* and transient method, *C* was determined.

In the SS method, *K* (Eq. 1) was derived by measuring the temperature difference ΔT at a separation ΔL under the SS heat flow Q [6, 7].

$$K = \frac{Q}{A \times \frac{\Delta T}{\Delta L}} \quad (1)$$

In the transient method, rate of cooling was measured and through FEA, the *C* was estimated.

Test

To eliminate the convection heat loss, the test was carried out in vacuum. The test set up (Fig. 2) consists of a

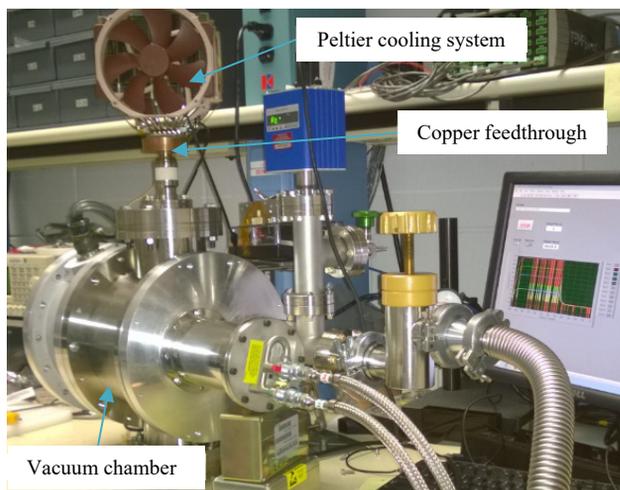


Figure 2: Test setup.

vacuum chamber connected to a roughing pump, a high-power copper feedthrough which holds the test piece and conduct heat to a heat sink, and Peltier cooling system to maintain constant sink temperature.

Figure 3 shows the preparation in detail for the test. Longer length and smaller diameter test piece are ideal for this type of one-dimensional conduction test as it gives higher thermal gradient and improve the measurement accuracy by reducing the effect of errors in temperature measurement. The best size the vendor could make is 1/2” diameter and 1.2” long *Ga*/*Ni* rod.

Ultramic advanced ceramic heater housed in a very low thermal conductive Macor was used. Macor minimizes the heat loss from the heater. Both ends of the test piece were glued to the copper and heater/Macor by high temperature thermal conductive epoxy-based adhesive. RTDs, which have higher accuracy and repeatability were used for the temperature measurement. To minimize the contact resistance, RTDs were attached to a test piece by silver conductive paste in-between and held firmly by Kapton tape. At the heat sink surface of copper feedthrough, Peltier cooling system was used to maintain 22 °C throughout the test. The temperatures were logged with time. The vacuum pressure level in the chamber is maintained at 10⁻⁴ torr by a roughing pump.

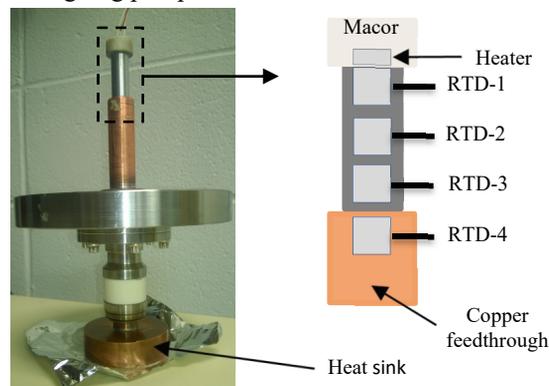


Figure 3: *Ga*/*Ni* test piece preparation for the test.

RESULTS

Figure 4 shows the temperature plots of *Ga*/*Ni* test at different locations (Fig. 3). As constant heater power (11 V and 1.63 Amps) was supplied, temperatures went up, then achieved SS. Once heater power was switched off, temperatures dropped to room temperature. Heat sink was always maintained at room temperature (22 °C). SS temperatures values were used to calculate *K* and cooldown plots in conjunction FEA were used to estimate *C*.

Calculation of thermal conductivity, *K*

The accuracy of *K* depends upon the accuracy of the *Q* value (Eq. 1). *Q* can be determined from supplied power to the heater and the radiation heat loss. Radiation heat loss is proportional to test specimen size and the test temperature.

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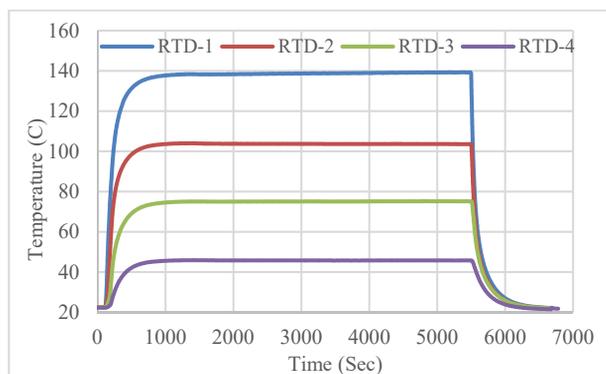


Figure 4: Temperature plots from Ga_4Ni test.

To determine the radiation heat loss during Ga_4Ni test, an identical test was carried out on low carbon steel (LCS) test specimen (same size of Ga_4Ni), whose thermal conductivity (51.9 W/m-C) is in close vicinity of Ga_4Ni . Heat loss was calculated to be 16.78 %. Since the temperature plots of LCS and Ga_4Ni were very similar, it was assumed that the same amount of heat loss would occur in both tests though it would be a little higher in Ga_4Ni considering the higher temperature values.

Table 1 illustrates the calculation of thermal conductivity (K) (Eq. 1) using SS temperature data, spacing between RTDs and net Q considering the heat loss.

Table 1: Thermal Conductivity Calculation From Ga_4Ni Test

Steady state power, $V = 11V, I = 1.629Amps$					
RTD	Position from heater end (m)	Temp ($^{\circ}C$)	% of heat loss from LCS test	K value (W/m- $^{\circ}C$)	Average K value (W/m- $^{\circ}C$)
1	0.005486	139.24	16.78	-	36.73
2	0.016256	103.61		35.58	
3	0.0254	75.188		37.87	

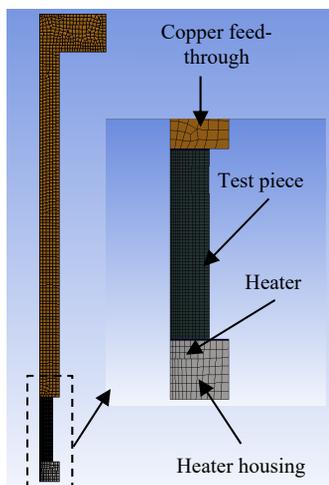


Figure 5: Axisymmetric finite element model of the test.

Estimation of specific heat, C

FEA was carried using ANSYS Workbench to recreate the experimental results. Finite element model is shown in Figure 5. It was done in two steps. First, the SS conditions were created through SS simulations and then its temperature results were used as initial condition in transient analysis to create the cool down plots. The SS simulation need only K whereas transient simulation needs K, C and density. In transient simulation different C values are tried until the cooldown plot matches to the experimental result.

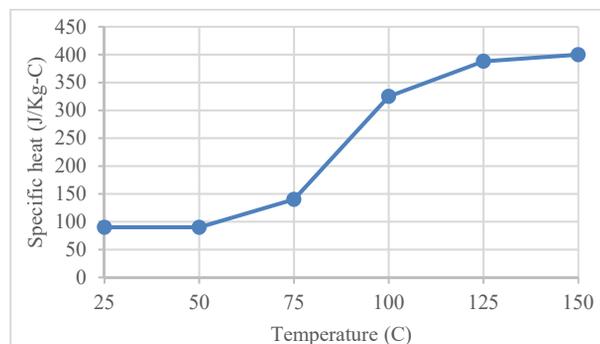


Figure 6: Specific heat, C of Ga_4Ni obtained from the test and FEA.

To validate the FEA, this analysis was done on the LCS test and then on Ga_4Ni . The cooldown plots created in FEA using LCS's known K, C and density values match well to the experimental results. The same methodology was applied to determine C of Ga_4Ni .

It was found that specific heat of Ga_4Ni varies substantially with temperature. After several runs of FEA, the temperature dependent C was determined (Fig. 6). The specific heat increases with temperature and reaches a plateau at about 150 $^{\circ}C$.

SUMMARY

The one-dimensional steady state thermal conduction test was successfully carried out on identical test specimens, low carbon steel (LCS) and Ga_4Ni . The Ga_4Ni 's thermal conductivity, K was determined to be 36.73 W/m-C. Specific heat values were established (Fig. 6) after several FEA runs. To evaluate C beyond 150 $^{\circ}C$, heater power needed to be increased to achieve higher SS temperature.

There are no published results to compare the findings from this test, however results determined from thermal test, FEA and through validation of a material with known thermal properties indicates that it should be very accurate. To improve the accuracy of the result, longer and smaller diameter specimen is desirable where more RTDs can be mounted, but such Ga_4Ni test piece could not be easily fabricated. Higher number of temperature data points obtained from RTDs would improve the accuracy of results. This test setup can be used to assess other targets in the future.

The Ga_4Ni target capsule was successfully designed and fabricated. The K value obtained from this test was used to run FEA to determine the maximum beam power this target

capsule can handle during irradiation without destruction. The target was successfully irradiated at 3147 W beam power for 11 days.

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