DEFECT FRACTIONS FOR FISSILE AND FERTILE TRISO COATED FUEL

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1.0 SUMMARY

High quality TRISO-coated UCO and ThO_2 particles with reference MHTGR dimensions were produced in a coating campaign in August and September 1986 for irradiation tests. The heavy metal contamination and the defect levels were below the following limits established for the MHTGR fuel:

<u>Property</u>	<u>Mean Value</u>
HM Contamination Fraction	$\leq 1 \times 10^{-5}$
SiC Defect Fraction	$\leq 5 \times 10^{-5}$
Missing Buffer Fraction	$\leq 5 \times 10^{-5}$
Missing or Defective IPyC	$\leq 4 \times 10^{-5}$
Missing or defective OPyC	$\leq 1 \times 10^{-4}$

Over 9 kg of uranium in UCO and 30 kg of thorium in ThO_2 were TRISO-coated in 4 fissile and 3 fertile batches in the 240mm Development Coater. These coated fuel particles will be used to produce fuel rods for testing in the irradiation validation tests to be conducted in capsules HRB-19,-20 and -21 on the DOE Fuel and Fission Product Technology Program.

2.0 INTRODUCTION

The purpose of this effort was to produce high quality TRISO-coated UCO and ThO_2 fuels for use in irradiation testing of MHTGR fuel. The irradiation capsule tests to be conducted in HRB-19,-20 and -21 will be the first test of the MHTGR fuel in which the exposed heavy metal contamination and coating defect levels have been reduced a factor of 10x relative to the specified fuel quality of prior HTGR designs. In August and September of 1986 the 240mm Development Coater with an improved draft tube coating zone configuration was used to successfully produce the required high quality UCO

and ThO_2 TRISO fuel particles in large batch sizes representative of production scale. The following sections describe the properties of the coated fuel particles.

3.0 UCO TRISO-COATED FUEL PARTICLES

TRISO-coated 350 μm diameter UCO (19.7% enriched) fuel particles produced in this campaign used UCO kernels prepared in the pilot plant to MHTGR specifications. Two UCO kernel composites (NU38-10001 and NU38-10003) were used to produce the 4 fissile coated fuel particle batches. Mean UCO kernel properties for the two composites are presented in Table 3.1 below:

Table 3.1

UCO (19.7% Enriched) Kernel Properties

Property	Specification	NU38-10001	NU38-10003
Density (Mg/m^3)	≥ 10.5	10.67	10.65
Mean Diameter (μm)	≤ 360	345	351
Weight % U	≥ 87.0	88.6	88.6
Carbon/Uranium (atomic ratio)	≤ 0.4	0.3	0.2
Oxygen/Uranium (atomic ratio)	1.8-1.6	1.64	1.65

Other UCO kernel properties for these composites are being measured and the data will be reported in a later report.

TRISO coatings were deposited on the UCO kernels in the 240mm Development Coater using the improved draft tube configuration at an initial UCO kernel batch size of 2215 grams of uranium (2500 gm of UCO kernels). Reference 350 μm fissile kernel batch sizes in the 240mm coater have historically been conducted at 3200 gm of uranium. However, it was not possible to operate in the development coater with water coolant in excess of 457 gm of U-235. The 2500 gm UCO (19.7 % enriched) kernel batch size containing approximately 2200 gm HM allowed these operations to proceed without modification to the coolant system. A batch coating sequence

(buffer-seal-IPyC, SiC, and OPyC) was used for this coating to allow sampling for all required QC measurements.

Mean dimensional particle properties for the 4 TRISO-coated UCO particle batches produced and the MHTGR product specifications are presented in Table 3.2 below:

Table 3.2

TRISO-Coated UCO Particle Dimensional Data

Coating Batch Number	UCO Kernel Batch Size (HM) (gm)	UCO Kernel Composite No. (NU38-)	Mean Coating Dimensions (μm)			
			Buffer	IPyC/Seal	SiC	OPyC
MHTGR Spec.	5000 ^(a)		90-110	40-60	30-40	30-50
8876-64	2215	10001	102.8	50.6	35.0	34.1
8876-66	2215	10001	105.5	50.0	35.2	41.8
8876-68	2215	10001	104.7	48.6	35.2	41.8
8876-70	2215	10003	105.0	52.8	32.6	42.8

a) Long range development UCO batch size goal for Heavy Metal content

Coating defects and exposed heavy metal contamination levels for the MHTGR fuels are normally measured at the fuel rod stage as required by the product specification. However, in order to assess the capabilities of the TRISO-coated UCO particles produced in this work to meet these requirements at the rod stage, an evaluation of the coated UCO particles was made. The defective particle and the exposed heavy metal contamination levels for the 4 fissile particle batches are presented in Table 3.3 below:

Table 3.3

Defective Particle and Exposed Heavy Metal Contamination
For TRISO-Coated UCO Particles

Coating Batch No.	Surface Contam. Fraction	Missing Buffer Fraction	Missing or Permeable IPyC Fraction	Defective SiC Fraction	Missing or Defective OPyC Fraction
MHTGR Spec.	$\leq 1 \times 10^{-5}$	$\leq 5 \times 10^{-5}$	$\leq 4 \times 10^{-5}(a)$	$\leq 5 \times 10^{-5}$	$\leq 1 \times 10^{-4}$
8876-64 ^(b)	7.4×10^{-7}	1.7×10^{-5}	(a)	8.9×10^{-6}	$\leq 6.7 \times 10^{-5}$
8876-66 ^(b)	6.6×10^{-7}	3.2×10^{-5}	(a)	5.0×10^{-6}	$\leq 7.1 \times 10^{-5}$
8876-68 ^(b)	8.0×10^{-6}	2.0×10^{-5}	(a)	1.5×10^{-5}	$\leq 7.4 \times 10^{-5}$
8876-70 ^(b)	7.4×10^{-8}	4.1×10^{-5}	(a)	2.5×10^{-5}	$\leq 7.5 \times 10^{-5}$

a) Measurement taken at fuel rod stage by fuel dispersion.

b) Defect and contamination taken after washing but before liquid elutriation and screening.

Defective coated fuel particle data given in Table 3.3 above were taken on washed but not liquid elutriated and screened material. Elutriation and screening has been shown to reduce defects by removing a few highly faceted particles from the parent batches (Ref.2). Therefore, some small additional improvement in particle quality maybe realized after these operations.

Examples of TRISO-coated UCO particle microstructure and overall particle shapes for these materials are shown in Figures 3.1, 3.2 and 3.3. Metallographic mounts were prepared to examine the coatings for key properties such as IPyC and OPyC porosity and SiC coating flaws. All coatings showed microstructures that were typical for high density, low porosity PyC coatings. Other PyC QC data taken confirmed these observations. Coated fuel particle faceting (aspect shape ratios) data were low being ~1.040 versus MHTGR product specification limits of 1.200 (maximum/minimum particle diameter ratio).

MP86008-1

Fig. 3-1. Metallographic examination showing field view of typical microstructure for TRISO-coated, 20% enriched, 350 μ m UCO kernel, fuel particles from batch 8876-70.

MP86008-2

Fig. 3-2. Photomicrograph showing typical microstructure of a TRISO-coated, 20% enriched, 350μm UCO kernel, fuel particle from batch 8876-70.

MP86008-3

Fig. 3-3. Photomicrograph showing microstructure of layers of a TRISO-coated, 20% enriched, 350μm UCO kernel, fuel particle from batch 8876-70.

4.0 TRISO-Coated 500 μ m ThO₂ Particle Data

Three batches of TRISO-coated 500 μ m ThO₂ fuel particles were prepared to produce high quality fuel for the MHTGR capsule test program. Thorium oxide kernel materials used as coating substrate for this coating were obtained from ORNL earlier this year. This ThO₂ kernel material had been produced at GA in the late 1970's and sold to ORNL for their coating development work. The GA ThO₂ kernel sintering process at that time included a final sintering temperature of ~1200 C which was found later to be too low to completely convert and sinter the ThO₂ kernels. Thirty five kilograms of the ThO₂ kernels obtained from ORNL as virgin material were re-sintered at 1600 C in air to produce coating substrate for this work (Ref. 3). The ThO₂ kernel QC data for this material are presented in Table 4.1 below:

Table 4.1

ThO₂ Kernel QC Data For Re-sintered Material

Property	MHTGR Specification	Measured Value
Impurity(ppm)		
Fe	≤ 100	≤ 50
Cr	≤ 100	≤ 5
Ni	≤ 100	≤ 1
Total impurities	≤ 5000	≤ 287
Density (Mg/m ³)	≥ 9.50	9.94
Mean Diameter (μ m)	≤ 510	512

4.1 TRISO-Coated ThO₂ Fuel Particles

Three batches of TRISO-coated 500 μ m ThO₂ fuel particles were coated in the 240mm Development Coater using the draft tube coating configuration. The purpose for these coated ThO₂ fuel particles was to produce high quality fertile particles² to be used in fuel rod fabrication for irradiation testing of the MHTGR fuel design in capsules HRB -19, -20 and -21. Initial ThO₂ kernel batch sizes of 10 kg were used in the three fertile TRISO coated fuel particle batches produced. A batch coating sequence

(buffer-seal-IPyC, SiC, OPyC) was used for the fertile TRISO coating to allow statistical QC sampling of the material at each coating stage to measure coating properties as required by the MHTGR product specifications. Key dimensional properties and the MHTGR product specifications are presented in Table 4.2 below:

Table 4.2

TRISO-Coated 500um ThO₂ Particle Dimensional Data

Coating Batch No.	Kg HM	Mean Dimension (μm)			
		Buffer	IPyC/Seal	SiC	OPyC
MHTGR Spec.	≥10 ^(a)	55-75	40-60	30-40	30-50
8876-52	10.0	63.1	54.6	33.0	39.0
8876-56	9.81	68.2	55.5	35.1	40.4
8876-58	9.81	67.1	56.4	36.0	41.1

a) Long range development goal.

Defective coating fraction measurements for key properties of the coated fuel particles are required at the fired fuel rod stage. However, defective fuel particle properties were measured at the coated fuel particle stage for the three fertile TRISO-coated ThO₂ particle batches produced to assess their capability to meet MHTGR product specifications. The defective coated fuel particle data for the three fertile batches are presented in Table 4.3 below:

Table 4.3

Coating Defects For TRISO-Coated ThO₂ Fuel Particles

Coating Batch No.	Surface Contam.	Missing Buffer	Missing or Permeable IPyC	Defective SiC	Missing or Defective OPyC
MHTGR Spec.	$\leq 1 \times 10^{-5}$	$\leq 5 \times 10^{-5}$	$\leq 4 \times 10^{-5}$	$\leq 5 \times 10^{-5}$	$\leq 1 \times 10^{-4}$
8876-52	3.7×10^{-7}	1.8×10^{-5}	(a)	3.3×10^{-5}	$\leq 7.0 \times 10^{-5}$
8876-56	$< 2.0 \times 10^{-7}$	7.3×10^{-6}	(a)	1.6×10^{-6}	$\leq 7.3 \times 10^{-5}$
8876-58	$< 1.5 \times 10^{-7}$	1.7×10^{-5}	(a)	3.2×10^{-5}	$\leq 6.5 \times 10^{-5}$

a) Measured at fuel rod stage by fuel dispersion.

Defective fuel particle data given in Table 4.3 were taken on washed but not liquid elutriated and screened material. Removing some small fractions of the fertile fuel particle batches which consist of coated kernel fragments and large diameter coated doublet materials should show further improvement in coating quality.

The coating microstructures for the TRISO-coated 500 μ m ThO₂ fuel particles produced were examined. Metallographic mounts were prepared and pictures of some typical fertile particles from the batches are shown in Figures 4.1, 4.2 and 4.3. The appearances of the IPyC and OPyC coating microstructures were typical of high density and low porosity PyC coatings. Overall particle shapes were well within product specification limits. Mean faceting (aspect ratio) values for the three fertile particle batches produced were 1.036 to 1.045 (minimum/maximum particle diameter) as compared to a allowable specification limit of 1.20.

MP86003-1

Fig. 4-1. Metallographic examination showing field view of typical microstructure for TRISO-coated, 500μm ThO_2 kernel, fuel particle from batch 8876-56.

MP86003-2

Fig. 4-2. Photomicrograph showing typical microstructure of a TRISO-coated, 500 μ m ThO₂ kernel, fuel particles from batch 8876-56.

MP86003-3

Fig. 4-3. Photomicrograph showing microstructure of layers of a TRISO-coated, 500μm ThO₂ kernel, fuel particle from batch 8876-56.

5.0 CONCLUSIONS

Four fissile and three fertile TRISO-coated UCO and ThO₂ fuel particle batches were produced in the 240mm Development Coater using the improved draft tube configuration. All key product properties for these materials met MHTGR product specifications which are a factor of 10x lower than previous HTGR fuel specifications.

6.0 REFERENCES

- 1) Scheffel, W. J., "HTGR Fuel Product Specification For MHTGR", HTGR-86-081, GA Doc. No. 901588/E, GA Technologies Inc., September 1986.
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