Report on the RIA Proposal - High Power ISOL Target Development*

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1. Introduction

The high power ISOL production target proposed for RIA will be based on fission from uranium or a compound of uranium to produce ion species far from stability [1]. The two-step target design employs neutrons first generated in a cooled, refractory, primary target which then induce fission in a surrounding assembly of uranium carbide. An engineering design of the ANL concept for the prototype target was done by TechSource, Inc. [2] with the fine-grained, high thermal conductivity UC_2 target material to be supplied by ANL. The primary target will be a liquid lithium cooled tungsten cylinder, irradiated by the RIA driver 1-GeV proton or deuteron beam. R&D on this aspect of the target assembly is firmly established using liquid Li metal cooling technology already employed for the fragmentation target and stripper films. The secondary target consists of a series of stacked, annular disk sections of pressed uranium carbide powder. Thermal analysis of a target geared towards a test at ISAC was performed by TechSource for primary and secondary target components for an incident proton beam of 500 MeV and intensity of 100 μA. A UC_2 density of 3.3 g/cm^3 is needed to keep the volume of the secondary target within design specifications. Fission rates have been calculated for the secondary UC_2 target and found to be 0.0329 fissions/proton for 2.5 g/cm^3 density and 0.0698 fissions/proton for 5.0 g/cm^3 density [3]. 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 [4]. 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. And so, the research covered by this proposal would be to manufacture and characterize the uranium carbide secondary target material, pressed into disks with densities greater than 3.3 g/cm^3 and thermal conductivities of at least 2 W/m-K in order to meet design criteria.**Uranium Carbide Manufacture**

As the original uranium carbide material used for this research is no longer commercially available, an effort was immediately mounted for in-house manufacture. Preparation has begun at ANL-E and ANL-W of this material by the method of arc melting uranium metal combined with an excess of solid carbon [5].

As grain size may 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. 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 microns. Sample powder was obtained from ANL-W with grain sizes of -325 and -400 mesh. A comparison photograph of this new material is shown in Fig. 1. Investigations are now underway employing samples prepared with this material.

3. Thermal Conductivity Measurements

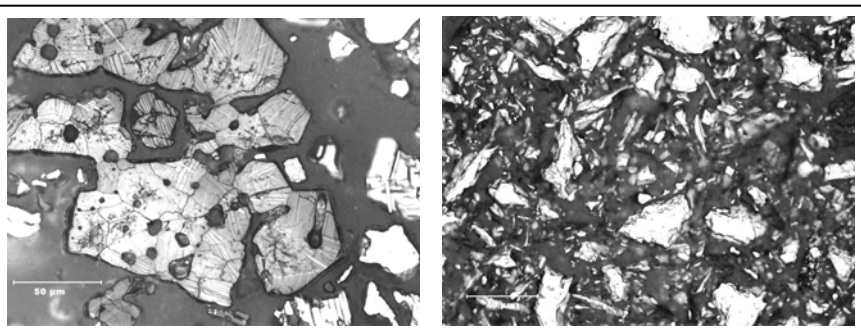

Figure 1. Uranium Carbide (Cerac material, left) 60 mesh and new UC_2 powder (-325 mesh, right).

The thermal conductivity measurement of sample disks are prepared in a laboratory hood by first weighing out UC_2 powder together with 40 mg of carbon (ratio of 8:1) in the form of high-purity synthetic graphite

powder. This synthetic graphite material is available with particle sizes of -200 and -325 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. The mixture is then poured into a 10 mm compaction die and pressed to 5 tons (10,000 psi) using a laboratory press. Densities of 5 g/cc and greater have been achieved, meeting design specifications.

These thin sample pellets then have their thermal conductivity measured using the method of electron bombardment recently developed at ANL [6], a simple extension of the technique employed by De Coninck for the determination of thermal diffusivity [7]. The sample was heated on the bottom face by a vertical electron beam source installed within a vacuum evaporator. After achieving thermal equilibrium, the temperature of both faces of the sample were measured with the aid of a two-color pyrometer.

Several measurements of the thermal conductivity for three samples of the new fine-grained uranium carbide powder have been carried out. The method is still being refined, checked for accuracy and overall degree of reproducibility. In Fig. 2 we give the results, with error bars for one of these sets of thermal conductivity measurements as a function of temperature. The earlier work [8] on reduced oxide samples prepared by the ISOLDE prescription gave densities of 3 g/cm3 with thermal conductivities of 1-2 W/m-K in rough agreement with calculated thermal properties by Drake and Talbert [9]. Densities and thermal conductivities for the more recent UC_2 samples exhibit improved properties, comparable to literature values [10,11].

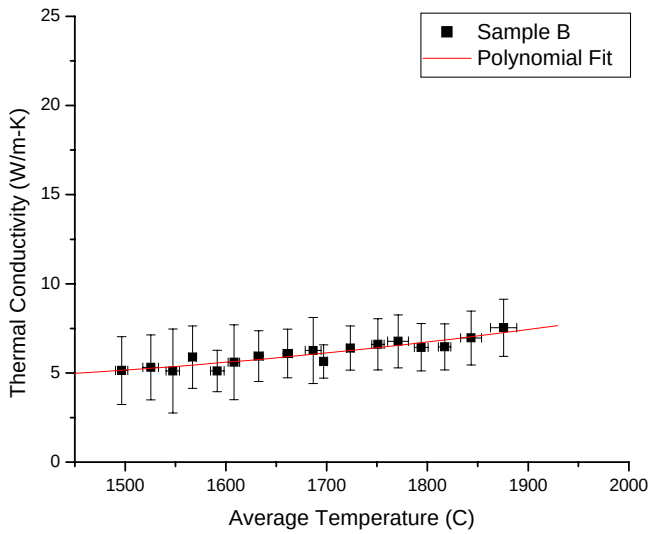

Figure 2. Plot of Thermal Conductivity vs. Temperature for one UC_2 pellet sample

4. Additional Thermal Analysis - Sintering

When heated, changes may occur in the density, thermal conductivity and the microscopic structure of the uranium carbide pressed powder samples and is being investigated for long term stability at elevated temperatures (2000 C). Access to a high-temperature vacuum furnace at ANL has been problematic. The availability of two furnaces at Argonne, one experiencing cooling difficulties after a high temperature test run, and the other developing a vacuum chamber leak has prompted the need for this baking to be carried out at ORNL. The acquisition of a high- temperature tungsten-heated vacuum furnace with a sufficiently large hot zone for acceptance of the actual production target disks is of paramount importance.

Duplicates of the above samples were forwarded to the Thermophysical Properties Users Center at Oak Ridge National Laboratory (ORNL) for independent thermal conductivity determinations. A set 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 time delay for 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 for the determination

of thermal conductivity. Recent thermal conductivity determinations (shown above) as well as new heat capacity calculations (150 J/mol-K) by Alton [12] should be confirmed by these measurements.

5. Release Studies at UNISOR

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 has become a high priority. The target/ion source of the UNISOR facility at Oak Ridge National Laboratory (ORNL) is being used for characterization of the ANL secondary target material release properties. We are working closely with Dan Stracener and Ken Carter and several sets of UC_2 pellets have been shipped to Oak Ridge for these measurements. Reduced oxide samples, sent earlier, have undergone a bake-out procedure at 1950 C to remove contaminants and have also been measured. An analysis of diffusion and effusion properties for this data was carried out by Antonio Villari [13]. Comparison of ORNL-RVC, reduced oxide and UC_2 commercial material supplied by ANL found mixed results as well as normalization inconsistencies in the data indicating different operating conditions in the target/ion source. This needs better understanding. Experimental studies of the release properties of radioactive ions from the newer, smaller grain size ANL material are to be performed soon after completion of the modifications to the UNISOR beam line.

6. Conclusion and Budget

With verification of the thermal properties coupled with new data from ORNL on the release properties of the fine-grain (-325 mesh) material, we hope to move towards fabricating actual secondary target disks using custom designed dies and a large area press. It is believed that the densities and thermal conductivity achieved for these UC_2 samples are sufficient for Talbert to proceed on the high-power RIA target design.

The allocation of $150K for this project during FY2004 was budgeted exclusively for the manpower from the ANL Physics, Chemical Technology, and Energy Technology Divisions. Five scientific staff from these divisions worked part time on this project to fabricate and characterize the material. The integral of the effort charged to this project was 0.6 FTE.

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7. References

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