

FABRICATION OF GRAPHITE MATRIX FUEL FOR PULSED REACTORS*

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Introduction

A program was initiated by Sandia Laboratories (SLA) to increase the total fluence in the experimental cavity of the Annular-Core Pulsed Reactor (ACPR) for fast reactor fuel element transient and safety testing. The ACPR, described by D. J. Sasnor et al.⁽¹⁾ is a TRIGA-type reactor with a 23-cm dry central cavity serving as the primary experiment irradiation test region. The ACPR is fueled with a $\text{UZrH}_{1.6}$ alloy arranged in a hexagonal "annulus" to provide an experimental region 23 cm diam x 30 cm long. The present ACPR capabilities are steady-state operation to 300 kW [$\phi = 10^{13}$ n/(cm² sec)], and pulsing mode operation with an initial period as short as 1.3 msec, a pulse width (FWHM) of 4.5 msec, and a total fluence of 3×10^{15} n/cm² for a maximum pulse.

The large central irradiation cavity and the short initial periods have created interest in using the ACPR to simulate the conditions required for LMFBR fuel element and transient safety testing. A total cavity fluence of 8×10^{15} n/(cm² sec) for a maximum pulse and to ~ 2 MW for the steady-state power level are the goals for the reactor performance.

A two region core was proposed consisting of an outer driver region of the hydride fuel and an inner region of fuel with higher temperature and enthalpy capabilities. This would permit a higher pulse fluence capability and maintain the operating temperature limitations of the hydride fuel. The safety aspects of a strong prompt negative temperature coefficient would be provided by the hydride fuel region.

Several candidate fuels have been considered for the high-temperature regions of the core. One of these is the (U,Zr)C-graphite composite fuel developed by the Los Alamos Scientific Laboratory (LASL) during the Rover (Space Nuclear Propulsion Reactor) Program.^(2,3) This experience has also recently been used to fabricate carbide-graphite composite fuel rods for the TREAT converter studies⁽⁴⁾ being performed by Argonne National Laboratories (ANL).

Materials

Developmental fuel extrusions were made by extruding either a mixture of graphite flour-Thermax, ZrC, and UO_2 or graphite flour-Thermax and (U,Zr)C solid solution powder. All extrusions were bonded with Varcum 8251, a partially polymerized furfuryl alcohol. If a pore former was added to the mix it was either wood flour or tapioca starch.

Thermax (carbon black) was added to the extrusion mixes in amounts varying from 5 to 15 pph of the graphite flour.

The reactor grade ZrC and the 93% enriched $^{235}\text{UO}_2$ powders had an average particle size of 3.5 μm and 5.0 μm respectively.

The solid solution (U,Zr)C powder was made by reacting a mixture of ZrO_2 , UO_2 and carbon to 2550 K. Several powders with varying U contents, having high density particles, all -325 mesh, were made.

Two high purity graphite flours having different particle sizes and having different coefficients of thermal expansion (CTE) were used for both the extrusion and hot pressing work. The flours were -200 mesh, -325 mesh and < 10 μm .

All hot pressings were made with combinations of solid solution (U,Zr)C and graphite flours including special smaller fraction sizes other than -325 mesh. Camphor was used as a fugitive binder to prevent segregation of the powders.

Fabrication Procedures

All extrusion batches were mixed in a Patterson-Kelley twin-shell blender equipped with an intensifier bar and liquid addition apparatus. The dry ingredients and 88% of the required Varcum binder were mixed in the blender. The remaining Varcum binder was added step-wise during the extrusion operation.

The mix was successively passed through a commercial meat chopper and extruded a number of times to thoroughly homogenize all ingredients. The 150-ton extrusion press is equipped with a vacuum system for evacuation of the extrusion die, chamber and mix before each extrusion. The final extrusion was made into graphite fixtures used to contain the extrusion during the first stage of heat treatment. Extrusions were heat treated slowly over a period of 9 days to the final desired temperature, usually ~ 2900 K.

Hot pressing mixes were made by blending the required dry ingredients with camphor and chlorothene in a Patterson-Kelley blender.⁽⁵⁾ The camphor acts as a fugitive binder to condition the surface of the particle to prevent the segregation of the powders, which have widely different densities, during die loading operations. Chlorothene is used to dissolve the camphor to ensure thorough mixing but is evaporated from the mix prior to loading the hot pressing dies.

The graphite hot pressing dies are 32 mm inside diameter. The hot pressings were usually about 75 mm long. Hot pressing conditions varied but good pressings were made at pressures of 24 MPa (3500 psi) and temperatures around 2800 K.

Nondestructive Testing

The as-fabricated extrusions and hot pressings were radiographed for defects such as cracks, voids and inclusions. Density variations in the hot pressings were also detected. The fuels were gamma scanned in 1-cm increments to determine the ^{235}U distribution uniformity.

Discussions

Fuel materials for the ACPR must be able to withstand high thermal stresses. Important mechanical properties contributing to thermal stress resistance are low coefficient of thermal expansion (CTE), low modulus and high fracture strength. A fuel having high thermal diffusivity and heat capacity is also desirable. Because of the short heating time, rapid dissipation of the heat from the fuel particles to the entire fuel is necessary. Although other proposed fuels for the ACPR have higher heat capacities than the (U,Zr)C-graphite, the high potential enthalpy and thermal stress resistance of the graphite fuel makes it a good candidate.

*Work supported by the U.S. Nuclear Regulatory Commission (Project No. A1032) and the U.S. Energy Research and Development Administration under Contract AT(29-1)-789.

The first developmental fuel extrusions, 35.5 mm diam, contained ZrC, UO₂ and carbonaceous materials. During heat treatment the UO₂ converted to the carbide and entered into solid solution with the ZrC. In converting the oxide to carbide, carbon is lost lowering the graphite matrix density. When the solid solution is formed, a continuous carbide network results. Thus the heat treated extrusion consists of intertwined continuous networks of graphite and carbide. The carbide phase, 35 vol% of the fuel, has a high CTE so a high CTE graphite was used to reduce the mismatch in thermal expansion. Although this material was strong, the high CTE, $6.7 \times 10^{-6}/K$, tended to reduce its thermal stress resistance.

Hot pressings, which had high (35 vol%) carbide contents, tended to have isolated carbide particles although there was agglomeration of the carbide. The CTE was $> 7 \times 10^{-6}/K$.

Ideally, graphitic fuels would consist of small, $< 20 \mu m$, isolated fuel particles surrounded by a high density graphite matrix. Several changes were made to achieve this structure. The carbide content was reduced as was the size of the graphite flour which had a lower CTE than used previously. The combination of these changes should result in improved thermal stress resistance. Also, in the case of the extrusions, the carbide phase was added to the extrusion mix as (U,Zr)C solid solution powder rather than ZrC and UO₂. The CTE was reduced to $\sim 4.4 \times 10^{-6}/K$ and the matrix density increased from ~ 1.5 to 1.6 g/cm^3 . A high matrix density is required to increase the enthalpy of the system thus tending to reduce the maximum temperature the fuel would experience when pulsed.

Hot pressings made with the above changes still had high CTEs, $\sim 8 \times 10^{-6}/K$. The graphite matrix densities were as high as 2.0 g/cm^3 .

Experimental Results

Extrusions made with ZrC plus UO₂ which had relatively high porosities and a continuous carbide (35-vol%) structure could be readily fractured when pulsed in the ACPR.

Two extrusions, one in the shape of an annulus and the other a core to fit inside the annulus, were used and the carbide phase was added as (U,Zr)C solid solution powder. The annulus contained 20 vol% carbide (CTE of $4.3 \times 10^{-6}/K$) while the core, having a higher U loading, contained 30 vol% carbide and a CTE of $5.8 \times 10^{-6}/K$. This fuel was subjected to multiple reactor pulses producing the most severe temperature distribution and thermal stresses possible in the ACPR. No fractures, cracks or fragments were detected. Peak temperatures were approximately 2400 K.

An extrusion containing 17 vol% (U,Zr)C, also made with low CTE graphite and solid solution powder, was extruded in the shape of a solid 35.5-mm-diam cylinder. Samples of this extrusion were pulsed 6 times at full power to peak temperatures of 2350 K without failure. The CTE of this material was $4.4 \times 10^{-6}/K$ with a room temperature modulus of 18 GPa ($2.6 \times 10^6 \text{ psi}$) and fracture strength of 34 MPa (4900 psi).

Hot pressed fuels containing 35 vol% (U,Zr)C solid solution were made with "high" and "low" porosity, 20% and 11% respectively. The high porosity material failed at modest energy depositions but the material with a lower pore volume performed very well in the

ACPR tests. One set of samples withstood 55 maximum pulses without fracturing or chipping. The peak temperature was approximately 2300 K.

Further hot pressings having lower carbide contents (17 to 32 vol%) made with low CTE graphite flour all showed excellent thermal stress resistance. Fuel pellets survived numerous full power pulses without damage. The high matrix density of the hot pressings increased the enthalpy of the fuel so that for a given power input the peak temperature would be about 10% less than for a similar extruded fuel. A typical hot pressing had a CTE of $7.7 \times 10^{-6}/K$, room temperature modulus of 17 KPa ($2.4 \times 10^6 \text{ psi}$) and fracture strength of 38 MPa (5500 psi).

Other properties measured on many samples, both extrusions and hot pressings, were heat capacity, thermal diffusivity, enthalpy, fracture strain and peak to minimum radial energy profile.

Conclusions

Extrusion and hot pressing techniques have been developed for fabricating developmental fuel for the ACPR. Both methods produced fuel materials which have been shown to survive more severe thermal stress environments than will be present in the upgraded version of the ACPR. An extensive data base exists for this type of fuel and the manufacturing procedures are considered state-of-the-art.

References

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