2003-164-N0 -- DEVELOPMENT OF HIGH DENSITY, HIGH THERMAL CONDUCTIVITY URANIUM CARBIDE FOR EFFICIENT ISOL TARGETS

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Purpose: A high flux of neutrons can be generated by using a high-Z target such as tungsten irradiated by high-energy light ions such as protons or deuterons. The geometry for producing the most useful flux of secondary neutrons depends on the specific application. Producing the most neutron-rich fission fragments requires a compact coaxial geometry**.** Liquid-metal cooling of the core of this target is useful in keeping the assembly compact while thermally decoupling the primary and secondary target components. The high neutron multiplicity of tungsten, combined with its high density, make it a good choice for a compact primary target, while lithium is a good choice of coolant. With this geometry 10^{14} fissions per second are generated in the secondary uranium carbide target with 100 kW of protons or deuterons at 1-GeV beam energy. The fissions induced by secondary neutrons in the few-MeV energy range are the most effective in producing the most neutron-rich fission products. The optimal primary/secondary target geometries for producing fission products for elements outside the standard asymmetric fission mass regions will be different since such isotopes are produced better by fission induced by higher energy neutrons. The forward-peaked high-energy neutrons produced via the breakup of energetic deuteron beams on low-Z targets can be used for this purpose. At low deuteron beam energies the 2-step geometry has been used in ISOL (isotope separator on line) laboratories because fission yields are increased due to the larger useful uranium target volumes.

The simulations of the fission yields have assumed a uranium carbide density of 2.5 g/cm^3, ¼ of the full theoretical density of UC_2. This type of material has been used extensively at ISOLDE, the ISOL facility at CERN. It is porous and releases a large range of isotopes quickly. However, the thermal conductivity of this material has been measured recently, and it has been found to be extremely low. A preliminary thermal analysis of this geometry for the secondary target has been done and it shows that higher density and higher thermal conductivity are both essential for optimal performance of this target configuration. Concurrently, work at GANIL in France has shown that very fine grained, full density graphite is very efficient at releasing short-lived noble gas isotopes. The goal of the present work is to develop a very fine-grained, higher density form of uranium carbide for use in a model RIA target.

Approach: The goal was to fabricate samples at twice the density of the commonly used material and to send the samples to ORNL for release time measurements. Many parameters must be explored: the grain size of the starting ingredients, working directly with fine-grained uranium carbide versus the present method of firing a mixture of uranium oxide and graphite, and the temperature/pressure combination for the compacting process.

Technical Progress and Results: Investigations using commercially obtained uranium carbide material and prepared into targets involving various binder materials have been carried out. 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.

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 Oak Ridge by D. Stracener. Progress is summarized below.

Experimental Method and Recent Results. Thin sample pellets had their thermal conductivity measured using the method of electron bombardment recently developed at ANL. 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 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.

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 on reduced UC_x samples prepared by the ISOLDE prescription give densities less than 3 g/cm^3 with thermal conductivities (k) of 1-2 W/m-K and emissivities (ε) of 0.7-0.8. 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). These samples show great promise for use in the high-power applications needed for the RIA target.

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 requires 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. 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.

Additional Analysis. When heated, changes may occur in the density, thermal conductivity and the microscopic structure of the sample pellets and this is, therefore, being investigated for three sintering temperatures, 1100, 1500 and 2000 C. 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. These measurements at ORNL are still in progress with direct funding from the DOE Office of Nuclear Physics RIA R&D program**.**

Uranium Carbide Manufacture. We have begun in-house preparation of this material by the method of arc melting uranium metal combined with an excess of solid carbon. 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 will continue under direct funding from the DOE Office of Nuclear Physics RIA R&D program**.**

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. Photo-micrographs of this new material confirm that it is much finer-grained than that pressed from the commercial material.

Release Studies at Oak Ridge. An ISOL 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 testing. 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 during FY 2004 after completion of the modifications to the ISOL beam line.

Specific Accomplishments: Preliminary results of these studies have been reported at three national and international conferences and one workshop, as listed below.

- ANS annual meeting, Accelerator Applications Division Symposium, San Diego, California (June 2003).
    - Measurements of the Thermal Conductivity of Uranium Carbide Samples by the Method of Electron Bombardment, John P. Greene, Jerry Nolen, Maria Petra and Tatiana A. Burtseva.
- DOE RIA R&D Workshop, Bethesda, Maryland (August 2003).
    - High Power ISOL Target Development – R&D Report, John P. Greene, Jerry Nolen, and Tatiana Burtseva.
- International Conference on Radioactive Nuclear Beams, Argonne, Illinois (September 2003).
    - Uranium Carbide Fission Target R&D for RIA – an Update, J.P. Greene, A. Levand, J. Nolen, and T. Burtseva.
- International Conference on Thermal Conductivity, Knoxville, Tennessee (October 2003).
    - Direct Measurements of the Thermal Conductivity of Uranium Carbide Samples by the Method of Electron Bombardment, John P. Greene, Jerry Nolen, Maria Petra and Tatiana A. Burtseva.