C70-799

~~CONFIDENTIAL~~

NASA CR-72666

(NASA-CR-72666) THERMAL CYCLING OF URANIUM
DIOXIDE-TUNGSTEN CERMET FUEL SPECIMENS
P.J. Gripshover, et al (Battelle Memorial
Inst.) 8 Dec. 1969 97 p

N72-75490

Unclas
00/99 45668

THERMAL CYCLING OF URANIUM DIOXIDE - TUNGSTEN
CERMET FUEL SPECIMENS

by

P. J. Gripshover and J. H. Peterson

prepared for

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

CONTRACT NAS-8568-H

NAS-8568

~~RESTRICTED DATA~~
~~ATOMIC ENERGY ACT OF 1954~~

BATTELLE
Memorial Institute

CLASSIFICATION CHANGED
TO...
By Authority of ID 7351 9/28/72

~~CONFIDENTIAL~~

X70-73337

(ACCESSION NUMBER)	(THRU)	(CODE)	(CATEGORY)
97			
(PAGES)	(NASA CR OR TMX OR AD NUMBER)	AVAILABLE TO U.S. GOVERNMENT AGENCIES AND CONTRACTORS ONLY	
NASA-CR-72666			

FF No. 602 (D)

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the National Aeronautics and Space Administration (NASA), nor any person acting on behalf of NASA:

- A.) Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or
- B) Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method or process disclosed in this report.

As used above, "person acting on behalf of NASA" includes any employee or contractor of NASA, or employee of such contractor, to the extent that such employee or contractor of NASA, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with NASA, or his employment with such contractor.

Requests for copies of this report should be referred to

National Aeronautics and Space Administration
Office of Scientific and Technical Information
Attention: AFSS-A
Washington, D.C. 20546

C70-799

~~CONFIDENTIAL~~

NASA CR-72666

THERMAL CYCLING OF URANIUM DIOXIDE - TUNGSTEN
CERMET FUEL SPECIMENS

by

P. J. Gripshover and J. H. Peterson

prepared for

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

December 8, 1969

NAS 3-8563
CONTRACT NAS-8563-H

Technical Management
NASA Lewis Research Center
Cleveland, Ohio
Nuclear Systems Division
Robert L. Brown

BATTELLE MEMORIAL INSTITUTE
505 King Avenue
Columbus, Ohio 43201

Battelle is not engaged in research for advertising, sales promotion, or publicity purposes, and this report may not be reproduced in full or in part for such purposes.

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

TABLE OF CONTENTS

	<u>Page</u>
SUMMARY	1
INTRODUCTION.	1
MATERIALS AND EQUIPMENT	2
EXPERIMENTAL WORK	3
Procurement of the Fuel and Coated Fuel Particles.	4
Fabrication of Phase I Fuel Pellets.	13
Thermal Cycling of the Phase I Pellets	18
Evaluation of Thermal Cycled Specimens - Phase I	19
PHASE II STUDIES.	53
Fabrication of Phase II Pellets.	54
Thermal Cycling of Phase II Pellets.	57
Evaluation of Thermal Cycles Specimens - Phase II.	60
CONCLUSIONS	82
REFERENCES.	84

~~CONFIDENTIAL~~

CONFIDENTIAL

LIST OF FIGURES

	<u>Page</u>
FIGURE 1. 30 MICRON DIAMETER UO_2 PARTICLES.	6
FIGURE 2. 30 MICRON DIAMETER CERIA STABILIZED UO_2 PARTICLES	6
FIGURE 3. 60 MICRON DIAMETER CERIA STABILIZED UO_2 PARTICLES	7
FIGURE 4. TYPE A MATERIAL AS COATED BY FIRST COATING SUBCONTRACTOR. . . .	10
FIGURE 5. DIAMETRAL GROWTH OF SELECTED PELLETS - PHASE I.	21
FIGURE 6. PELLETS AFTER THERMAL CYCLING TO FAILURE.	23
FIGURE 7. PELLETS AFTER THERMAL CYCLING TO FAILURE.	24
FIGURE 8. PELLETS B2 AND E1 AFTER 125 CYCLES.	25
FIGURE 9. PELLET A PRIOR TO THERMAL CYCLING	26
FIGURE 10. PELLET A1 AFTER 75 THERMAL CYCLES	27
FIGURE 11. PELLET A2 AFTER 25 THERMAL CYCLES	28
FIGURE 12. PELLET B PRIOR TO THERMAL CYCLING	29
FIGURE 13. BONDING BETWEEN MATRIX AND CLAD IN PELLET B	30
FIGURE 14. PELLET B1 AFTER 100 THERMAL CYCLES.	31
FIGURE 15. CLADDING FAILURE IN THIN END OF PELLET B1 AFTER 100 CYCLES. . .	32
FIGURE 16. PELLET B2 AFTER 125 THERMAL CYCLES.	33
FIGURE 17. PELLET B1-2 AFTER HEAT TREATMENT AT 2000 C FOR 5 HR IN VACUUM .	34
FIGURE 18. PELLET B1-1 AFTER 100 THERMAL CYCLES	35
FIGURE 19. PELLET B1-3 AFTER 75 THERMAL CYCLES	36
FIGURE 20. PELLET C PRIOR TO THERMAL CYCLING	37
FIGURE 21. BONDS IN PELLET C PRIOR TO THERMAL CYCLING.	38
FIGURE 22. PELLET C1 AFTER 50 THERMAL CYCLES	39
FIGURE 23. TYPE OF FAILURES OCCURRING BETWEEN FUEL FORM AND CLADDING AND IN THE THIN END PLUG OF PELLET C-1.	40

CONFIDENTIAL

CONFIDENTIAL

LIST OF FIGURES (continued)

	<u>Page</u>
FIGURE 24. PELLET D PRIOR TO THERMAL CYCLING.	41
FIGURE 25. PELLET D1 AFTER 5 THERMAL CYCLES	42
FIGURE 26. PELLET E PRIOR TO THERMAL CYCLING.	43
FIGURE 27. PELLET E1 AFTER 125 THERMAL CYCLES	44
FIGURE 28. PELLET E2 AFTER 100 THERMAL CYCLES	45
FIGURE 29. PELLET F PRIOR TO THERMAL CYCLING.	46
FIGURE 30. PELLET F1 AFTER 100 THERMAL CYCLES	47
FIGURE 31. PELLET F2 AFTER 75 THERMAL CYCLES.	48
FIGURE 32. PELLET G PRIOR TO THERMAL CYCLING.	49
FIGURE 33. PELLET G1 AFTER 25 THERMAL CYCLES.	50
FIGURE 34. DIAMETRAL GROWTH OF SELECTED PELLETS - PHASE II.	62
FIGURE 35. GROWTH CHARACTERISTICS OF PHASE I AND II UNCLAD FUEL FORMS . . .	64
FIGURE 36. PELLET A AFTER FAST AND EXTENDED THERMAL CYCLES.	65
FIGURE 37. PELLET E AFTER FAST AND EXTENDED THERMAL CYCLES.	66
FIGURE 38. BOND OF END PLUG TO TUNGSTEN CLAD IN PELLET E.	67
FIGURE 39. CRACK IN THICK END PLUG OF PELLET E AFTER THERMAL CYCLING. . . .	67
FIGURE 40. PELLET G AFTER FAST AND EXTENDED THERMAL CYCLES.	68
FIGURE 41. PELLET TYPE H PRIOR TO THERMAL CYCLING	69
FIGURE 42. PELLET H2 AFTER FAST AND EXTENDED THERMAL CYCLES	70
FIGURE 43. PELLET H1 AFTER 200 FAST THERMAL CYCLES.	71
FIGURE 44. BOND OF FUEL FORM TO CLAD IN PELLET H1 AFTER THERMAL CYCLING . .	72
FIGURE 45. BOND OF END PLUG TO CLAD AFTER THERMAL CYCLING OF PELLET H1. . .	72

CONFIDENTIAL

CONFIDENTIAL

LIST OF FIGURES (continued)

	<u>Page</u>
FIGURE 46. PELLETT K AFTER 25 FAST CYCLES AND 2 EXTENDED CYCLES.	73
FIGURE 47. FUEL FORM TYPE C AFTER 200 FAST THERMAL CYCLES	74
FIGURE 48. FUEL FORM TYPE D AFTER 200 FAST THERMAL CYCLES	75
FIGURE 49. FUEL FORM TYPE F AFTER 200 FAST THERMAL CYCLES	76
FIGURE 50. FUEL FORM TYPE G AFTER 200 FAST THERMAL CYCLES	77
FIGURE 51. FUEL FORM TYPE H AFTER 200 FAST THERMAL CYCLES	78
FIGURE 52. PELLETT H1 AFTER 200 THERMAL CYCLES AND PELLETT H3 AFTER 150 THERMAL CYCLES	79

CONFIDENTIAL

CONFIDENTIAL

LIST OF TABLES

	<u>Page</u>
TABLE 1. PELLETS TYPES AS SPECIFIED IN ORIGINAL CONTRACT.	5
TABLE 2. FUEL DATA FOR PHASE I PELLETS	8
TABLE 3. NEW PELLET TYPES AS SPECIFIED IN CONTRACT MODIFICATION.	11
TABLE 4. CHEMICAL ANALYSES OF ITEMS E, F, AND G.	12
TABLE 5. ANALYSES OF NASA-FURNISHED TUNGSTEN COATED UO_2	14
TABLE 6. DENSITY DATA ON CONSOLIDATED FUEL FORMS - PHASE I	16
TABLE 7. THERMAL CYCLE DATA - PHASE I.	20
TABLE 8. ANALYSES OF TUNGSTEN COATED FUEL PARTICLES - TYPE H	55
TABLE 9. PELLET AND FUEL FORM TYPES SCHEDULED FOR EVALUATION IN PHASE I. . .	58
TABLE 10. DESCRIPTION OF CLADDING DEFECTS PRIOR TO THERMAL CYCLING - PHASE II PELLETS.	59
TABLE 11. CLAD PELLET THERMAL CYCLE DATA - PHASE II	61
TABLE 12. FUEL FORM THERMAL CYCLE DATA - PHASE II	63

CONFIDENTIAL

~~CONFIDENTIAL~~

ABSTRACT

In phase I tungsten clad cermet fuel specimens were thermal cycled, to study the effects of fuel loading, fuel particle size, stabilized fuel, duplex coatings, and fabrication techniques on dimensional stability during thermal cycling. In phase II the best combination of the factors studied in phase I were combined in one specimen for evaluation.

~~CONFIDENTIAL~~

~~Confidential~~

SUMMARY

(CRD) This program was concerned with the fabrication, thermal cycling, and metallographic examination of tungsten clad cermet fuel pellets. The program was divided into two phases. In Phase I, seven pellet types were studied to determine the effect of fuel loading, fuel-particle size, stabilized fuel, duplex coatings, and fabrication techniques on the pellet growth characteristics. In Phase II the best combination of the factors studied in Phase I were combined into a single pellet type for further evaluation.

(CRD) The work in Phase I indicated that small UO_2 fuel particles (30 μ diam) which were stabilized with ceria performed the most satisfactorily. A fuel loading of about 50 v/o was found to be optimum. An observation was made that the pellets with lower density fuel forms from a previous study had more desirable growth characteristics. In Phase II the most desirable combination of factors studied in Phase I were evaluated in a newly fabricated group of pellets. These pellets showed no improvement in growth characteristics over the best Phase I pellets but did withstand 200 thermal cycles to 1800 C rather than the 125 cycles achieved in Phase I.

INTRODUCTION

(U) The Lewis Research Center of the National Aeronautics and Space Administration has conducted research on the fabrication and evaluation of tungsten cermet fuel pellets at Battelle's Columbus

~~Confidential~~

~~Confidential~~

2

Laboratories since July, 1963. During this period research on two contracts NAS3-4255 and NAS3-8563-H have been conducted at BCL and procedures have been developed for the fabrication, thermal cycling and evaluation of these fuel pellets. Contract NAS3-4255 was concerned with the fabrication of not only clad fuel pellets but also with the fabrication of containment-vessel assemblies.⁽¹⁾ These containment-vessel assemblies were subsequently operated and evaluated in the NASA-Plumbrook Reactor.

(CRD) As a result of these studies, additional work under this present contract (NAS3-8563-H) was initiated at BCL to further evaluate the growth and structural stability of these clad fuel pellets. During this program the first phase was concerned with the evaluation of the effect of fuel loading, stabilized fuel, fuel-particle size and duplex coatings on the thermal cycling, growth and structural characteristics of these pellets. In the second phase, the best combination of these variables was investigated by thermal cycling and subsequent evaluation.

(U) This final report on NASA Contract NAS3-8563-H covers all the work performed from the initiation of the program in July, 1966, to the conclusion in July, 1969.

MATERIALS AND EQUIPMENT

(U) The materials used in the preparation of the clad-fuel pellets consisted of tungsten cladding materials and coated fuel particles. A description of the coated fuel particles is given in the Experimental Work section of this report. The tungsten cladding components were fabricated from high-purity arc-cast tungsten rod.

~~Confidential~~

CONFIDENTIAL

3

(U) In addition to the above materials, gas-pressure-bonding containers for the fuel pellet were also utilized. Arc-cast molybdenum was selected for the bonding container because of its weldability and ease of removal from the pellet by selective leaching in dilute nitric acid. Tantalum was used to double-can the bonding container (i.e., to encase the molybdenum container to provide increased reliability during bonding).

(U) Equipment used during this program consisted of specialized gas-pressure-bonding equipment capable of temperatures to 3200 F and pressure in excess of 30,000 psi. Also utilized were machine-shop facilities, high-temperature vacuum and hydrogen furnaces, helium leak detectors, the metallographic laboratory, and spectrographic and chemical analysis facilities. Radiographic and dye-penetrant test equipment were used for nondestructive testing.

EXPERIMENTAL WORK

(CRD) The experimental work on this program was divided into two phases. In Phase I seven types of clad fuel pellets were fabricated, thermal cycled, and evaluated. These fuel pellets were so designed that the effect of fuel loading, fuel particle size, duplex coatings on the fuel particles, and fuel-core fabrication technique could be evaluated. From the data obtained in Phase I a selection of the "best" pellet type or combination of types would be made for further study in Phase II. The pellets fabricated in Phase II would be evaluated by thermal cycling and metallographic examination.

CONFIDENTIAL

~~CONFIDENTIAL~~

4

Phase I Studies

(U) The experimental work conducted in Phase I of this program consisted of procurement and analyses of fuel particles and coated fuel particles, fabrication of the clad pellets, thermal cycling of the pellets and evaluation of these pellets. These procedures and the results of this phase are discussed below.

Procurement of the Fuel and Coated Fuel Particles

(CRD) The initial contract prior to any modifications called for the procurement of materials by Battelle and fabrication of seven types of clad fuel pellets. These pellet types are shown in Table 1. The fuel for these pellets was ordered from NUMEC. Three types of fuel were ordered. These were 30 μ diameter UO_2 , 30 μ diameter UO_2 + 10 mole percent CeO_2 , and 60 μ diameter UO_2 + 10 mole percent CeO_2 . These particles were metallographically examined for free metal upon receipt from NUMEC and were submitted for determination of O/U ratio, chemical analysis, and density. Figures 1, 2, and 3 show the microstructures of these fuel particles. No free uranium was noted in any of the samples. However, the density of the 30 μ diameter UO_2 particles did not appear to be as high as the values stated by NUMEC. The density data, fuel particle size, and Battelle chemical analyses are given in Table 2. Both groups of ceria stabilized UO_2 samples contained about 500 ppm of Al which is probably in the form of alumina. NASA did not reject the fuel because of this impurity although a higher purity level would have been desirable. The density of the particles as determined by Battelle was lower than that

~~CONFIDENTIAL~~

CONFIDENTIAL

TABLE 1. PELLET TYPES AS SPECIFIED IN ORIGINAL CONTRACT

Pellet Type	Fuel	Fuel Size	Coating	Fluorine Content	Fuel Loading	Quantity
A	UO ₂	30 ± 10 μ	W-1/2 vapor ^(a)	<40 ppm	50 V/O (UO ₂)	5
B	UO ₂	30 ± 10 μ	W-1/2 vapor ^(a)	150-200 ppm	50 V/O (UO ₂)	3
C	UO ₂ + CeO ₂	30 ± 10 μ	W-all vapor	<40 ppm	73 V/O (ceramic)	3
D	UO ₂ + CeO ₂	30 ± 10 μ	W-all vapor ^(a)	<40 ppm	57 V/O (ceramic)	3
E	UO ₂ + CeO ₂	60 ± 10 μ	W-1/2 vapor ^(a)	<40 ppm	57 V/O (ceramic)	3
F	UO ₂	30 ± 10 μ	Mo & W ^(b)	<40 ppm	50 V/O (UO ₂)	3
G	UO ₂ + CeO ₂	30 ± 10 μ	W-1/2 vapor ^(a)	<40 ppm	57 V/O (ceramic)	3

(a) The remainder of the tungsten coating would consist of a uniform coating of tungsten powder.

(b) The coated particle would consist of the fuel coated with 1-2 microns of molybdenum followed by vapor coating with tungsten to obtain the desired fuel loading.

CONFIDENTIAL

~~CONFIDENTIAL~~

0345

6

1000X

As Polished

9A805

FIGURE 1. 30 MICRON DIAMETER UO_2 PARTICLES

1000X

As Polished

9A800

FIGURE 2. 30 MICRON DIAMETER CERIA STABILIZED UO_2 PARTICLES

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

0345

7

1000X

As Polished

9A803

FIGURE 3. 60 MICRON DIAMETER CERIA STABILIZED UO_2 PARTICLES

~~CONFIDENTIAL~~

CONFIDENTIAL

8

TABLE 2. FUEL DATA FOR PHASE I PELLETS

Element	Fuel Type		
	30 μ dia. UO ₂ ^(a)	30 μ dia. UO ₂ +CeO ₂	60 μ dia. UO ₂ +CeO ₂
Impurity Level - ppm			
B	0.3	<0.2	<0.2
Fe	5	<5	<5
Mn	<1	<1	<1
Mg	10	1	1
Cr	10	<5	<5
Si	5	5	10
Ni	<2	<2	<2
Al	100	500	500
Mo	5	50	50
Cu	1	15	1
Ca	100	70	50
Cl(b)	<10	<10	<10
F (b)	<10	<10	<10

Determination			
NUMEC density - per cent	96.8	99.5	97.8
Battelle density - per cent	92.8	94.9	95.8
Average particle size - diameter	35.8	36.7	62.1
Per cent of particles within $\pm 10\%$ of nominal diameter	96	86	88

(a) O/U ratio of 30 μ dia. UO₂ was determined by NUMEC to be 2.005; by Battelle to be 2.002.

(b) Halide analyses furnished by NUMEC.

CONFIDENTIAL

~~CONFIDENTIAL~~

9

determined by NUMEC but was still greater than the minimum density of 92 percent specified by the contract.

(U) The coating of the fuel particles was subcontracted to an organization other than NUMEC on the basis of cost and indicated ability to meet the halide impurity specification. These coating requirements are indicated in Table 1. The coating subcontract was terminated after approximately 9 months of effort because they were unable to furnish the required coated particles which would meet the specifications of the contract. In fact, none of the coated material appeared to have good adherent coatings and the fluorine content was very high--830 ppm in some samples. A photomicrograph of some Type A material is shown in Figure 4. Because of the inability of the subcontractor to furnish the coated material and the delay caused by the problem of obtaining the desired material, the program was modified so that NASA would furnish some of the coated material and Battelle would obtain the remainder of the coated particles from NUMEC. Sufficient material remained from the initial NUMEC fuel order to meet these requirements. The new pellet types as specified in the modified contract are shown in Table 3. The impurity content of the retained halides was relaxed to 75 ppm for these revised pellet types.

(U) An order was let to NUMEC to coat the last three types of material shown in Table 3. The first four types were furnished by NASA. The coated material from NUMEC was submitted for chemical analysis. The results of this work are shown in Table 4. Also included in this table are the halide analyses furnished by NUMEC with the material. In all cases the Battelle analyses showed higher values than those obtained by NUMEC. A sample of Type G was vacuum heat treated at 1500 C for 1 hr to determine the effect

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

0277

10

250X

Murakami's Etchant

4B124

FIGURE 4. TYPE A MATERIAL AS COATED BY FIRST COATING SUBCONTRACTOR

Note the poor bonding between coating layers.

~~CONFIDENTIAL~~

CONFIDENTIAL

TABLE 3. NEW PELLET TYPES AS SPECIFIED IN CONTRACT MODIFICATION

<u>Pellet Type</u>	<u>Fuel</u>	<u>Fuel Size</u>	<u>Coating</u>	<u>Fuel Loading</u>	<u>Quantity</u>
A	UO ₂	38-78	30 v/o W ^(a)	50 v/o UO ₂	3
B	UO ₂	38-78	50 v/o W	50 v/o UO ₂	5
C	UO ₂	38-78	30 v/o W	70 v/o UO ₂	3
D	UO ₂	78-150	50 v/o W	50 v/o UO ₂	3
E	UO ₂ +CeO ₂	30±10	50 v/o W	50 v/o Ceramic	3
F	UO ₂	30±10	50 v/o Mo+W	50 v/o UO ₂	3
G	UO ₂ +CeO ₂	60±10	50 v/o W	50 v/o Ceramic	3

(a) Tungsten powder to be added as a uniform coating to obtain desired fuel loading.

CONFIDENTIAL

~~CONFIDENTIAL~~

12

TABLE 4. CHEMICAL ANALYSES OF ITEMS E, F, and G

Element	Impurity Level-ppm		
	Item E	Item F	Item G
Fe	20	60	5
Mn	< 1	< 1	< 1
Mg	< 1	< 1	< 1
Al	5	10	10
Mo	100	(a)	< 10
Cu	10	3	3
Ni	20	100	10
Ca	< 1	< 1	< 1
Cr	< 10	< 10	< 10
F(b)	56	41	79
Cl(b)	135	88	167
F(c)	19	10	43
Cl(c)	19	64	19

- (a) Contains molybdenum coating over UO_2 substrate
(b) BM1 determinations
(c) NUMEC determinations

~~CONFIDENTIAL~~

CONFIDENTIAL

13

of this treatment on the halide content. After treatment, chlorine was reduced from 167 ppm to 53 ppm and fluorine from 79 ppm to 68 ppm. A sample of this same material was sent to the AEC New Brunswick Laboratory, Brunswick, New Jersey, for analysis. Their results indicated a chlorine content of 10 ppm and a fluorine content of 120 ppm. Comparing these data with the values obtained by NUMEC and Battelle (Table 4) it would appear that the Battelle chlorine values are high and there is no close agreement among any of the three fluorine values. As a result of these higher fluorine values NASA again relaxed the retained halide specification and all material was accepted for use on the program.

(U) The material for the next four pellet types was received from NASA and analyzed for chlorine and fluorine content. The results are shown in Table 5. All material met the desired specification of <75 ppm halides except for part of the material required for Pellet D. This did, however, fall within the new specification received from NASA. It was decided by Battelle and NASA that all material would be vacuum treated at 1500 C for 1 hr to reduce the halide content and provide uniform processing for all the material.

Fabrication of Phase I Fuel Pellets

(CRD) All the fuel forms with the exception of Type A consisted entirely of coated particles. The Type A pellets were similar to those fabricated on the previous program. The coated particles for the Type A pellets had a 30 v/o W coating. Tungsten fines (<1 μ diameter particle size) were added to these particles by wax-coating techniques developed on the previous contract to provide a final fuel loading of 50 v/o UO_2 .

CONFIDENTIAL

~~CONFIDENTIAL~~

14

TABLE 5. ANALYSES OF NASA-FURNISHED TUNGSTEN COATED UO_2

Delivery Identification	Contract Identification	Impurity Level-ppm	
		Cl	F
1	Item D	106	43
2	Item D	10	53
3	Item B	< 5	< 10
1A	Items A & C	34	< 10

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

15

These fuel forms were cold compacted and vacuum outgassed at 1500 C for 1 hr. The coated particles for the other fuel forms were also vacuum treated at 1500 C for 1 hr prior to loading into molybdenum pressing containers. The loaded containers for all seven pellet types were outgassed at 2000 F in vacuum for 1 hr, backfilled with argon after cool-down, evacuated, and sealed in an electron beam welder. These molybdenum containers were then double canned in tantalum to enhance reliability during consolidation. The consolidation parameters for these specimens were 2900 F and 15,000 psi for a period of 3 hr. The containers were removed from the fuel forms by selective leaching. Following visual inspection, the fuel forms were ground to dimensions of 0.460-in. diameter by 0.980 in. long. Densities of the seven fuel form types were determined and are listed in Table 6. Practically no variation in density existed within a given type.

(U) During the period that the coated particles were being obtained and the fuel forms consolidated, the tungsten cladding materials were being prepared. The procedures for this had been developed on Contract No. NAS3-4255.⁽¹⁾ All the tungsten material was high-purity, arc-cast tungsten and was described in the Materials and Equipment section of this report. The rod for the tubular portion of cladding was purchased from Climax Molybdenum and had an impurity level of about 300 ppm molybdenum. The material used for the end plugs was obtained from Universal Cyclops.

(U) The rod for the tubular portion of the cladding was nominally 9/16-in. diameter. A through hole, 0.445 in. in diameter was EDM'd through a piece 1.200 in. long. Following a stress relief, the tubes were honed to an inside diameter of 0.462 in. and were ground on the OD to

~~CONFIDENTIAL~~

CONFIDENTIAL

16

TABLE 6. DENSITY DATA ON CONSOLIDATED FUEL
FORMS - PHASE I

Fuel Form Identification	Average Density Percent of Theoretical
A	95.7
B	95.2
C	95.4
D	96.2
E	92.7
F	95.4
G	94.7

CONFIDENTIAL

CONFIDENTIAL

17

0.530 in. End plugs, 0.460 in. in diameter were prepared by grinding from 1/2-in.-diameter rod.

(CRD) The fuel forms and the tungsten cladding components were prepared for gas-pressure bonding by hydrogen heat treatment prior to assembly. The fuel forms were first given a 1-minute dip in dilute nitric acid to remove any smeared UO_2 . The fuel forms were hydrogen treated at 1750 to 1800 C for 1-1/2 hr. Heat-up and cool-down was accomplished at 500 C per hr. After treatment it was noted that the Type F fuel forms had increased in diameter and length. The diameter had increased to 0.465 in. from 0.460 in. Therefore, the tungsten-cladding components for the Type F pellets were honed on the ID to 0.467 in. diameter. New end plugs were ground to 0.465 in. diameter. The tungsten cladding components were then hydrogen treated at 1850 F for 1-1/2 hr. The lower temperature was used to reduce the amount of grain growth in the material that would have occurred at the 1800 C treatment temperature. The molybdenum bonding containers were also heat treated with the tungsten-cladding components to provide a very clean bonding system.

(CRD) The fuel forms and the tungsten-cladding components were assembled and placed in the bonding containers, welded in an inert gas chamber, and vacuum treated at 2000 F for 1 hr. After cooldown the vacuum chamber was backfilled with argon and the specimens immediately placed in an electron beam welder and evacuated. After four hr under vacuum the containers were sealed by the electron beam. After leak checking, the molybdenum containers were double canned in tantalum. Bonding conditions for these specimens were 3150 to 3200 F for 1-1/2 hr at a gas pressure of 30,000 psi. The gas was bled from the autoclave at a rate of 100 psi/min while maintaining temperature.

CONFIDENTIAL

~~CONFIDENTIAL~~

18

Inspection of the containers indicated that all containers had deformed satisfactorily. Removal of the pellets from the bonding containers was accomplished by selective leaching.

(U) The pellets were visually inspected and no apparent defects were evident. The clad pellets were then ground to provide a cladding thickness of 0.018-0.022 in. on the diameter and one end. The other end was ground to provide a clad thickness of 0.125 in. Following grinding, the pellets were inspected by Zyglo and helium leak check. No defective pellets were found.

(U) A preliminary metallographic examination was made on one pellet each of Types B and F prior to the initiation of thermal cycling. In both cases, the fuel form was well bonded to the unfueled tungsten cladding. The tungsten-to-tungsten bonds at the joints between the end plugs and the exterior tube were well bonded with no noticeable voids.

Thermal Cycling of the Phase I Pellets

(U) Two pellets of each type fabricated plus two Type B pellets, which were to be heat treated at 2000 C in vacuum for 4 to 6 hr, were selected for thermal cycling. The heat treatment for the two Type B pellets consisted of a vacuum heat treatment at 2000 C for 5 hr. These pellets, which were now designated as Type B1, were inspected and one was cracked on the thin end. Another Type B pellet was given an identical heat treatment and it also cracked in a similar manner. This cracking is discussed in the Phase II section of this report.

(U) The thermal cycling of the pellets was initiated using the thermal cycle as specified in the contract. This consisted of the following:

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

19

1. Heat to 1800 C in vacuum in 20 min.
2. Hold at temperature for 5 min.
3. Cool to 1200 C in 5 min. in vacuum.
4. Cool to 700 C in 5 min. in static helium.
5. Cool to 200 C or below in 5 min. in flowing helium.

Inspection of the pellets would be made after 5, 10, 15, 25, 50, 75, 100, 125, 150, 175, and 200 cycles. This inspection consisted of weighing and measuring each pellet to determine changes as a result of the thermal cycling, helium leak checking, and finally dye-penetrant inspection for cracks. The helium leak check as conducted by Battelle consisted of subjecting the pellets to an external helium pressure (50 psi for 10 min), removing the pellets from the pressurizing chamber, and placing in a vacuum chamber which was directly attached to a mass spectrometer.

(U) The thermal cycle data obtained during this phase for each pellet is listed in Table 7. These data are graphically shown in Figure 5; included with these growth curves are growth data from two specimens on the previous contract. Pellet T2 is the tungsten-clad pellet from the third thermal-cycle experiment and Pellet E7 is a typical enriched pellet from the previous contract.

Evaluation of Thermal Cycled Specimens - Phase I

(U) After thermal cycling the pellets were sectioned so as to remove a thin transverse section from the center of the pellet. Each of the remaining ends were then sectioned longitudinally so that the bonds at the interface between the end plug and the tubular section of the cladding could be examined. One pellet of each type which had not been thermal

~~CONFIDENTIAL~~

TABLE 7. THERMAL CYCLE DATA - PHASE I

Pellet Identification	Percent Growth							
	5 Cycles	10 Cycles	15 Cycles	25 Cycles	50 Cycles	75 Cycles	100 Cycles	125 Cycles
A1 Diameter	0.56	0.97	1.27	1.92	3.16	4.24 ^(a)	--	--
A1 Length	0.50	0.79	1.05	1.54	2.45	3.23	--	--
A2 Diameter	0.52	0.88	1.21	1.79 ^(a)	--	--	--	--
A2 Length	0.46	0.76	0.99	1.45	--	--	--	--
B1 Diameter	0.06	0.12	0.22	0.38	0.91	1.55	2.25 ^(a)	--
B1 Length	-0.05	-0.09	-0.10	-0.10	0	0.17	0.42	--
B2 Diameter	0.02	0.10	0.18	0.32	0.77	1.35	1.97	2.84 ^(a)
B2 Length	-0.05	-0.06	-0.06	-0.05	0.09	0.36	0.65	1.07
B1-1 Diameter	0.04	0.14	0.22	0.42	0.91	1.55	2.13 ^(a)	--
B1-1 Length	-0.01	0	0.03	0.09	0.31	0.58	0.95	--
B1-3 Diameter	0.04	0.10	0.18	0.38	0.91	1.53 ^(a)	--	--
B1-3 Length	0.02	0.05	0.05	0.07	0.31	0.62	--	--
C1 Diameter	0.06	0.16	0.28	0.54	1.75 ^(a)	--	--	--
C1 Length	0	0.07	0.15	0.38	1.43	--	--	--
C2 Diameter	0.04	0.12	0.20	0.42	1.57 ^(a)	--	--	--
C2 Length	0	0.04	0.10	0.28	1.07	--	--	--
D1 Diameter	0.30 ^(a)	--	--	--	--	--	--	--
D1 Length	0.35 ^(a)	--	--	--	--	--	--	--
D2 Diameter	0.28	--	--	--	--	--	--	--
D2 Length	0.27	--	--	--	--	--	--	--
E1 Diameter	0.14 ^a	0.20	0.30	0.44	0.85	1.33	1.93	2.39 ^(a)
E1 Length	0.13	0.16	0.19	0.33	0.63	1.08	1.52	1.85
E2 Diameter	0.16	0.24	0.28	0.46	0.93	1.49	2.12 ^(b)	--
E2 Length	0.10	0.14	0.18	0.27	0.61	1.04	1.47	--
F1 Diameter	0.74	1.28	1.74	2.62	4.07	5.39	6.56 ^(a)	--
F1 Length	0.65	1.14	1.54	2.20	3.28	4.05	4.72	--
F2 Diameter	0.68	1.18	1.15	2.54	4.09	5.46 ^(a)	--	--
F2 Length	0.65	1.16	1.59	2.39	3.52	4.58	--	--
G1 Diameter	0.18	0.32	0.48	0.56 ^(a)	--	--	--	--
G1 Length	0.14	0.29	0.37	0.64	--	--	--	--
G2 Diameter	0.18	0.34	0.50	0.88 ^(a)	--	--	--	--
G2 Length	0.17	0.27	0.34	0.61	--	--	--	--

(a) Removed from thermal cycling due to cracking defect.

(b) Removed from thermal cycling for metallographic examination.

CONFIDENTIAL

CONFIDENTIAL

~~CONFIDENTIAL~~

21

~~CONFIDENTIAL~~

FIGURE 5. DIAMETRAL GROWTH OF SELECTED PELLETS - PHASE I

Pellet AI was heat treated at 2000 C for 24 hr after 25 cycles. Pellet T2 is the tungsten clad pellet from the third thermal cycle experiment on Contract NAS 3-4255. Pellet E7 is a typical enriched pellet from this previous contract.

~~CONFIDENTIAL~~

22

cycled was also examined in this manner. One pellet, designated as B1-2, which had been heat treated at 2000 C for 1 hr but not thermal cycled due to cracking, was also examined.

(CRD) The external appearance of these pellets prior to metallographic examination is shown in Figures 6, 7, and 8. The photomicrographs of these specimens are shown in Figures 9 through 33. Examination of these photomicrographs and the corresponding growth curves resulted in the following conclusions concerning the thermal cycling and subsequent results of the pellets fabricated in Phase I.

- (1) Larger fuel particles cause cladding cracks to develop with fewer thermal cycles and less growth. This is shown by comparing pellets B and D and pellets E and G. The larger size particles contained in pellets D and G were removed from cycling due to cracking after 5 and 25 cycles, respectively.
- (2) Deterioration of the matrix occurs more rapidly at higher fuel loadings. This can be shown by comparing pellet types B and C. The higher fuel loading also results in weak fuel form to cladding bonds due to the small amount of matrix available for bonding to the cladding.
- (3) The use of molybdenum in the tungsten matrix results in more rapid growth during cycling. However, more growth can occur before failure than with the other pellets evaluated.

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

23

1.75X

42256

FIGURE 6. PELLETS AFTER THERMAL CYCLING TO FAILURE

c 345

CONFIDENTIAL

1.75X

FIGURE 7. PELLETS AFTER THERMAL CYCLING TO FAILURE

42257

24

CONFIDENTIAL

~~CONFIDENTIAL~~

0345

25

1.75X

42425

FIGURE 8. PELLETS B2 AND E1 AFTER 125 CYCLES

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

0340

26

100X

a. Murakami's Etch

9C666

500X

b. Murakami's Etch

9C691

FIGURE 9. PELLET A PRIOR TO THERMAL CYCLING

The Type A fuel pellets consist of 50 v/o UO_2 , 22.7 v/o tungsten coating and 27.3 v/o tungsten powder. Fuel particle size is 38-78 μ diameter.

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

0345

27

100X

a. Murakami's Etch

9C667

500X

b. Murakami's Etch

9C692

FIGURE 10. PELLET A1 AFTER 75 THERMAL CYCLES

This pellet was heat treated for 24 hr
at 2000 C in vacuum after 25 cycles.

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

28

100X

a. Murakami's Etch

9C668

500X

b. Murakami's Etch

9C693

FIGURE 11. PELLET A2 AFTER 25 THERMAL CYCLES

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

0345

29

100X

a. Murakami's Etch

9C669

500X

b. Murakami's Etch

9C694

FIGURE 12. PELLET B PRIOR TO THERMAL CYCLING

The Type B fuel pellets consist of approximately 50 v/o UO_2 and 50 v/o tungsten coating. Fuel particle size is 38-78 μ diameter.

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

0345

30

500X

Murakami's Etchant

9C688

FIGURE 13. BONDING BETWEEN MATRIX AND CLAD IN PELLET B

This is typical of the bonds obtained in the Phase I pellets.

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

0345

31

100X

a. Murakami's Etch

9C670

500X

b. Murakami's Etch

9C695

FIGURE 14. PELLET B1 AFTER 100 THERMAL CYCLES

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

0240

32

30X

Murakami's Etchant

9C714

FIGURE 15. CLADDING FAILURE IN THIN END OF PELLET B1
AFTER 100 CYCLES

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

33

C345

100X

a. Murakami's Etch

9C674

500X

b. Murakami's Etch

9C699

FIGURE 16. PELLET B2 AFTER 125 THERMAL CYCLES

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

1345

34

100X

a. Murakami's Etch

9C671

500X

b. Murakami's Etch

9C696

FIGURE 17. PELLET B1-2 AFTER HEAT TREATMENT AT 2000 C
FOR 5 HR IN VACUUM

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

1345

35

100X

a. Murakami's Etch

9C672

500X

b. Murakami's Etch

9C697

FIGURE 18. PELLET B1-1 AFTER 100 THERMAL CYCLES

This pellet was vacuum heat treated
at 2000 C for 5 hr prior to thermal
cycling

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

0345

36

100X

a. Murakami's Etch

9C673

500X

b. Murakami's Etch

9C698

FIGURE 19. PELLET B1-3 AFTER 75 THERMAL CYCLES

This pellet was vacuum heat treated
at 2000 C for 5 hr prior to thermal
cycling.

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

0345-

37

100X

a. Murakami's Etch

9C675

500X

b. Murakami's Etch

9C700

FIGURE 20. PELLET C PRIOR TO THERMAL CYCLING

The Type C fuel pellets consist of 68.6 v/o UO_2 and 31.2 v/o tungsten coating. The fuel particle size is 38-78 μ diameter.

~~CONFIDENTIAL~~

FIGURE 21. BONDS IN PELLET C PRIOR TO THERMAL CYCLING

Note the excellent bonding between the cladding and the matrix and the end plug - tubular clad interface.

~~CONFIDENTIAL~~

0345

39

100X

a. Murakami's Etch

9C676

500X

b. Murakami's Etch

9C701

FIGURE 22. PELLET C1 AFTER 50 THERMAL CYCLES

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

0345

40

30X

Murakami's Etchant

9C716

FIGURE 23. TYPE OF FAILURES OCCURRING BETWEEN FUEL FORM AND CLADDING AND IN THE THIN END PLUG OF PELLET C-1

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

0345

41

100X

a. Murakami's Etch

9C677

500X

b. Murakami's Etch

9C702

FIGURE 24. PELLET D PRIOR TO THERMAL CYCLING

The Type D fuel pellets consist of approximately 50 v/o UO_2 and 50 v/o tungsten coating. The fuel particle size is 78-150 μ diameter.

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

0345

42

100X

a. Murakami's Etch

9C678

500X

b. Murakami's Etch

9C703

FIGURE 25. PELLET D1 AFTER 5 THERMAL CYCLES

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

0345

43

100X

a. Murakami's Etch

9C679

500X

b. Murakami's Etch

9C704

FIGURE 26. PELLET E PRIOR TO THERMAL CYCLING

The Type E fuel pellets consist of 47.5 v/o ceria stabilized UO_2 and 52.5 v/o tungsten coating. The fuel particle size is 20-40 μ diameter.

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

C345

44

100X

a. Murakami's Etch

9C680

500X

b. Murakami's Etch

9C705

FIGURE 27. PELLET E1 AFTER 125 THERMAL CYCLES

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

0345

45

100X

a. Murakami's Etch

9C681

500X

b. Murakami's Etch

9C706

FIGURE 28. PELLET E2 AFTER 100 THERMAL CYCLES

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

0345

46

100X

a. Murakami's Etch

9C682

500X

b. Murakami's Etch

9C707

FIGURE 29. PELLET F PRIOR TO THERMAL CYCLING

The Type F fuel pellets consist of 47.2 v/o UO_2 , a 16.6 v/o molybdenum coating on the UO_2 particle and a final coating of 36.2 v/o tungsten. The fuel particle size is 20-40 μ diameter.

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

0345

47

100X

a. Murakami's Etch

9C683

500X

b. Murakami's Etch

9C708

FIGURE 30. PELLET F1 AFTER 100 THERMAL CYCLES

~~CONFIDENTIAL~~

0345

48

100X

a. Murakami's Etch

9C684

500X

b. Murakami's Etch

9C709

FIGURE 31. PELLET F2 AFTER 75 THERMAL CYCLES

100X

a. Murakami's Etch

9C685

500X

b. Murakami's Etch

9C710

FIGURE 32. PELLET G PRIOR TO THERMAL CYCLING

The Type G fuel pellets consist of 52 v/o ceria stabilized UO_2 and 48 v/o tungsten coating. The fuel particle size is 50-70 μ diameter

~~CONFIDENTIAL~~

0345

50

100X

a. Murakami's Etch

9C686

500X

b. Murakami's Etch

9C711

FIGURE 33. PELLET G1 AFTER 25 THERMAL CYCLES

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

- (4) The use of ceria-stabilized fuel particles results in less matrix deterioration during thermal cycling. This can be illustrated by comparing pellet types B and E. Type G pellets which, like the type E pellets, contain stabilized fuel also show very little deterioration although these pellets received only 25 thermal cycles. The pellets which do not contain ceria have deteriorated more in all cases but not in the same manner. Types B and C appear to have been attacked at the tungsten grain boundaries and this in turn results in large voids at these locations. However, pellets A and F do not show this type of behavior but appear to have had considerable readjustment of the tungsten resulting in areas that are completely lacking in tungsten between UO_2 particles and other areas which show increases in tungsten thickness.
- (5) The use of powder to form part of the tungsten matrix results in greater growth during thermal cycling. This is shown by comparing pellet types A and B. A possible cause may be a nonuniform distribution of tungsten which causes some thin ribs and, therefore, a weaker structure than that obtained by applying all the tungsten by vapor deposition. However, pellet type C, which contains the same coated particles as pellet type A and would have ribs as thin or thinner, did not show the rapid rate of

~~CONFIDENTIAL~~

~~Confidential~~

52

growth of pellet Type A during thermal cycling. Even though the fuel and tungsten coating were the same for pellet Types A and C, the type of matrix deterioration was different. The type of deterioration in the matrix is very similar in Types A and F which showed the most rapid rates of growth during thermal cycling. No explanation for the differences in matrix deterioration of the unstabilized fueled pellet is offered, but it must be recognized that different modes of deterioration exist. Because Types A and C were made from the same lot of material and the addition of tungsten fines would not have introduced any additional halides, differences in fluorine and chlorine content would probably not be the dominant factor.

- (6) Another observation is that the probably densities after fabrication and prior to thermal cycling of Pellet Type A from this contract and Types T2 and E7 from the previous contract were not the same. In all three cases the pellets were fabricated in much the same manner with the exception of the final bonding conditions which would control the final density since these were the maximum temperature and pressure parameters. All three pellet types contained coated particles supplemented by the addition of tungsten fines to bring the fuel loading to 50 v/o. The fuel was not stabilized in any of the

~~Confidential~~

~~CONFIDENTIAL~~

three cases. Bonding conditions for Pellet T2 were 3100-3200 F and 16,000 psi for 1-1/2 hr; conditions for Pellet E7 were 3150-3200 F and 20,000 psi for 2 hr; and conditions for Pellet A of this contract were 3150-3200 F and 30,000 psi for 1-1/2 hr. By comparing the growth curves in Figure 5 it can be seen that the higher the bonding pressure the greater the growth of the fuel pellet. It can be assumed that since the bonding temperature for these three similar pellet types was the same, the density would be controlled solely by the compacting pressure.

Phase II Studies

(CRD) As a result of the above conclusions Phase II of this program was designed to study the effect of bonding parameters, extended hold times during the thermal cycling, and stabilized fuel on the growth characteristics and microstructure of the clad fuel pellets. In Phase II three types of pellets were scheduled for fabrication. These were:

- (1) Pellet Type H which would be the same as Type E of Phase I.
- (2) Pellet Type J which would be fabricated using a spare Type A core from Phase I.
- (3) Pellet Type K which would be fabricated using a spare Type E core from Phase I.

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

54

Bonding conditions for these would be 3150 F and 16,000 psi for a period of 1-1/2 hr. These conditions would duplicate the bonding parameters of the third thermal cycle experiment from the previous contract which gave the most desirable growth characteristics of any pellet type evaluated on either this contract or the previous contract.

(U) The thermal cycling of the pellets was of two different types. The fast cycle used in Phase I was again the primary means of evaluation, but an extended cycle was also used for selected pellets. The particular details relating to those thermal cycles are discussed in another section of this report. The means for evaluation during thermal cycling and after thermal cycling were the same as those used in Phase I.

Fabrication of Phase II Pellets

(U) The fabrication procedures for the Phase II pellets were identical to those used in Phase I with the exception of the final bonding cycle. The fuel used for the Type H pellets was the same as that used for the Type E pellets in Phase I. NUMEC supplied the coating services for this fuel and the analyses of the coated particles is given in Table 8. The retained halides met the requirements of the contract and good agreement was obtained between the mass spectrographic and chemical techniques.

(U) The consolidation of the Type H fuel forms and the subsequent preparation of these fuel forms, the fuel forms for Types J and K pellets, and the tungsten cladding components were handled in exactly the same manner described in Phase I of this report. The density of the Type H fuel forms after consolidation was 92.7 percent which compared favorably with the Type E fuel forms of Phase I. Prior to the high temperature hydrogen

~~CONFIDENTIAL~~

TABLE 8. ANALYSES OF TUNGSTEN COATED FUEL PARTICLES - TYPE H

Element	Impurity Level, ppm	
Na	6	
Mg	6	
Al	30	
Si	20	
P	3	
S	6	
Ca	40	
Cr	50	
Mn	6	
Fe	150	
Ni	20	
Cu	1	
Zr	30	
Mo	300	
La	10	
Cl	30(a)	30(b)
F	20(a)	42(b)

(a) Cl and F as determined by mass spectrographic analysis.

(b) Cl and F as determined by chemical analysis.

~~Confidential~~

56

treatment for the fuel forms (Types H, J, and K), they were lightly abraded and dipped in a 20 percent nitric acid solution. At this time small microcracks were noted on the ends of some of Type H fuel forms. Additional abrading of the fuel forms did not appear to remove the cracks. Of the five Type H fuel forms fabricated, one did not show cracks, one showed cracks only on one end and the remaining three showed cracks on both ends. The high-temperature hydrogen treatment did not appear to change the cracking in any manner.

(CRD) The consolidation run for the clad fuel pellets was made at parameters of 3150 F and 16,000 psi for a period of 1-1/2 hr. Toward the end of the pressure release the furnace temperature dropped to about 2800 F rather than being maintained at the desired 3100-3200 F level. This lower temperature is in excess, however, of the 2500 F which is considered the minimum desired temperature while still under pressure. At 2500 F a tensile stress of 1000 psi completely relaxes in 5 min., so full relaxation of the stresses in the pellets did occur. Following the bonding runs the specimens were removed from their containers by selective leaching and ground to final dimensions.

(U) The pellets scheduled for evaluation in Phase II consisted of four Type H pellets, one Type J pellet, one Type K pellet, and one each of the following pellet types remaining from Phase I - Types A, C, E, and G. Because of a crack in the exterior cladding of the Type C pellet, the clad was removed and it was reclad with the Phase II pellets. Thermal cycling of some fuel forms from both Phases I and II was also scheduled because a few extras were made but had not been clad. These fuel forms included Types C, D, F, G, and H. In

~~Confidential~~

~~CONFIDENTIAL~~

57

Table 9 is given a listing of the Pellet and fuel form types which were scheduled for evaluation in Phase II.

(U) Following grinding of the pellets clad in the Phase II bonding cycle, inspection of all pellets scheduled for thermal cycling was made. During this inspection it was determined that small cracks in the tungsten clad could possibly be concealed by smeared metal on the surface. (This is possibly the reason for two of the Type B1 pellets in Phase I showing cracks after the 2000 C heat treatment.) This smeared metal was removed by lightly abrading the surface and then placing the pellet in a solution of Murakami's etchant for about 5 min. Pellet Types C, J, and one of the H pellets designated as H4 had obvious cracks in the as-ground condition. The remaining pellets were given the light abrading and etching described above to expose cracks. An evaluation of the Phase II pellets is given in Table 10. A very thin transverse section of Type J pellet was removed from the cracked end to determine if further processing of this pellet was advisable. It was determined by NASA that no further work would be done on this pellet because some deterioration of the cermet fuel form had already begun to occur. The pellets for thermal cycle evaluation were Types A, E, G, H1, H2, H3, and K.

Thermal Cycling of Phase II Pellets

(CRD) As mentioned previously two types of thermal cycles were to be used. Pellets H1 and H3, as well as all the unclad cores were to be thermal cycled with the standard fast cycle used in Phase I with inspection at 10, 25, 50, 75, 100, 125, 150, 175, and 200 cycles. Pellets A, E, G, H2, and K would receive 50 fast cycles but would also receive an extended thermal cycle between fast cycles 10, 11, 25, and 26, and after fast cycle

~~CONFIDENTIAL~~

TABLE 9. PELLETT AND FUEL FORM TYPES SCHEDULED FOR
EVALUATION IN PHASE II

Clad Pellet Type ^(a)	Fuel	Fuel Size Dia-Microns	Coating	Fuel Loading	Quantity
H	UO ₂ + CeO ₂	30±10	50 v/o W	50 v/o Ceramic	4
J	UO ₂	38-78	30 v/o W ^(b)	50 v/o UO ₂	1
K	UO ₂ + CeO ₂	30±10	50 v/o W	50 v/o Ceramic	1
A	UO ₂	38-78	30 v/o W ^(b)	50 v/o UO ₂	1
C	UO ₂	38-78	30 v/o W	70 v/o UO ₂	1
E	UO ₂ + CeO ₂	30±10	50 v/o W	50 v/o Ceramic	1
G	UO ₂ + CeO ₂	60±10	50 v/o W	50 v/o Ceramic	1
<u>Fuel Form Type (c)</u>					
C	UO ₂	38-78	30 v/o W	70 v/o UO ₂	1
D	UO ₂	78-150	50 v/o W	50 v/o UO ₂	1
F	UO ₂	30±10	50 v/o Mo + W	50 v/o UO ₂	1
G	UO ₂ + CeO ₂	60±10	50 v/o W	50 v/o Ceramic	1
H	UO ₂ + CeO ₂	30±10	50 v/o W	50 v/o Ceramic	1

(a) Clad Fuel Pellet Types H, J, and K were clad in Phase II; Types A, C, E, and G, were left from Phase I.

(b) Tungsten powder added as a uniform coating to obtain desired fuel loading.

(c) All fuel forms except Type H were left from Phase I.

~~CONFIDENTIAL~~

TABLE 10. DESCRIPTION OF CLADDING DEFECTS PRIOR TO
THERMAL CYCLING - PHASE II PELLETS

Pellet Identification	Description of Defects
A	A very small defect on thick end
C	Poor bonding between tungsten end plugs and cladding - not cycled
E	Four very fine cracks on thick end. The longest crack extending about one-third of the way across end
G	One crack extending about one-third of the way across thin end
H1	No defects
H2	No defects
H3	Very faint dye-penetrant indications (widely separated dots) at the bond interface of the end plugs to the clad on both the thick and thin ends
H4	Crack on thin end. This pellet will be used for metallographic examina- tion of Type H pellets prior to thermal cycling.
J	Crack across thin end - not cycled
K	Crack across thin end

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

50. Thermal cycling would then be terminated on these pellets. Inspection of these pellets would be made before and after each extended cycle. The extended thermal cycle consisted of a hold time of 50-100 hr at 1800 C. The purpose of this extended cycle was to determine if any of the undesirable growth that had occurred during the fast cycle could be recovered during a long hold time at temperature.

(U) The growth characteristics of the clad fuel pellets are shown in Table 11. These data are plotted graphically in Figure 34. The growth of the unclad fuel forms are given in Table 12. These data are presented in a graphical manner in Figure 35.

Evaluation of Thermal Cycles Specimens - Phase II

(U) After removal from thermal cycling the clad pellets were sectioned in the same manner as those in Phase I. The unclad fuel forms were examined metallographically on a transverse section only. The photomicrographs of these specimens are shown in Figures 36 through 51. In Figure 52 are shown Pellets H1 and H3 after thermal cycling.

(CRD) Examination of these photomicrographs and comparison with the thermal cycle data from Phase I resulted in the following conclusions:

- (1) No marked change in the growth during thermal cycling was gained from the extended thermal cycles. The growth characteristics of the pellets having the extended thermal cycle were not substantially different from those receiving the fast cycles only.
- (2) The structure of the fuel and the matrix in the pellets that received the extended thermal cycle

~~CONFIDENTIAL~~

TABLE 11. CLAD PELLET THERMAL CYCLE DATA - PHASE II

Pellet Identification	Percent Growth											
	10 Fast Cycles	First Extended Cycle	25 Fast Cycles	Second Extended Cycle	50 Fast Cycles	Third Extended Cycle	75 Fast Cycles	100 Fast Cycles	125 Fast Cycles	150 Fast Cycles	175 Fast Cycles	200 Fast Cycles
A Diameter	0.86	0.92	1.73	1.75	3.00	2.96	--	--	--	--	--	--
A Length	0.70	0.81	1.46	1.50	2.48	2.48	--	--	--	--	--	--
E Diameter	0.18	0.16	0.44	0.44	0.97	0.97	--	--	--	--	--	--
E Length	0.17	0.16	0.41	0.39	0.89	0.89	--	--	--	--	--	--
G Diameter	0.28	0.30	0.84	0.86	1.81	1.81	--	--	--	--	--	--
G Length	0.25	0.29	0.67	0.67	1.31	1.34	--	--	--	--	--	--
H1 Diameter	0.32	--	0.68	--	1.30	--	1.88	2.34	2.73	3.06	3.49	3.78
H1 Length	0.24	--	0.53	--	1.06	--	1.50	1.85	2.12	2.38	2.67	2.92
H2 Diameter	0.27	0.27	0.70	0.64	1.44	1.40	--	--	--	--	--	--
H2 Length	0.22	0.21	0.53	0.53	1.06	1.06	--	--	--	--	--	--
H3 Diameter	0.31	--	0.70	--	1.38	--	1.90	2.38	2.72	3.09(c)	--	--
H3 Length	0.24	--	0.53	--	0.97	--	1.37	1.69	1.95	2.23	--	--
K Diameter	0.26	0.20	0.36	0.32	(a)	--	--	--	--	--	--	--
K Length	0.24	0.15	0.28	0.24	--	--	--	--	--	--	--	--

- (a) Initial defects noted in Table 9 did not increase in severity during cycling, but became less noticeable.
- (b) Crack in thin end did increase in width but did not propagate any farther than initial length.
- (c) Although defects noted in Table 9 did not appear to increase in severity, specimen showed helium leak after 150 cycles.
- (d) The crack indicated in Table 9 widened and showed severe leak after second extended cycle - very slight helium leak noted after first 10 fast cycles.

CONFIDENTIAL

62

CONFIDENTIAL

FIGURE 34. DIAMETRAL GROWTH OF SELECTED PELLETS - PHASE II

- (a) Extended cycle Pellets A, E, G, and H2, held for 65 hr. between cycles 10 and 11.
- (b) Extended cycle Pellets A, E, G, and H2 held for 51 hr between cycles 25 and 26.
- (c) Extended cycle Pellets A, E, G, and H2 held for 51 hr after cycle 50.
- (d) H3 removed from cycling due to helium leak.

TABLE 12. FUEL FORM THERMAL CYCLE DATA - PHASE II

Fuel Form Identification	Percent Growth on Length								
	10 Fast Cycles	25 Fast Cycles	50 Fast Cycles	75 Fast Cycles	100 Fast Cycles	125 Fast Cycles	150 Fast Cycles	175 Fast Cycles	200 Fast Cycles
C	0.07	0.46	1.18	1.94	2.65	3.27	3.68	4.08	4.29
D	0.59	1.48	2.80(a)	3.76	4.68	5.90	7.32	8.95	10.99
F	2.25	4.02	6.03	6.03	6.58	7.31	8.04	8.78	9.41
G	0.22	0.54	1.07	1.78	2.19	2.90	3.91	5.24	6.77
H(b)	0.35	0.95	1.72	2.53	3.64	5.07	6.38	7.90	9.32

(a) Showed slight cracking on one end after 50 cycles.

(b) Fuel form had cracks on each end prior to thermal cycling. These cracks did not increase in size or length during cycling.

CONFIDENTIAL

CONFIDENTIAL

FIGURE 35. GROWTH CHARACTERISTICS OF PHASE I AND II UNCLAD FUEL FORMS

100X

a. Murakami's Etchant

2E892

500X

b. Murakami's Etchant

2E893

FIGURE 36. PELLET A AFTER FAST AND EXTENDED THERMAL CYCLES

This pellet structure is very comparable to Pellet A1 from Phase I.

CONFIDENTIAL

~~CONFIDENTIAL~~

0345

66

100X

a. Murakami's Etchant

2E900

500X

b. Murakami's Etchant

2E901

FIGURE 37. PELLET E AFTER FAST AND EXTENDED THERMAL CYCLES

The same matrix stability found in Phase I is still evident.

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

0345

67

100X

Murakami's Etchant

5E320

FIGURE 38. BOND OF END PLUG TO TUNGSTEN CLAD IN PELLET E

100X

Murakami's Etchant

5E319

FIGURE 39. CRACK IN THICK END PLUG OF PELLET E AFTER THERMAL CYCLING

This crack partially healed during thermal cycling.

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

C3425

68

100X

a. Murakami's Etchant

2E904

500X

b. Murakami's Etchant

2E905

FIGURE 40. PELLET G AFTER FAST AND EXTENDED THERMAL CYCLES

The integrity of the UO_2 may have been enhanced by the extended thermal cycle. See Figure 33.

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

0345

69

100X

b. Murakami's Etchant

2E916

500X

b. Murakami's Etchant

2E917

FIGURE 41. PELLET TYPE H PRIOR TO THERMAL CYCLING

The Type H Pellets are similar to the Type E pellets from Phase I except for the final bonding pressure. Slightly more porosity is evident as a result of this lower pressure.

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

0345

70

100X

a. Murakami's Etchant

2E912

500X

b. Murakami's Etchant

2E913

FIGURE 42. PELLET H2 AFTER FAST AND EXTENDED THERMAL CYCLES

~~CONFIDENTIAL~~

71

100X

a. Murakami's Etchant

2E910

500X

b. Murakami's Etchant

2E911

FIGURE 43. PELLET H1 AFTER 200 FAST THERMAL CYCLES

This structure is very similar to that of Pellet E1 (Figure 27) after 125 thermal cycles.

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

72

100X

Murakami's Etchant

5E318

FIGURE 44. BOND OF FUEL FORM TO CLAD IN PELLETT H1
AFTER THERMAL CYCLING

100X

Murakami's Etchant

5E317

FIGURE 45. BOND OF END PLUG TO CLAD AFTER THERMAL
CYCLING OF PELLETT H1.

BATTELLE MEMORIAL INSTITUTE - COLUMBUS LABORATORIES

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

0345

73

100X

a. Murakami's Etchant

2E920

500X

b. Murakami's Etchant

2E921

FIGURE 46. PELLET K AFTER 25 FAST CYCLES AND 2 EXTENDED CYCLES

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

74

100X

a. Murakami's Etchant

2E896

500X

b. Murakami's Etchant

2E897

FIGURE 47. FUEL FORM TYPE C AFTER 200 FAST THERMAL CYCLES

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

0345

75

100X

a. Murakami's Etchant

2E898

500X

b. Murakami's Etchant

2E899

FIGURE 48. FUEL FORM TYPE D AFTER 200 FAST THERMAL CYCLES

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

0345

76

100X

a. Murakami's Etchant

2E902

500X

b. Murakami's Etchant

2E903

FIGURE 49. FUEL FORM TYPE F AFTER 200 FAST THERMAL CYCLES

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

0345

77

100X

a. Murakami's Etchant

2E906

500X

b. Murakami's Etchant

2E907

FIGURE 50. FUEL FORM TYPE G AFTER 200 FAST THERMAL CYCLES

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

0345

78

100X

a. Murakami's Etchant

2E908

500X

b. Murakami's Etchant

2E909

FIGURE 51. FUEL FORM TYPE H AFTER 200 FAST THERMAL CYCLES

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

03 45

79

1.75X

46815

FIGURE 52. PELLET H1 AFTER 200 THERMAL CYCLES AND
PELLET H3 AFTER 150 THERMAL CYCLES

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

appeared about the same as the pellets cycled in Phase I. Some slight improvement in the fuel integrity in Pellet G may have been gained by the extended cycle.

- (3) The change in the bonding cycle did not have any marked effect on the growth of the pellets during thermal cycling. The growth characteristics of Type H pellets in Phase II and Type E pellets in Phase I were very similar. The very similar growth characteristics show that processing parameters and the density of the resulting structure do not control the growth pattern of the pellets.
- (4) Good reproducibility was obtained in the pellets cycled in Phase II. The Type H pellets were nearly identical and these in turn were about the same as Type E from Phase I which the Type H pellet was to duplicate. Also, the pellets which received the extended cycle responded in a similar manner to their growth in Phase I.
- (5) The tungsten-to-tungsten bonds obtained at 16,000 psi bonding pressure in Phase II were not of the quality obtained at 30,000 psi in Phase II. This can be seen by comparing Figures 38 (Pellet E) and 45 (Pellet H1). The number of thermal cycles is much greater for Pellet H1 but the extent of grain growth across the interface is not as great. Also dye-penetrant inspection picked up some defects at the bond interfaces in the Phase II

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

pellets (see Table 10) while none were detected in Phase I pellets. Part of this may be due to the removal of any smeared metal from the Phase II pellets prior to inspection.

- (6) The growth of the unclad pellets is definitely much greater than that of the clad fuel pellets. This can be seen by comparing the growth curves of unclad pellets with the clad pellets. The macroscopic integrity of the unclad pellets appeared to be good throughout the thermal cycling with the exception of the Type D fuel form. This type had a large fuel size and the clad pellets of this type were removed after 5 thermal cycles in Phase I. The fuel forms with the most desirable growth characteristics were Types C and G. Although Type C had less total growth the integrity of the matrix was very poor. Type G showed better matrix integrity. This is probably due to the greater proportion of matrix in this fuel form. The Type F fuel form showed a very rapid growth rate for the first 50 cycles but appeared to follow a more normal growth pattern after this period of high initial growth.

~~CONFIDENTIAL~~

~~Confidential~~

82

CONCLUSIONS

(CRD) As a result of the research work on this program and the previous program (NAS3-4255) the following conclusions concerning the fabrication, thermal cycling, and evaluation of tungsten clad cermet fuel pellets have been made:

- (1) The fabrication process developed during the previous contract provides clad fuel pellets of good integrity and fair reproducibility.
- (2) The factors controlling the growth of the clad cermet fuel pellets during thermal cycling are not fully understood. Certain generalizations can be made, however. These are:
 - (a) A relatively small fuel particle size--approximately 30 microns in diameter--results in less growth than larger fuel particles.
 - (b) Stabilized fuel appears to give more desirable growth characteristics.
 - (c) Fuel particles with the full coating obtained by vapor deposition appear to withstand growth as well or better than particles made up of coated particles and tungsten fines.
 - (d) Tungsten plus molybdenum duplex coatings do not result in desirable growth characteristics.

~~Confidential~~

~~Confidential~~

83

- (e) The presence of a strong cladding material of high integrity is helpful for controlling the growth of the fuel form.
- (f) Long-term heat treatments either prior to initiation of thermal cycling or during the normal fast thermal cycling procedure do not have any affect on the growth of the pellet.
- (g) Bonding parameters and the resulting variations in density of the fuel form do not appear to have a marked effect on the growth characteristics of the clad fuel pellet.

~~Confidential~~

~~Confidential~~

84

REFERENCES

1. Gripshover, P. J., and Peterson, J. H., "Development of Uranium Dioxide-Tungsten Cermet Fuel Specimens for Thermionic Applications (U)" (NASA CR-1059, Confidential) July, 1968.

~~Confidential~~

Distribution List

	No Copies
Lewis Research Center	
21000 Brookpark Road	
Cleveland, Ohio 44135	
Attention: R. L. Brown, MS 49-2	3
W. Hassel, MS 500-312	1
N. T. Musial, MS 500-311	1
Library, MS 60-3	2
Report Control Office MS 5-5	1
J. Creagh, MS 49-2	1
N. Saunders, MS 105-1	1
B. Lubarsky, MS 500-201	1
P. Donoughe, MS 49-2	1
L. Humble, MS 49-2	1
 NASA Scientific and Technical Information Facility	
Box 5700	
Bethesda, Maryland	
Attention: NASA Representative	6
 NASA Washington	
Document Control	
600 Independence Avenue SW	
Washington, D.C. 20546	
Attention: J. J. Lynch, Code RNW	1
F. Schulman, Code RNP	1
 Jet Propulsion Laboratory	
California Institute of Technology	
Central Document Control	
4800 Oak Grove Drive	
Pasadena, California 91103	
Attention: J. Davis	1
P. Rouklove	1
 NASA Ames Research Center	
Document Control Section	
Moffett Field, California 94035	
Attention: Library	1
 NASA Goddard Space Flight Center	
Security Officer 236	
Greenbelt, Maryland 20771	
Attention: Library	1
 NASA Langley Research Center	
Langley Field, Virginia 23365	
Attention: Librarian	1

NASA Marshall Space Flight Center
Huntsville, Alabama 35812
Attention: Library 1

Battelle Memorial Institute
Columbus Laboratories
Security Officer
505 King Avenue
Columbus, Ohio 43201
Attention: J. H. Peterson 3
P. J. Gripshouer 3
D. Kizer 1
D. Keller 1

Battelle Memorial Institute
Pacific Northwest Laboratories
Technical Information Center
P.O. Box 999
Richland, Washington 99352
Attention: Library 1

General Electric Company
Knolls Atomic Power Laboratory
Document Custodian
P.O. Box 1072
Schenectady, New York 12301
Attention: R. Ehrlich 1

General Electric Company
Nuclear Systems Programs
P.O. Box 15132
Cincinnati, Ohio 45215
Attention: J. W. Stephan 1
for J. A. McGurty

General Electric Company
Atomic Power Equipment Department
P.O. Box 1131
San Jose, California 95108
Attention: A. Thompson 2
for J. Danko
J. VanHoomissen

North American Aviation Inc.
Atomics International
Document Control
P.O. Box 309
Canoga Park, California 91304
Attention: R. C. Allen 1
C. K. Smith 1

Aerojet General Corporation San Ramon Plant P.O. Box 86 San Ramon, California 94583 Attention: S. Johnson for Library	1
Los Alamos Scientific Laboratory Mail and Records Group Los Alamos, New Mexico 87544 Attention: G. McGrover R. Salmi	1 1
Mr. I. M. O'Leary U. S. Atomic Energy Commission Technical Reports Library Washington, D.C. 20525	3
U.S. Atomic Energy Commission Division of Technical Information Extension P.O. Box 62 Oak Ridge, Tennessee 37831	3
Mr. Donald S. Beard Space Nuclear Systems Division U.S. Atomic Energy Commission Washington, D.C. 20545	1
Mr. Patrick O'Riordan Space Nuclear Systems Division U.S. Atomic Energy Commission Washington, D.C. 20545	1