High Power ISOL Target Development - R&D Report*

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R&D Category: ISOL Systems

Abstract

For the Rare Isotope Accelerator (RIA) facility, ISOL targets employing refractory compounds of uranium are being developed to produce radioactive ions for post-acceleration. In our two-step target design, a neutron generator induces fission in a surrounding assembly of uranium carbide. The availability of refractory uranium compounds in forms that have good thermal conductivity, relatively high density, and adequate release properties for short-lived isotopes remains an important issue.

Investigations using commercially obtained uranium carbide material and prepared into targets involving various binder materials have been carried out at ANL. Thin sample pellets have been produced for measurements of thermal conductivity using a new method based on electron bombardment with the thermal radiation observed using a two-color optical pyrometer. These measurements have been performed on uranium carbide powder samples as a function of grain size, pressing pressure and sintering temperature. Comparisons of the data with literature values are presented. Additional uranium carbide powder has been prepared by the ANL Energy Technology Division and also manufactured at ANL-West. In addition, thermal modeling of uranium carbide target disks with modest densities and thermal conductivities has been carried out at ANL and also by W. Talbert of Tech Source. Talbert and collaborators at Tech Source have also performed simulations of the thermal behavior of the secondary target assembly incorporating various heat shield configurations.

Sets of sample pellets using various binders have been forwarded to Oak Ridge National Laboratory for independent thermal diffusivity analysis at ORNL. Measurements of fission release properties for these samples are being carried out at the UNISOR facility at HRIBF by D. Stracener. Collaborative efforts are also underway with G.D. Alton of ORNL involving high power carbide targets formed using the painting technique.

Finally, we are presently investigating spray coatings along with the possibility of employing Atomic Layer Deposition in collaboration with the ANL Materials Science Division as a uranium compound coating technique for thin metallic foils in the construction of a “stacked foil” target for possible in-beam testing at high power at TRIUMF.

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1. Introduction

Production targets proposed for RIA will be based on fission targets employing the compounds of uranium to produce ion species far from stability [1]. In our two-step target design (Figure 1), neutrons are first generated in a cooled, refractory, primary target which then induce fission in a surrounding assembly of uranium carbide.

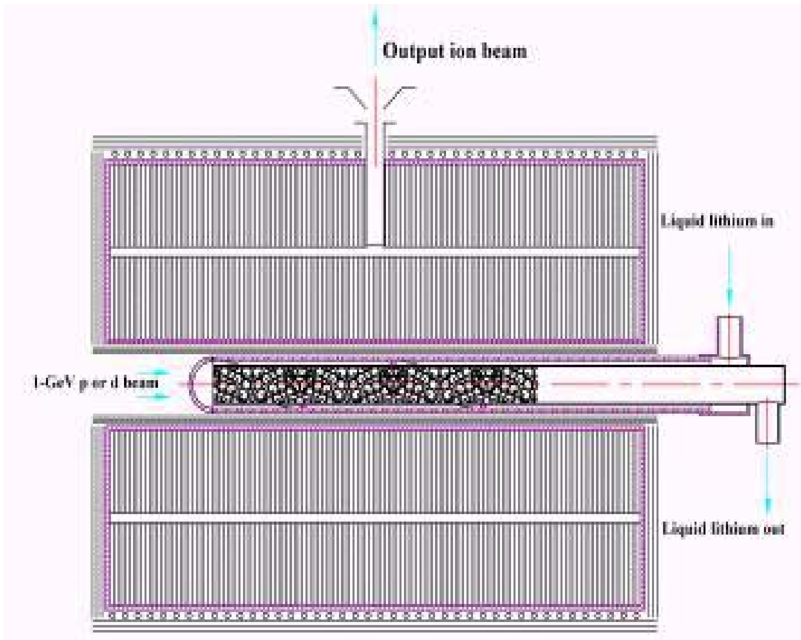

Figure 1. Two-step RIA production target design.

The prototype target design is being developed at TechSource, Inc. [2] and the fine-grained, high thermal conductivity UC_2 is being prepared at ANL.

2. Current Target Design

The primary target is a liquid lithium cooled tungsten foam cylinder, irradiated by the RIA driver beam. The secondary target consists of a series of spaced, annular disks of pressed uranium carbide with an outer dimension of 6 cm and inner hole of 3 cm diameter. Thermal analysis of the two-step target concept performed with a realistic primary target, including coolant components resulted in 800 W of deposited energy (as compared to 20 kW in the neutron generator) for an incident proton beam of 500 MeV and intensity of 100 µA. Energy deposition and calculated fission rates determined that a density of, or greater than, 3.3 g/cm^3 would reduce the volume of the secondary target (UC_2) while keeping the fission rate constant.

3. Investigations of Uranium Carbide Properties

3.1 Fission Rates

Fission rates calculated for the secondary UC_2 target, using the prototype target material parameters. Total fission rates for UC_2 material were 0.0329 fissions/proton for 2.5 g/cm^3 density, 0.0698 fissions/proton for 5.0 g/cm^3 density, and 0.1077 fissions/proton for 7.5 g/cm^3 density [3]. Shown below is a plot of fissions/p for these three densities as a function of annular target disk (radial) thickness.

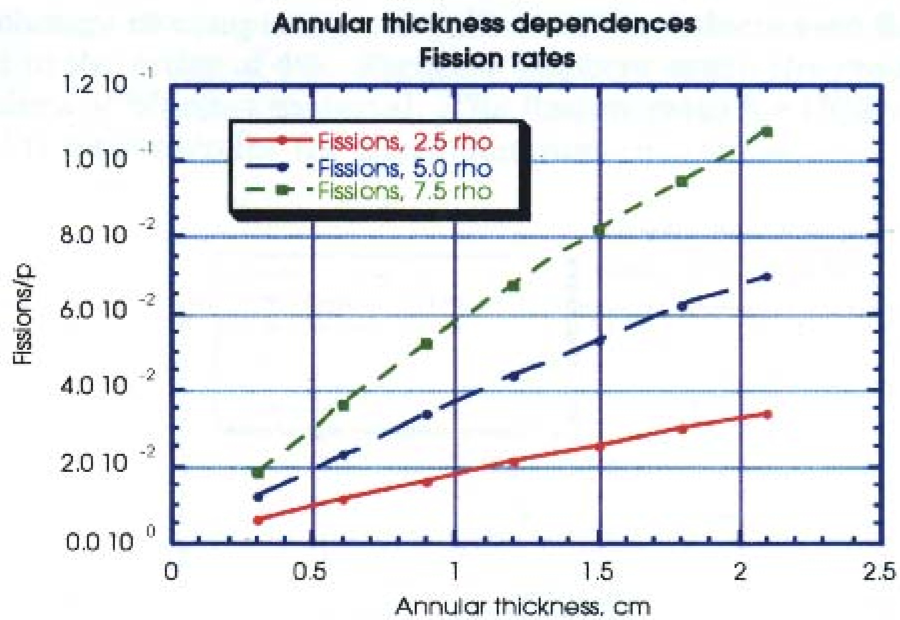

Figure 2. Fission rates as a function of annular target disk thickness.

4. Thermal Conductivity Measurements

4.1 Sample Preparation

The thermal conductivity measurement sample disks are prepared in a laboratory hood by first weighing out 0.32 g of UC_2 powder (-60 mesh) together with 40 mg of carbon (ratio of 8:1) in the form of high-purity synthetic graphite powder (-200 mesh). The weighed material is transferred to a 10 ml glass beaker where two drops of albumin are added as a binder and mixed thoroughly. Next, the mixture is poured into a 10 mm compaction die and pressed to 5 tons (10,000 psi) using a laboratory press.

4.2 Experimental Method and Recent Results

These thin sample pellets have their thermal conductivity measured using the method of electron bombardment recently developed at ANL [4]. The sample was heated on the bottom face by a vertical electron beam source installed within a vacuum evaporator. The sample was supported on a 95% transmission molybdenum mesh that was positioned central to the beam axis using a tantalum metal ring on top of a 6 in. glass tube where the beam spot size is approximately the size of the sample pellet. After achieving thermal equilibrium, the temperature of both faces of the sample were measured with the aid of a two-color pyrometer first set up to measure the bottom face of the sample then re-positioned above to view the top face. For a complete measurement, the experiment consisted of the sample being irradiated twice. Shown below is a typical thermal conductivity measurement, plotted as a function of temperature.

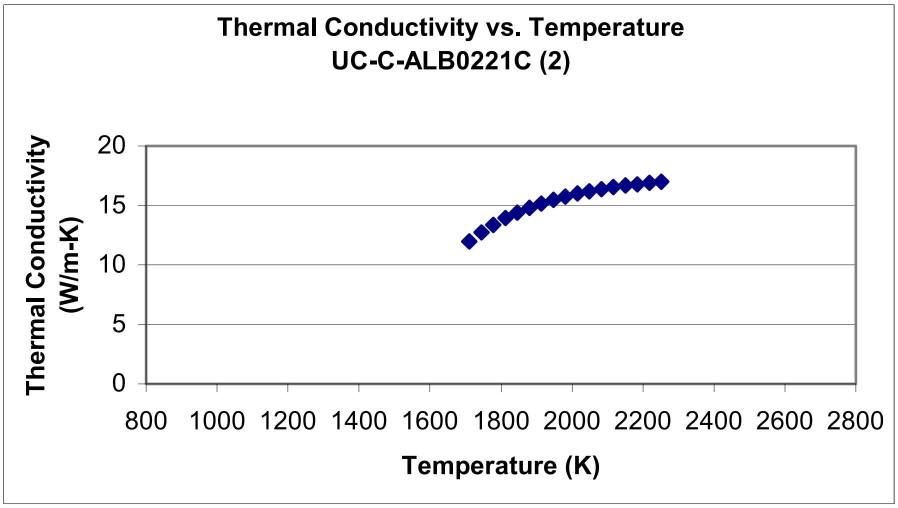

Figure 3. Thermal conductivity of a UC_2 target as a function of temperature

Several measurements of the thermal conductivity of small samples of uranium carbide materials have been carried out. The method is still being refined, checked for accuracy and overall degree of reproducibility. The results of earlier work [5] on reduced UC_X samples prepared by the ISOLDE prescription give densities on the order of 3 g/cm^3 with thermal conductivities of 1-2 W/m-K and emissivities of 0.7-0.8 in rough agreement with calculated thermal properties by Talbert [6]. Densities and thermal conductivities for the more recent UC_2 samples exhibit improved properties (k = 2-12 W/m-K), (ε = 0.4-0.5) and comparable to literature values [7,8]. These samples show great promise for use in the high-power applications needed for the RIA target. This work is still in progress.

4.3 Thermal Simulations

The small energy deposition rate in the surrounding UC_2 secondary target material is not sufficient to heat the target to the desired operating conditions necessary to promote effective release of fission products. To obtain a temperature range of 1600 to 2100°C prompted the introduction of heat shielding into the secondary target design, both between the primary target and the uranium carbide as well as outside the secondary target [9]. Figure 4 shows the maximum temperatures obtained using six external heat shields. The heat shields have been removed to show the surface temperatures of the target. Thermal conductivities on the order of 2 W/m-K (or greater) over the operating temperature range are required for a viable secondary target design containing 5 internal heat shields surrounding the primary target.

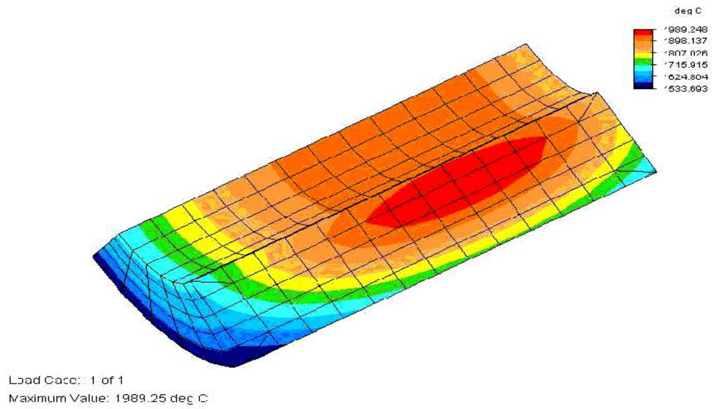

Figure 4. Temperature profiles for a target with six external heat shields.

4.4 Additional Analysis

When heated, changes may occur in the density, thermal conductivity and the microscopic structure of the sample pellets and is therefore being investigated for three sintering temperatures, 1100, 1500 and 2000°C. The sample disks were placed in a tantalum crucible and heated in a Type CH-10 crucible heater within a vacuum evaporator under a nominal pressure of a few 10^{-6} Torr. Currents of 300-400 A were applied as in a standard thermal evaporation with the temperature monitored using an optical pyrometer. After heating, the samples were observed using scanning electron microscopy (SEM).

Duplicate samples have been forwarded to the Thermophysical Properties Users Center at Oak Ridge National Laboratory (ORNL) for independent thermal conductivity determinations. Two sets of six sample pellets are being measured using their Laser Flash Thermal Diffusivity System. In this method, a short pulse of heat is applied to the front face of the pellet using a laser, with the temperature change of the rear face measured with an infrared detector. To determine the specific heat, a Differential Scanning Calorimeter is used to measure the thermal response of the UC_2 pellet as compared to a standard while heating uniformly at a constant rate. These measurements taken together are then used in the determination of the thermal conductivity.

4.5 Uranium Carbide Manufacture

We have begun in-house preparation (ANL-E & ANL-W) of this material by the method of arc melting uranium metal combined with an excess of solid carbon [10]. Sample preparation using various carbon and/or graphite starting materials is being investigated.

As grain size will prove to play an important role in ultimate densities and thermal conductivities achieved, this prepared material can be further characterized using sieves and new pellets pressed with enhanced properties. The desired release of the fission products produced under sample irradiation also needs to be explored as a function of density/grain size. Research into this aspect of our UC_2 samples as a step toward a RIA target is just now beginning.

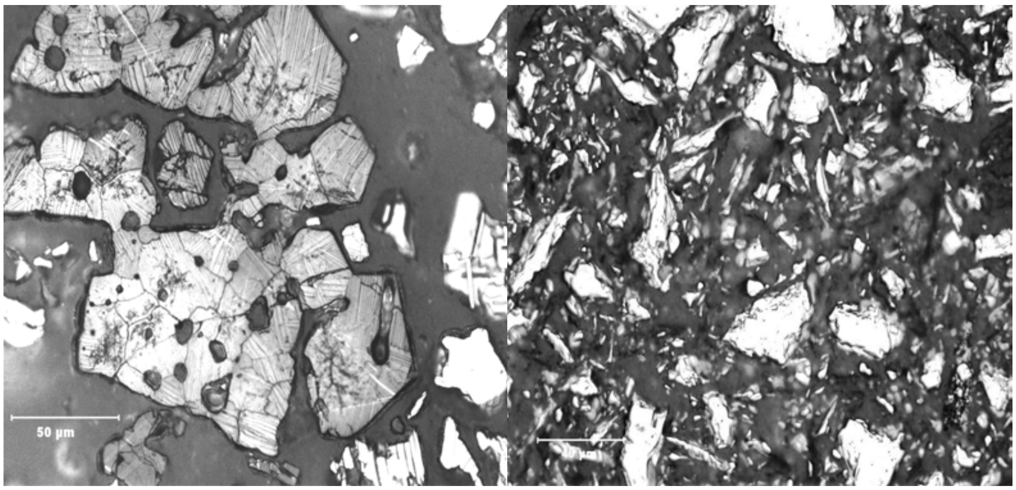

Figure 3. Micrographs (500x) of uranium carbide (Cerac material) -60 mesh (a) and ANL UC_2 powder, -325 mesh (b).

An initial run produced a 25 g ingot which was crushed under a nitrogen atmosphere using a tool steel mortar & pestle to yield 20 g of powder (-325 mesh), grain size 44 mm. A comparison photograph of this new material is shown in Figure 3. Investigations will begin employing samples prepared with this material.

5. Release Studies at UNISOR

The UNISOR facility at ORNL is used for decay studies of radioactive nuclei. The chemical selectivity capabilities of the target/ion source allow for characterization of "thick" target release properties. A second set of UC_2 pellets have recently been shipped to Oak Ridge for experiments at UNISOR. The previously shipped UC_x samples have now been baked out for several hours at 1950°C to remove contaminants. Measurements of the release of radioactive ions from these pellets will be performed in mid to late September after completion of the modifications to the UNISOR beam line.

6. Multi-Foil Target Approach

An alternate target approach to thin, pressed UC_2 disks is to stack a collection of refractory metal foils, such as Nb or Zr, regularly spaced within the target volume. This approach has been used successfully at ISOLDE and TRIUMF.

6.1 Spray Painting

In addition to using the properties of metal foils for ISOL targets, coatings with enhanced properties may be applied to the foils to improve production of rare ion species using various techniques. A common approach used for targets at ANL has been spray painting and was used recently for the preparation of MoS_2 targets for a GAMMASPHERE experiment at ATLAS [11]. Alton has employed a painting technique [12] for coating fiber targets and, with ANL, has begun a collaboration to provide uranium carbide for painting targets for use at HRIBF. Another technique being investigated is Electrospray or "powder coating" commonly used in decorative automotive finishes.

6.2 Atomic Layer Deposition

An exciting new prospect, working with the Material Science Division, is a new deposition technique employing Physical Vapor Deposition (PVD) onto metal (or foam) substrates using Atomic Layer Deposition (ALD). In this method monolayer films are grown on a substrate of choice using alternating reactive gas phases in a small furnace under computer control. In theory, the process can be applied to uranium compounds – UO_2 on W foils, but more promising might be uranium nitride (UN) coatings.

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