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CHLM-5177

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Contract AF(30-1)-2789
May 21, 1963

Mr. L. M. Haring

M. A. DeCrescente

SNAP-90 Fuel Development,
January-March, 1963

cc: Messrs.

W. G. Kennedy ✓

R. H. Johns

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FUELS RESEARCH AND DEVELOPMENTURANIUM CARBIDE

Irradiation Testing: Post irradiation evaluation was completed on capsule RW26-190 (stoichiometric UC with 1 mil tungsten foil). Evaluation of capsule RW26-151 (5.3% carbon with 2 mil tantalum barrier) was started. This latter capsule was tested at 2175F for 10,027 hours with a power density of approximately 1.7 kw/cc. Preliminary examination showed no fuel-cladding reaction, thus further confirming the effectiveness of tantalum as a fuel-clad barrier, as indicated by out-of-pile tests. Fission gas release was about 7% and a slight amount of diametrical and longitudinal swelling of the pins was noted. Detailed data for this capsule and also for capsule RW26-190 are summarized below. The marked difference in gas release between these 2 capsules is notable and is presumably associated with the difference in density of the fuel pellets. Work currently in progress to improve the strength of the fuel matrix, to improve the strength of the clad material, and to eliminate any possibility of fuel-clad reactions are all intended to effectively deal with a possible gas release problem.

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BY: TED REDMON DATE: 11-3-73

SPECIAL REREVIEW FINAL DETERMINATION	Reviewers	Class.	Date
Class: <u>U</u>	HFC	<u>U</u>	11/30/81
	KAW	<u>U</u>	12/2/81

To: Mr. L. M. Raring

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	Capsule	
	<u>W26-151</u>	<u>W26-190</u>
Enrichment (% U^{235})	10	10
Density (% theoretical)	95.2	92.9
Composition (% O)	5.3	4.8
Barrier	Tantalum	Tungsten
Temp. (°F)	2175	2055
Time (Hrs.)	1027	1069
Power Density (W/cc)	1.64-1.74	1.5-1.7
Burnup (% U^{235})	6.6-7.0 ⁽¹⁾	6.2-6.5 ⁽²⁾
Gas Release (max. %)	7.2-7.5	76
Pin Swelling	Slight ⁽³⁾	None
Fuel-Clad Interaction	None	None ⁽⁴⁾

(1) G-137 analysis - preliminary

(2) Mass spectrometer analysis - final

(3) Swelling was highest in the middle pin; in diameter - 5 mils (1.7%), in length - 4.4 mils (0.3%).

(4) Some reaction was noted in unprotected regions where the clad was directly exposed to the UC.

The over-all status of the uranium carbide irradiation program is summarized in Table I.

Powder Preparation: The silver-palladium hydrogen diffusion cell, which is being evaluated as a hydrogen getter to replace uranium metal, was installed in a test stand

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for preliminary trial operation prior to installation in the UC synthesis rig. Observation of pressure changes in mixed samples of methane and hydrogen introduced into the apparatus, indicated that hydrogen was removed selectively from the mixed gas. Based upon these promising preliminary trials, the equipment will be installed with the pilot synthesis rig. If successful, the synthesis cycle time will be significantly reduced.

Raw Materials: Difficulties in achieving consistently high density in sintered UC and UN have been traced to the quality of the starting metals. The problem has been identified with specific lots of metal. Chemical and spectrographic analysis of material from good and poor sintering batches have, so far, given no indications of any significant differences. An investigation has been started to determine whether the physical condition, particularly cold reduction, of the starting uranium has any effect on the sinterability of the final product.

Pellet Fabrication - Powder Lubricants and Solvents: Continuation of the evaluation of internal lubricants and solvent carriers showed that normal hexane, which has a very low affinity for water, was preferable as a solvent for lubricants with UC. Several internal lubricants are being investigated as possible substitutes for the external die lubricants presently used for hand-fabricated pellets. Additions of about 2% of cetyl alcohol to UC powder resulted in small increases in both the green and sintered densities. Slight carbon pick-up occurred and the need for an external lubricant, though reduced, was not fully eliminated.

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Table 1

Status of UC Irradiation Testing

10% Enriched, 2200F

<u>Capsule Designation</u>	<u>Percent Carbon</u>	<u>Barrier</u>	<u>Pellet Density</u>	<u>Scheduled Exposure</u>	<u>Status</u>
PW26-150	5.2	Ta	93.8	3,000	At MTR
151	5.3	Ta	95.2	1,027	Evaluated
152	5.3	Ta	94.3	10,000	At MTR
153	5.3	Ta	95.2	10,000	At MTR
154	5.3	Ta	-	3,000	Pellets prepared
160	5.3	Ta	95.9	1,000	At MTR
170	5.0	None	97.0	3,000	At MTR
171	5.0	None	97.0	1,065	At Hot Lab.
173	5.3	None	96.1	10,000	At MTR
174	5.3	None	96.1	10,000	At MTR
181	4.8	W	93.5	3,000	At MTR
190	4.8	W	92.9	1,069	Evaluated
191	4.8	W	92.9	2,000	At Hot Lab.
192	4.8	W	92.9	10,000	At MTR
195	4.8	W	-	3,000	Being Prepared
196	4.8	W	-	1,000	Being Prepared

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UC Procurement: A sample of gas-synthesized uranium carbide received from United Nuclear was subjected to a chemical and sintering analysis. Results of the chemical analysis (tabulated below) meet the CANEL specifications for starting material.

Chemical Analysis of Commercial Powder (Batch E-432)

Carbon	4.75%
Oxygen	900-1000 ppm
Nitrogen	175-210 ppm

Sintered densities were low, however, 92% of theoretical density being the maximum achieved from this material as opposed to 96% from UC powder synthesized according to the procedures developed at CANEL. The differences are believed to be associated with the particle size of the material.

UC Prototype Facilities: The Stokes mechanical press, containing an integral vacuum chamber, and specially designed for SNAP-50 fuel production, was received in late March. The equipment is being installed in the Fuel Fabrication Shop. Production parameters will be determined for the fabrication of UC pellets as soon as installation is completed. Prior to delivery, a visit was made to the Stokes plant where the equipment was observed in operation and pellets of UC were pressed in vacuo.

UC Out-of-Pile Compatibility Evaluation:

Out-of-pile compatibility testing of uranium carbide with Cb-1 Zr alloy was continued under two sets of conditions: (1) between compatibility couples (with and without barriers) comprising small specimens of the materials in intimate contact with each other and (2) between fuel and clad materials simulating the SNAP-50 fuel pin configuration and tested in a lithium environment. Results of the experiments evaluated in this period are presented below:

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Tungsten-Barrier Evaluation: Compatibility couples with a one mil tungsten foil barrier between Cb-1 Zr alloy and UC (approximately 4.8% C) have now been run for periods of up to 3000 hours at 2200F. Results of these longer test periods confirm previously reported data, viz., a slight depletion of the concentration of acicular UC_2 phase near the pellet surface and scattered Ch_2C precipitate in the clad. The appearance of the test materials after furnace testing was found to be consistent with the 1000 hour impile results reported for irradiation capsule BW26-190.

Samples of Cb-1 Zr tubing with a vapor deposited tungsten coating on the inside surface are being obtained from two sources. This composite tube may provide an improvement over loose tungsten foil as a barrier and, in addition may offer a considerable strength advantage.

Hyperstoichiometric UC-Cb-1 Zr Alloy Evaluation: In another test, free uranium was formed at the interface between UC (5.3% carbon) and Cb-1 Zr alloy in a 1500 hour at 2200F. Figure 1 shows uranium penetration into the Cb-1 Zr clad to a depth of approximately 10 mils. This is not consistent with other tests in which no significant reactions were observed under similar conditions. It is thought that the degree of surface contact is important. Where close contact exists, a thin layer of ChC is formed on the clad surface in contact with the UC. This layer appears to serve as a barrier to the diffusion of uranium. However, the practical conclusion to be drawn from the results as illustrated in Figure 1 is that excess carbon (as U_2C_3) cannot be depended upon to prevent serious reaction with Cb-1 Zr alloy clad.

Lithium Testing: Compatibility experiments simulating SNAP-50 fuel pin configurations were removed from test after 3000 hours at 2200F in a lithium environment. These tests represent continuation of the 1000 hour tests reported in BWAC-624.

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The fuel was hyperstoichiometric (5.2% C) with both bare Gb-1 Zr clad and tantalum-lined Gb-1 Zr clad. A 1/16" diameter hole was drilled through the clad of one half of the specimens prior to test to permit lithium penetration during test. The purpose of this was to assess the effect of lithium leakage on the interaction between the clad and fuel. Data obtained from both the 1000 and 3000 hour tests are compiled in Table 2. Figure 2 is a photograph of the exterior surface of pins after exposure for 3000 hours at 2200°F. Among the significant points derived from the analysis are:

Bare Gb-1 Zr Alloy: After 1000 hours, only a small amount of carbide phase was noted on the clad surface while the 3000 hour tests showed widely scattered carbides throughout the full clad thickness. In contrast to the results shown in Figure 1, there is neither a uniform layer of $\text{U}_3\text{Zr}_7\text{C}_2$ nor any evidence (by radiography) of uranium diffusion into the clad. As mentioned previously, these differences are believed to be caused by differences in contact conditions between the fuel and clad surface.

The posttest structure of the defected pins, tested for 1000 hours, is illustrated in Figure 3. Note the apparent volume increase of the cladding (thickness increased from about 22 mils to between 27 and 40 mils. See Figure 2d). Radiographic analyses showed uranium through virtually the entire clad thickness of both the 1000 hour and 3000 hour tests.

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Table (2)

Summary of 1019 and 3000 hour, 2200F Hyperstoichiometric Uranium Carbide -
Columbium-1 Zr-Lithium Compatibility Tests

<u>Cladding Material</u>	<u>Cladding Condition</u>	<u>Fuel</u>			<u>Clad Attack</u>				<u>Barrier Attack</u>			
		<u>Carbon Content (%)</u> <u>Pretest</u>	<u>Posttest</u>		<u>Carbide</u> <u>Penetration</u>		<u>U Penetration</u> <u>(mils)</u>	<u>Carbide</u> <u>Penetration</u>		<u>U</u> <u>Penetration</u>		
			<u>1019</u>	<u>3000 (3)</u>	<u>1019</u>	<u>3000</u>		<u>(mils)</u>		<u>(mils)</u>		
								<u>hr.</u>	<u>hr.</u>	<u>hr.</u>	<u>hr.</u>	<u>hr.</u>
.....												

- (1) Lithium attack to Columbium-1 Zr suspected
 (2) 1/16" dia. hole drilled in one end of pin.
 (3) Carbon analysis in process
 (4) Reacted zone observed.

Key: Comp. - Complete

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Tantalum-lined U-1 Zr Alloy: The effectiveness of tantalum in preventing reactions between U₂ and U-1 Zr alloy is illustrated by Figure 4. Carbon diffusion has not progressed beyond the U-1 Zr/tantalum interface and there is no evidence of uranium diffusion in the U-1 Zr clad. The defected pins tested for 1000 hours, showed no uranium diffusion but the 3000 hour tests revealed significant uranium diffusion into the U-1 Zr alloy clad. These tests indicated, at least, that a tantalum barrier effectively reduces the rate of reaction between U-1 Zr and U₂ in the presence of lithium.

IRADIATION HISTORY

Irradiation: Examination of the first 2200F U₂ irradiation capsule, W26-210, which was irradiated for 1000 hours, showed (1) no swelling, (2) slight reaction with the cladding, and (3) low fission gas release. Tungsten foil, which has been found to be effective in eliminating the fuel-clad interaction in out-of-pile tests, was not used in this test but has since been incorporated in the irradiation program. A summary of the results of the post-irradiation evaluation of W26-210 is presented below together with previous tests for comparison.

	Capsule		
	<u>W26-200(2)</u>	<u>W26-201(2)</u>	<u>W26-210(2)</u>
Enrichment (% U ²³⁵)	93	93	30
Density (% theoretical)	90	91	96
Barrier	none	none	none
Temp. (F)	1720	1990	2120
Time (Hrs.)	391	1595	1080
Power Density	0.4	0.6-1.2	1.1
Burnup (% U ²³⁵)			
Gas Release (%)	0.1	3.5-5.1	2.5-3.8
Pin Swelling	none	none	none
Fuel-Clad Interactions	slight	slight	slight

Table 3 is a status summary of the full U₂ irradiation schedule.

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Table (3)

Status of UN Irradiation Testing

<u>Capsule Designation</u>	<u>Temp. (°F)</u>	<u>Barrier</u>	<u>Enrichment (% U-235)</u>	<u>Scheduled Exposure</u>	<u>Status</u>
PW26-200	1750	None	93	390	Evaluated
201	2000	None	93	1,600	Evaluated
210	2200	None	30	1,000	Evaluated
220	2200	None	10	3,000	At MTR
230	2200	W	10	1,000	At Hot Lab
231	2200	W	10	3,000	At MTR
240	2200	W	10	10,000	At MTR
241	2200	W	10	4,000	At MTR
242	2200	W	10	6,000	Pins Complete
243	2200	W	10	8,000	Pins Complete
244	2200	W	10	1,000	Pins Complete
250	2200	W	10	1,000	Being Prepared
251	2200	W	10	3,000	Being Prepared

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UN Synthesis and Fabrication: The current production capacity for high purity UN powder, prepared by the CAMEL process, is between 50 and 60 grams in three days. Particle size of the powder is approximately 2 microns. Analyses show close to stoichiometric compositions with maximum contamination limits of 2000 ppm oxygen and 600 ppm carbon. Design has been completed and procurement of parts and construction started on apparatus for the synthesis of larger batch sizes. Final densities consistently above 96% of theoretical can be achieved by cold pressing and sintering.

UN Procurement: A sample of UN powder has been received from Namco and subjected to a chemical and sintering analysis. The sintering behavior of this material has been good - densities of 97% of theoretical being achieved in spite of a relatively high oxygen content (2800 ppm). However an unidentified phase was observed in the microstructure. A second sample has been ordered.

UN Compatibility: Diffusion couples of uranium mononitride and Cb-1 Zr alloy have shown interaction between the materials above 2000F. The reaction products are a (U-Cb) alloy and Cb_2N precipitate. Although the interaction of UN is considerably less than that of UC at comparable temperatures, even small amounts of uranium in the clad is intolerable and has led to the evaluation of barrier materials.

Tungsten Barrier Evaluation: Tungsten foils 1 mil thick have been found to effectively prevent the interaction between UN and Cb-1 Zr alloy for periods of up to 1700 hours at 2200F and 100 hours at 2800F. Additional diffusion couples of these materials are being prepared for a more complete evaluation.

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SOLID SOLUTION CAPSULES

Irradiation: Three capsules containing varying compositions of UC-ZrC are scheduled for irradiation. Based upon out-of-pile experimental data it is believed that through solid solutioning of UC with other refractory carbides such as ZrC and CbC, a stable fuel matrix may be achieved which will reduce fission gas release and possibly swelling. Compositions which will be evaluated include: 95 m/o UC-5 m/o ZrC; 83 m/o UC-17 m/o ZrC; and 71 m/o UC-29 m/o ZrC. The last composition, containing the highest concentration of ZrC, will have the same U loading as UO_2 . The anticipated irradiation conditions are 2200F for 1000 hours. Material for the first capsule (95 m/o UC and 5 m/o ZrC) is now being prepared.

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FIG 1

2598-170

Cb-1 Zr ALLOY COMPATIBILITY SPECIMENLEGEND

- A - CbC - 2 MILS
- B - Cb-U ALLOY - 8 MILS
- C - Cb₂C PARTICLES

CONDITIONS: STAIN-ETCHED

MAGNIFIED: 100X

SHOWING REACTION BETWEEN URANIUM CARBIDE (5.3 W/O C)
AND Cb-1 Zr ALLOY AFTER 1500 HOURS AT 2200F.

(LITHIUM ENVIRONMENT)

LEGEND

C - TANTALUM LINED DEFECTIVE PINS
D - UNLINED DEFECTIVE PINS

A - TANTALUM LINED SOUND PINS
B - UNLINED SOUND PINS

CONDITION: STAIN-ETCHED

MAGNIFIED: 100X

EXTERIOR OF Cb-1 Zr ALLOY FUEL PINS CONTAINING
URANIUM CARBIDE (5.0-5.2 W/O C) AND EXPOSED
3000 HOURS AT 2200F IN A LITHIUM ENVIRONMENT.

FIG 2

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FIG 3

Cb-1 Zr ALLOY VS. URANIUM CARBIDE COMPATIBILITY SPECIMENS (LITHIUM ENVIRONMENT)

LEGEND

- A CbC
- B Cb-U ALLOY CONTAINING Cb₂C PRECIPITATE
- C Cb-1 Zr ALLOY CONTAINING Cb₂C PRECIPITATE

CONDITION: STAIN-ETCHED

MAGNIFIED: 100X

SHOWING THE EFFECTS OF THE PRESENCE OF LITHIUM ON THE REACTION BETWEEN URANIUM CARBIDE (5-1 W/O C) AND Cb-1 Zr ALLOY AFTER 1000 HOURS AT 2200F

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URANIUM CARBIDE VS. TANTALUM-LINED Cb-1 Zr ALLOY
COMPATIBILITY SPECIMEN (LITHIUM ENVIRONMENT)

CONDITION: UNETCHED

MAGNIFIED: 200X

ILLUSTRATING THE ABILITY OF TANTALUM TO PREVENT
UC-Cb1 Zr INTERACTION AFTER 1000 HOURS EXPOSURE
AT 2200F

FIG 4

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