

W-UO₂ CERMET FUEL DEVELOPMENT VIA PULSED ELECTRIC CURRENT SINTERING. J. A. Webb¹, J. Werner¹, J. Harp¹, ¹Idaho National Laboratory (1765 N. Yellowstone Hwy, Idaho Falls, ID 83415 and jon.webb@inl.gov)

Introduction: Historical W-UO₂ development programs have shown the feasibility of CERMET fuels for ultra-high temperature space reactors. However, previous programs have also shown the importance of ultra fine-grained tungsten structures within W-UO₂ fuel-meat [1], [2], [3], [4], [5].

Pulsed Electric Current Sintering (PECS) with post processing by low temperature Hot Isostatic Pressing (HIP) may be a viable route to produce full density W-UO₂ fuel specimens and elements for Nuclear Thermal Propulsion (NTP). Pulsed Electric Current Sintering is similar to hot pressing in the fact that a uniaxial stress is applied to a specimen and heated such that diffusional mechanisms of mass transport consolidate powdered metal particles together into a bulk, highly dense aggregate. Heating in PECS is accomplished by Joule heating, and allows for ultra-rapid densification that can minimize the grain size of the sintered aggregate [6].

A series of initial experiments have been carried out to determine the kinetic parameters for PECS consolidation of tungsten particles. The kinetic studies have led to a greatly enhanced understanding of the microstructural evolution of tungsten that occurs during PECS processing and has also led to an understanding of the underlying mechanisms associated with PECS processing.

Lastly, a second series of experiments has been conducted to determine the microstructural properties of W-CeO₂ specimens produced where CeO₂ is used as a surrogate for UO₂. Experiments investigated the differences between the sintered properties of specimens fabricated from mixed particles of W and CeO₂ as well as W coated kernels of CeO₂.

Kinetic Studies: Tungsten particles with average size of 0.471 μm and 4.24 μm were sintered at differing conditions in an effort to determine the activation energies for consolidation. Particles of both sizes were sintered to a peak temperature of 1873 K with a 5 minute hold time of 5 minutes at a ramp rate of 20 K/min, 50 K/min and 100 K/min. The specimen displacement, temperature, applied stress and other parameters were recorded at one second intervals. The recorded data was used in Continuous Rate Heating models to determine the activation energy for consolidation and the pre-exponential frequency factors involved during the first stage of sintering. The activation energy was found to be 402 ± 20 kJ/mol, which is well in agree-

ment with the grain boundary diffusion activation energy of 387 kJ/mol.

Another set of experiments ramped specimens at a rate of 100 K/minute to a temperature of 1173 K for a 2 minute isothermal soak. The temperature was again ramped at a rate of 100 K/minute to a temperature of 1273 K for an isothermal soak. The process or increasing the temperature by 100 K at a rate of 100 K/minute was repeated up to a peak temperature of 1973 K. The displacement and temperature was used in two different models to determine the sintering kinetics over the entire sintierng cycle. The data confirmed that grain boundary diffusion (Cobble creep) is the dominant mechanism during the initial and intermediate sintering stages and that lattice diffusion (Nabarro-Herring and Power-Law creep) takes over as the dominant mechanism at approximately 1773 K. While these experiments were carried out for pure tungsten, the mechanisms are expected to be the same for W coated particles of UO₂ or CeO₂.

W-CeO₂ Sintering Studies: Some question has been raised as to wether or not fuel surrogate particles (CeO₂ or UO₂) need to be coated with tungsten prior to consolidation. In an attempt to answer this questions particles of CeO₂ were mixed with particles of tungsten in a tubular mixer. The powders were mixed to yield 40 vol.% CeO₂ within the mixer. In one set of experiments, the CeO₂ particles were smaller than the tungsten particles, in another experiment, the CeO₂ particles were larger than the tungsten particles, and in the last set of experiments CeO₂ particles were coated with tungsten via a CVD process.

To date the only specimens sintered, were the compacts where the CeO₂ particles were smaller than the W particles, each having an average size of 281 nm and 471 nm respectively. The CeO₂ particles easily migrated into the tungsten grain boundaries and diffused away from the center of the specimen creating an area of larger CeO₂ concentration at the specimen periphery than at the center. In both the areas of high and low CeO₂ concentration, the CeO₂ ended up encapsulating the tungsten. In the region of lowest CeO₂ content, the CeO₂ and W usually ended up as an interdispersed matrix with no true regions of W or CeO₂ encapsulation. In a few isolated areas tungsten grains ended up encapsulating CeO₂ that had diffused to the triple points.

Nanometric particles of CeO₂ have been spray dried to batch sizes of 10 μm, 50 μm and 100 μm and

spherodized via a plasma process. Five hundred grams of the 100 μm batch are currently being mixed with 471 nm tungsten to 40 vol.% CeO_2 . These powder compacts will be sintered in the same conditions as the first mixed particle batch to demonstrate the effects of sintering mixed particles where the tungsten particle size is much less than that of the CeO_2 .

The remaining 100 μm , 50 μm and 10 μm CeO_2 particles are currently being coated with tungsten via the hydrogen reduction of WF_6 . The particles will be sintered in the near future to demonstrate the microstructure obtained when monosize coated particles and mixed size coated particles are sintered together within the PECS process. It is expected that the coated particles will segregate and encapsulate CeO_2 particles in a much better fashion than can be obtained by mixing W and CeO_2 particles of any size ratio.

References: [1] (1968) *Nuclear Rocket Terminal Report*, ANL-7236 [2] 710 *High Temperature Gas Reactor Closeout Report*, Vol. I and VI, GEMP-600, [3] Baker R.J. (1966) *Basic Behavior and Properties of W-UO₂ CERMETS*, BWNL-394 [4] Marlowe M.O. and Kaznoff A.I., *Development of Low Thermal Expansion Tungsten-UO₂ CERMET Fuel*, GESP-9014, [5] MacInnis M.B. and Schulze H.O. (1965) *Tungsten Cladding of Tungsten-Uranium Dioxide Composites by Chloride Vapor Deposition*, NASA CR 54728, [6] Munir Z.A., et al., (2006) The Effect of Electric Field and Pressure on the Synthesis and Consolidation of Materials; A Review of the Spark Plasma Sintering Method, *J. Mater. Sci.* 41, 763-777,