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HEAD-END REPROCESSING STUDIES WITH IRRADIATED HIGH TEMPERATURE GAS-COOLED REACTOR (HTGR) FUELS*

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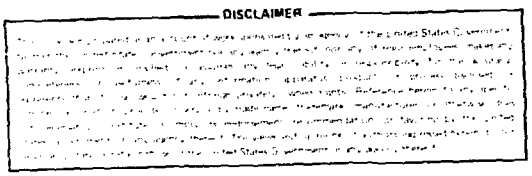

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The concept for the HTGR fuel is a feed-breed system based on uranium-235 and bred uranium-233 fissile particles mixed with thorium fertile particles in graphite fuel elements. The breeding ratio is less than unity, so these reactors are classed as converters. To prevent the accumulation of ^{236}U in the fissile fuel, it is necessary to keep the ^{235}U from mixing with the ^{233}U. The feed and breed particles are fabricated into microspheres of different sizes and coated with pyrolytic carbon and, in some cases, with an intermediate layer of silicon carbide. These coatings perform as miniature pressure vessels to contain the fission products during irradiation and as combustion barriers when the graphite is removed in the primary burning step to permit the separation of the two isotopes.

Head-end reprocessing studies with irradiated HTGR-type fuels have been in progress for several years.[1-6] These studies at Oak Ridge National Laboratory are part of a larger effort at developing a commercial reactor system by the Gulf-General Atomic Company and the Department of Energy.[7] The HTGR reprocessing flowsheet consists of the following steps: fuel block crushing, fluidized bed primary burning, coated particle separation, coated particle crushing, fluidized bed secondary burning, dissolution, clarification, and solvent extraction.

The fuel used for this experiment was (U-2.75 Th)C_2 TRISO-coated (contains SiC layer) fissile particles, and ThC_2 BISO-coated (no SiC layer) particles bonded into rods that were 1.25 cm diameter and 4.93 cm long. The fuel was contained in test element FTE-1 and was irradiated in the Peach Bottom Reactor at 1034 to 1177°C for a period of 448.8 effective full power days from July 11, 1[illegible] to [illegible] 14, 1973. The composite FIMA (fissions per initial metal atom)

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was a peak of 7.11% with a mean value of 5.64%. Fifty fuel rods, 631.0 g, were used for this study.

The fuel rods were visually inspected, crushed, and burned in the fluidized-bed primary burner at 875°C in 95% N_2-5% O_2 at a fluidizing velocity of 0.8 m/s. The cyclone fines and filter fines, collected from the off-gas during the burning operation, were weighed and analyzed. The burner product was weighed, and the fertile fraction (bare ThO_2 microspheres) was separated from the fissile fraction (SiC coated, $^{235}UC_2$ particles) by dissolving in 13 M HNO_3-0.1 Al^{3+}-0.05 F^- (Thorex reagent). The insoluble fraction was sieved through a 60-mesh sieve (250 μm) to recover the coated particles from the insoluble fission products. The insoluble fraction and samples of the dissolver solution were analyzed. The coated particles were processed by crushing in a roll crusher with a 280 μm gap and by burning in the fluidized-bed secondary burner at 875°C in 80% N_2-20% O_2 at a velocity of 0.76 m/s. The burner product was weighed and dissolved in Thorex reagent. The insoluble residue was dried and weighed. Samples of the dissolver solution and residue were submitted for analyses.

The experiment was analyzed by three methods: (1) weight balances measured for each operation, (2) carbon balance calculations based on in-line flow and in-line CO and CO_2 analyzers, and (3) heavy metals and fission product data obtained by analytical methods. The results of these methods were compared with each other and with the expected values based on fuel fabrication parameters and the irradiation history.[8]

Calculations of carbon balances from on-line instrumentation showed that 307.8 g of carbon could be accounted for in the off-gases from the primary burner. The expected weight loss was 303 g. The primary burner product ash, however, was about 20 g short of the expected value. The SiC coated particle

fraction was within 1.5% of the expected value after dissolution. Analysis of the dissolved fertile ash indicated that the ThO_2 content was about 20 g short. The missing 20 grams of material was accounted for in the next experiment, so it was clear that a temporary holdup was experienced in our equipment.

The crushed SiC coated fissile particles were burned. Analyses of the secondary burner off-gases accounted for 112% of the expected carbon. The secondary burner product was 94.5% of the expected value, and the dissolver residue (SiC hulls) was 86% of the expected result. These results indicate that the SiC coated particles contained slightly more carbon, and less SiC, than expected from fuel fabrication parameters. All the other data agree with expected results within about 5%. We conclude that good control was experienced in these experiments.

The analysis of the data generated by analytical methods can be facilitated by separating into the fertile and fissile groups. Two solid samples, the cyclone fines and filter fines, are included with the fertile fraction.

The recovered values of heavy metals averaged 2.01 g Th and 0.33 g U per fuel rod for the fifty rods in this experiment. These results can be compared with end of life (EOL) calculations, based on the irradiation history and average beginning of life (BOL) metal loadings, which predicted values of 2.54 g Th and 0.39 g U per fuel rod. Our experimental values represent 80 to 84% of the predicted values for the heavy metals.

The measured isotopic uranium distribution agrees very well with calculations and analyses of the fertile dissolver solution clearly shows that only about 5% of the coated fissile particles had been broken up to the time of leaching. The distribution of fission products also supports this conclusion, and in addition shows that about 40% of the ruthenium is contained in the insoluble dissolver residues. The recovered totals of fission products agrees with previous non-destructive analyses done at ORNL on similar fuel rods from the same test element.

In summary, our experiments with FTE-4 fuel have been consistent with predictions, in general, and the measured internal consistency of the data provide reasonable assurance that the reprocessing of TRISO-BISO fuel can be accomplished by the proposed flowsheet steps.

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