

MATERIALS UNDER IRRADIATION BY HEAVY IONS AND PERSPECTIVES FOR FRIB*

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Abstract

High energy heavy ion beams that are planned for the Facility for Rare Ion Beams (FRIB) will deliver power at very high densities and will produce significant radiation damage in materials with which they interact. Reliable predictions of material and component life times for FRIB are needed, yet the tools used to make the necessary predictions, for example heavy ion radiation transport codes, provide damage estimates whose levels have in the past varied significantly. In addition, there are very few appropriate data sets to validate code predictions. We will present examples of components, for example the beam dump system for FRIB, with attending predicted levels of damage obtained by radiation transport codes. We will summarize results from an experiment to produce and to quantify damage in a controlled way. Finally, we will show examples of targets used in experiments at the National Superconducting Cyclotron Laboratory (NSCL) where damage has been observed, and will present results from transport codes to quantify the damage.

INTRODUCTION

Michigan State University has prepared a conceptual design for a U.S. Department of Energy (DOE) Office of Science National User Facility for scientific research with rare isotope beams. This facility [1], the "Facility for Rare Ion Beams" (FRIB), will provide intense beams of rare isotopes to be used for cutting edge nuclear science research. The rare isotope beams will be created from intense beams of stable isotopes accelerated in a superconducting-radio-frequency linear accelerator to kinetic energies above 200 MeV/nucleon for all ions including uranium with beam power up to 400 kW. There are significant technical challenges associated with the high-power density caused by the interaction of the high-power primary heavy ion beam with matter, and with the high radiation levels associated with the nuclear interactions.

The systems most strongly affected by these challenges are the rare isotope production target, the primary beam dump, and various magnet systems. Research and development (R&D) is being performed to develop viable technical solutions. Even within previous MSU-led R&D efforts [2], it was recognized that radiation damage by high power heavy ion beams interacting with target and beam dump materials will be significant. It was also recognized that there is scant experimental information available at power and energy appropriate for FRIB. Attempts were made to use existing radiation transport

codes to predict levels of damage in the developed beam dump concept, a rotating water-filled aluminum shell. Stein et al. [3] estimated the damage using the PHITS [4] code system version available at that time, that for a 320 MeV/nucleon ²³⁸U beam having 366 kW (3e13 ions/s) passing through a 1 – 2 mm aluminum shell over a 5 cm x 220 cm area (in the case of rotation for an approximately 70 cm diameter drum) the resulting radiation damage is approximately 7e-2 dpa/day. The term "dpa" stands for displacements per atom. In metallic structures, displaced atoms result in often undesirable property changes, such as swelling and embrittlement. If the allowable dose is 5 dpa, this could be reached in about 10 weeks if the beam position on the dump is unchanged.

Currently available data suggest that the displacement damage caused by energetic heavy ions has a significant contribution from electronic stopping of the beam particles, and this contribution can be orders of magnitude larger than the damage caused by nuclear stopping. This "swift heavy-ion effect" has a strong dependence on the projectile energy. The relation of actual material damage from heavy ion radiation to dpa values calculated with commonly available transport codes is practically unknown. It is very important to FRIB design efforts to better understand heavy ion radiation damage mechanisms and to improve models and predictability.

PERSPECTIVES FOR FRIB

The preferred concept for a beam dump for FRIB at present is a water-filled rotating aluminium-shell system having approximately 70 cm diameter and approximately 1.5 mm shell thickness. This concept is shown in Figure 1. Damage predictions (in terms of dpa) were carried out for 1.5 mm aluminium using TRIM [5]. The TRIM code was chosen because it predicted higher values of dpa compared to older versions of MARS15 [6] and PHITS [4]. The representative heaviest ion beam was approximately 200 MeV/nucleon ²³⁸U. The representative "light" heavy ion was approximately 190 MeV/nucleon ⁴⁸Ca. The results are summarized in Table 1. Drum rotation and variation of beam position on the dump as a function of beam-target-rare isotope combinations that are expected during operations increase the lifetime. In addition, a mix of light and heavy ion beams is expected to be required to satisfy the science needs. Overall, the beam dump life is expected to exceed a year if our assumptions and code predictions of damage are reasonable. However, if radiation damage estimates are a factor of 10 too low, dump lifetimes of several months to several years can still be expected, depending on facility operation.

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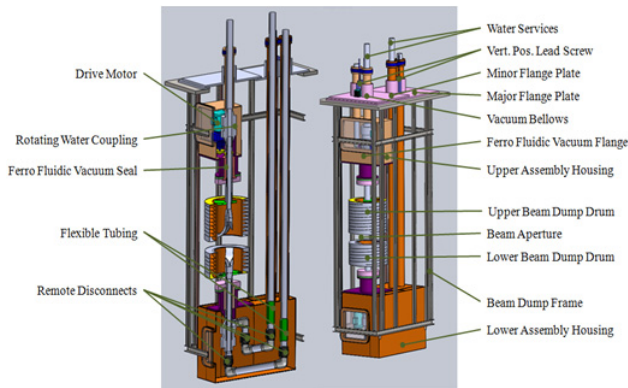


Figure 1: Locations and descriptors of the main mechanical components of the rotating water-cooled beam dump concept. The left panel shows a cut view of the assembly. The right panel is slightly rotated and shows transparent upper and lower assembly housing panels.

Table 1: Summary of damage and lifetime predictions using TRIM for the FRIB beam dump concept's aluminium shell

Beam	Effective Irradiation Area	DPA Rate (s ⁻¹)	Predicted Lifetime
238U ~ 200 MeV/nucleon	4 cm x 0.16 cm	4 e -4	7 hours if static and the beam is on the same spot
238U ~ 200 MeV/nucleon	8 cm x 70 π cm Increased by rotation, variation of beam position	1.5 e-7	Approximately 2 years
48Ca ~ 190 MeV/nucleon	0.5 cm x 70 π cm Increased by rotation	4e-10	Life of facility

RESULTS FROM RADIATION DAMAGE EXPERIMENT AT NSCL

A radiation damage experiment was carried out at NSCL [7] using a stack of 30 aluminum foils each 0.25

mm thick. The stack was designed to stop the 122 MeV/nucleon ⁷⁶Ge beam (stopping range is about 4.8 mm). Prior to the experiment the foils were annealed in a vacuum furnace. The sample is shown in Figure 2. The stack was cooled with chilled air directed at the sample in an attempt to keep the temperature below 100° C for a maximum of 5 W beam power.

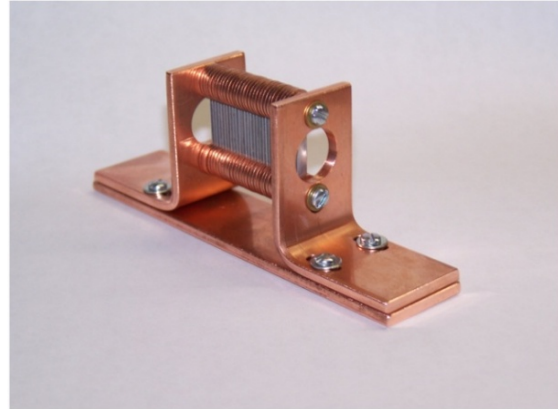


Figure 2: Air-cooled stack of 30 aluminum foils mounted in copper sample holder.

Transmission electron microscopy (TEM) measurements were then carried out at Low Activation Materials Design and Analysis (LAMDA) facility at ORNL. The images of foil 2, 4, 8, 14, 17, and 20 are shown in Figure 3. The significant concentration of dislocation loops observed by TEM in foil 2 indicates that the high energy ion beam did have a considerable effect in generating displacement damage through electronic stopping. However, the number of dislocations appeared to fall sharply with depth, in contradiction with code predictions. While displacement damage was observed through the development of dislocation loops in all the irradiation samples examined, the network of dislocation lines, tangles, and subgrain boundaries dominate the microstructure. Therefore, the level of defect damage generated during irradiation was not enough to show up over the statistical averaged values of electrical resistivity and hardness of the as-annealed samples. It is important to improve on such heavy ion beam-induced radiation damage experiments with a goal to induce enough damage so that one can tie bulk properties to known levels of displacement damage.

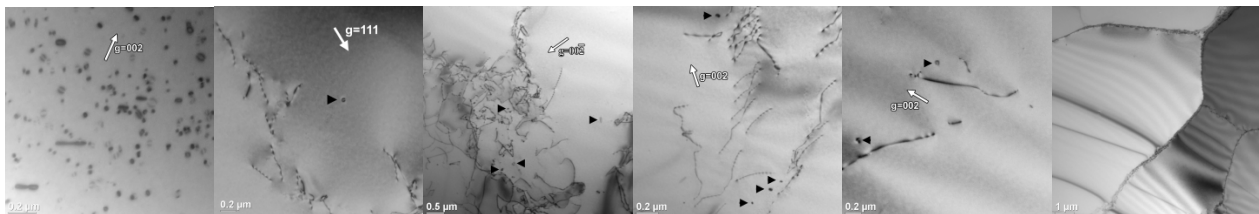


Figure 3: TEM images of irradiated aluminium sample number 2, 4, 8, 14, 17, and 20 (left to right, respectively). Black arrows help to indicate locations of radiation induced dislocation loops. Foil 2 is the most upstream foil analyzed and shows many radiation induced dislocation loops. Foil 14 is nearer the stopping depth and shows few loops. Sample 20 was beyond the stopping depth of the ion beam and revealed no radiation induced defects.

DAMAGE OF RARE ISOTOPE BEAM PRODUCTION TARGETS AT NSCL

In an effort to prevent damage of rare isotope beam production targets the NSCL Coupled Cyclotron Facility Beam Delivery Group routinely performs a priori thermal calculations for targets designated for use in scheduled experiments. Nevertheless, damage of targets has been observed even when levels of power delivered to these targets were significantly below those where melting could be expected.

Damage to a Tungsten Target

During a target change it was noticed that a tungsten target (580 mg/cm^2) used with experiment 08024 has a visible, crater-like surface modification where it was traversed by a $0.6 - 0.8 \text{ mm}$ -diameter 130 MeV/u ^{76}Ge beam (see Figure 4). Initially it was speculated that the target temperature exceeded melting temperature of tungsten (3410°C). However, the power deposition in the target was 88 W and thermal calculations suggested that melting temperature would be reached for power deposition exceeding 250 W (assuming a 1 mm diameter beam spot size).

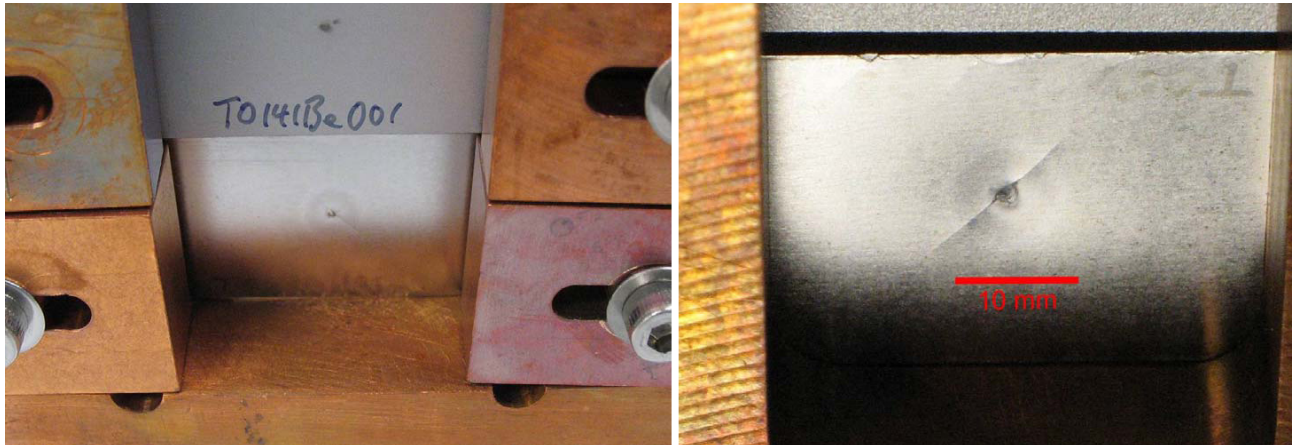


Figure 4: 580 mg/cm^2 tungsten target damaged by the 88 W , 130 MeV/u ^{76}Ge beam. The left panel shows the target (front view) in its position as the lower-most target in a water-cooled copper target ladder. The right panel shows the back view of the target, with a crater-like area around the beam spot. An approximate scale is also shown.

Using the target thickness and density, beam spot size, and the energy deposition and the total fluence of beam ions (5.77×10^{16}) that impinged on the target, a total absorbed dose was calculated to be approximately $7.9 \times 10^{12} \text{ Gy}$. Based on the collected data, it appears that the target was not damaged due to overheating and melting of the target but rather radiation damage induce swelling and embrittlement, leading to the observed crack. Another mechanism that may have contributed is melt layer erosion. In this process, the radiation induced defects of the metal lattice reduce the thermal conductivity, and thereby enable local melting of the tungsten material. Due to the thermal tension in the material a small crater can form, such as observed.

Calculations of dpa for the Tungsten Target

A calculation of damage using TRIM, based on the beam isotope, beam energy, target material and target thickness, indicates roughly 9700 displacements per beam ion. Factoring in the total number of beam ions and number of target atoms in the irradiated volume yields 74 dpa . However, the effects of target damage were noticed in particle-identification spectra when 51 dpa were accumulated. Calculations of dpa using then-available versions of the radiation transport codes MARS15 and PHITS provided 2.83 and 0.92 dpa respectively. Both

values are significantly below that obtained using TRIM (TRIM:MARS15 = 1:0.04, TRIM:PHITS = 1:0.01)

Very recently the dpa models within both MARS15 and PHITS have been significantly improved [8,9] by inclusion of and careful consideration of electromagnetic processes such as Coulomb scattering and electromagnetic showers [9,10]. Much better agreement between TRIM, MARS15, and PHITS is now reported [8,9] (TRIM:MARS15, TRIM:PHITS = 1:0.18, 1:0.21).

Damage to Beryllium Targets

Beryllium targets used in experiments 09030 and 09040 at NSCL were also found to be damaged even though precautions were made with respect to expectations based on power and absorbed dose. In these experiments a 140 MeV/u ^{48}Ca ion beam was used.

Targets Used in Experiment 09030

Two targets were used, “Be 1269 a” and “Be 1269 b”. Each consisted of a sandwich of two pieces of Be (approximately 1175 mg/cm^2 and approximately 94 mg/cm^2). The effective thicknesses of these targets were determined to be 1273.8 mg/cm^2 (Be 1269 a) and 1277.9 mg/cm^2 (Be 1269 b) by measuring energy losses of the incident beam. The uncertainty in the energy loss measurement is about 0.02% . Each target received a similar dose, about $4.5 \times 10^{12} \text{ Gy}$.

Targets Used in Experiment 09040

Two targets were used, “Be 1316 a” and “Be 1316 b”. Each consisted of a sandwich of two pieces of Be (approximately 846 mg/cm² and approximately 470 mg/cm²). The effective thicknesses of these targets were determined to be 1340.6 mg/cm² (Be 1316 a) and 1341.1 mg/cm² (Be 1316 b) by measuring energy losses of the incident beam. The uncertainty in the energy loss measurement is about 0.02%. Each target received a similar dose, about 4.5e12 Gy.

Observations of Target Damage

The radiation damage of the targets was manifested by an increased energy loss in the target at the location of beam spot and by increased energy straggling. By adjusting the target position so that the beam impinged either above or below the nominal position, it was found surrounding areas were not affected. An example of the beam images at the dispersive mid-plane of the A1900 fragment separator, at the beginning of experiment 09040 and after an accumulated dose of 7.6e12 Gy is shown in Figure 5.

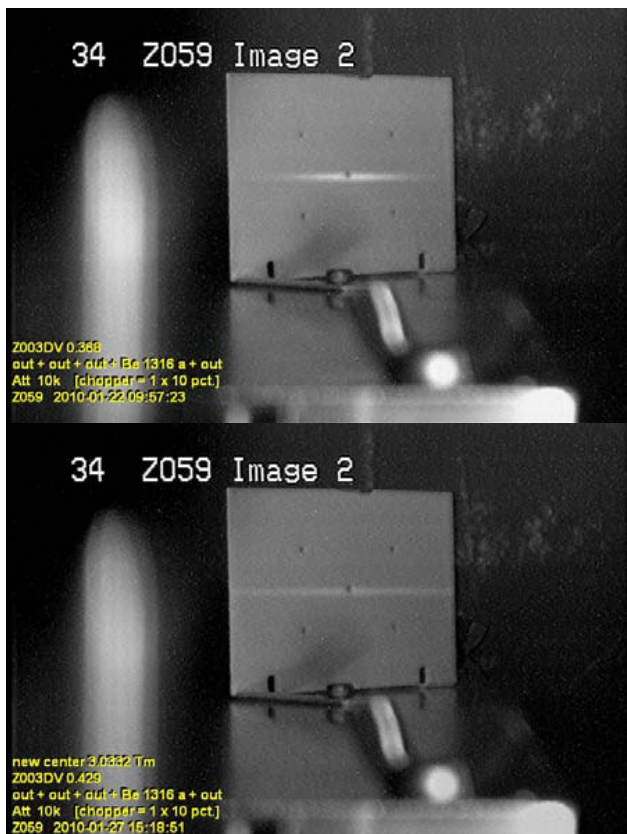


Figure 5: Images of the beam going through target Be 1316 a on the viewer at the dispersive mid-plane of the A1900. The upper image was taken at the beginning of the experiment; the lower image shows evidence of target thickness and energy straggling increase due to radiation damage after receiving 7.6e12 Gy.

In experiment 09030 the effective target thickness was measured once per day, and the production target was

swapped roughly at the midpoint of the experiment. The two targets, Be 1269 a and b, received a similar total dose of roughly 4.5e12 Gy. The measured increases in energy loss corresponded to a thickness increase of 2.3% (Be 1269 a) and 1.2% (Be 1269 b). The two targets responded differently to a similar dose.

In experiment 09040 the effective target thickness for target Be 1316 a was measured about five days into the experiment, at about the midpoint of the experiment. By this time, that target had received a dose of 7.6e12 Gy, and a significant increase in energy loss was observed. The measured effective thickness increased by 3.9%.

For the second half of the experiment, the second target was used and the effective thickness was measured once per day. Each subsequent measurement showed an increased effective thickness. The second target received a total dose of 7.5e12 Gy, but the increase in thickness amounts to only 1.2%.

Figure 6 shows a summary of the measured effective target thicknesses of the four targets. It is interesting to note that targets (a) were mounted higher in the target ladder than targets (b). The higher ladder position is further away from the water-cooled base of the ladder.

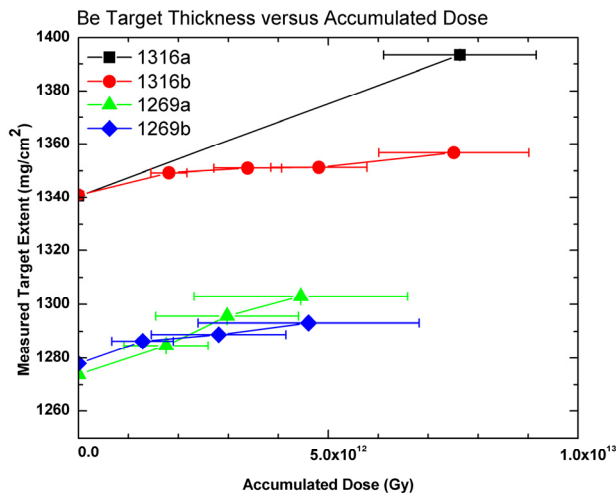


Figure 6: Measured effective target thickness for each Be target versus accumulated radiation dose.

Calculations of dpa for a Be Target

A calculation of damage [9] using TRIM was performed for the experimental conditions of 09040 and target Be 1316 b. The calculation yields 0.31 dpa. A calculation of dpa [9] using PHITS yields 0.24 dpa respectively, in very good agreement with TRIM.

SUMMARY

New high energy heavy ion beam facilities currently planned or being established, for example FRIB, will encounter significant levels of radiation damage to materials exposed to beam ions. Even at a currently operating lower-power heavy-ion-beam rare-isotope production facility, in this case NSCL, damaged rare isotope production targets have been experienced in spite

of planning efforts to avoid this. Not only do thermal properties of materials change with radiation damage but so do other fundamental properties necessary for experiment planning such as target thickness. It is thus becoming increasingly important for design and planning efforts that levels of damage can be predicted. Very recently heavy ion transport codes have made significant progress to improve models used to predict radiation damage, such as via levels of dpa. These codes currently appear to agree very well with each other. However, how the predicted levels of dpa relate to actual levels of dpa created by heavy ion beams, and to changes in material bulk properties, are still open questions. Benchmark experiments, or even heavy ion induced damaged materials (for example, targets) where parameters such as ion energy, fluence, power density, material temperature were collected, would be extremely valuable for validation purposes.

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