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DISPERSION OF URANIUM CARBIDES IN ALUMINUM PLATE-TYPE RESEARCH REACTOR FUEL ELEMENTS

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operated by
UNION CARBIDE CORPORATION
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SUMMARY

The technical feasibility of employing uranium carbide—aluminum dispersions in aluminum-base research reactor fuel elements was investigated. This study was motivated by the need to obtain higher uranium loadings in these fuel elements. Although the MTR-type unit, containing a 13 – 18 wt % U-Al alloy is a proven reactor component, fabrication problems of considerable magnitude arise when attempts are made to increase the uranium investment in the alloy to more than 25 wt %. A circumventive approach to these fabrication difficulties is to select a compound with significantly higher density than UAl_4 or UAl_3 compounds of the alloy system which, when dispersed in aluminum powder, will reduce the volume occupied by the brittle, fissile phase. The uranium carbides, with densities ranging from 11.68 to 13.63 g/cm^3, appear to be particularly attractive for this application and were selected for development as a fuel material for aluminum-base dispersions.

Since the conventional processing temperatures for roll-bonded aluminum fuel plates range between 580 and 620°C, studies were conducted in this temperature region to determine the chemical compatibility of carbides with aluminum in sub-size cold-pressed compacts as well as in full-size fabricated fuel plates. Procedures were also developed to prepare uranium carbides, homogeneously disperse the compounds in aluminum, roll clad the dispersions to form composite plates, and braze the plates into fuel assemblies. Corrosion tests of the fuel material were conducted in 20 and 60°C water to determine the integrity of the fuel material in the event of an inadvertent cladding failure. In addition, specimens were prepared to evaluate performance under extensive irradiation.

Prior to studying the uranium carbide—aluminum system, methods for preparing the carbides were investigated. Arc melting uranium and carbon was satisfactory for obtaining small quantities of various carbides. Later, reaction of graphite with UO_2 was successfully employed in the preparation of large quantities of UC_2.

Studies of the chemical compatibility of cold-pressed compacts containing 50 wt % uranium carbide dispersed in aluminum revealed a marked trend toward stability as the carbon content of the uranium carbide increased from 4.46% C to 9.20% C. Severe volume increases occurred in monocarbide dispersions with attendant formation of large quantities of the uranium–aluminum intermetallic compounds. Dicarbide dispersions, on the other hand, exhibited negligible reaction with aluminum after extended periods at 580 and 620°C. However, it was demonstrated that hydrogen can promote a reaction in UC_2–Al compacts. The hydrogen appears to reduce the UC_2 to UC which can subsequently react with aluminum producing the previously noted deleterious effects.

A study of the growth at 605°C of composite fuel plates containing 59 wt % UC_2 revealed insignificant changes within processing periods envisioned for fuel element processing. However, plate elongations as high as 2.5% were observed after 100 hr at this temperature.

Severe blistering which occurred on fuel plates fabricated in the initial stages of the investigation was attributed to gaseous hydrocarbons, and the condition was eliminated by vacuum degassification of cold-pressed compacts. With the exception of the degassification requirement, procedures for manufacturing UC_2-bearing fuel elements were identical to those specified for the Geneva Conference Reactor fuel elements.

Dispersions of uranium dicarbide corroded catastrophically in 20 and 60°C water, thus limiting the application of this material. However, specimens were prepared and inserted in the MTR to evaluate the irradiation behavior of this fuel because of its potential application in organic-cooled reactors.

INTRODUCTION

Under the Atoms-for-Peace Plan, foreign countries have designed and are presently constructing research reactors quite similar to the Materials Testing Reactor at Arco, Idaho, or the Bulk Shielding Reactor at Oak Ridge, Tennessee, which require plate-type aluminum fuel elements.[1] Although the

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[1] J. E. Cunningham and E. J. Boyle, "MTR-Type Fuel Elements," Proceedings of the International Conference on the Peaceful Uses of Atomic Energy 9, p 203 (1955).

technology of manufacturing such elements is generally well developed, problems are created because of the limited U^{235} enrichment of the uranium leased under the Plan. The 20% maximum U^{235} enrichment, coupled with the desire to maintain the standard MTR fuel element design, results in a five-fold increase in the total uranium over that required for domestic fuel elements. Such a condition leads to high concentrations of uranium in aluminum, particularly when U-Al alloys are considered. This, in turn, creates fabrication difficulties and higher fuel element costs. Since the problem of fabrication is intimately associated with the volume occupied by the brittle fuel compound, an obvious approach is to substitute a fissile compound with significantly higher density than the U-Al intermetallic compounds which exist in the alloy system. The reduction in the volume occupied by the brittle compounds greatly minimizes, or completely eliminates problems which arise during fabrication.

Uranium carbides possess several inherent properties which make them attractive for this purposed application. Foremost is the fact that the densities of the carbides range from 11.68 g/cm^3 for dicarbide to 13.63 g/cm^3 for the monocarbide, and the contained uranium in the compounds ranges from 90.8 wt % to 95.2 wt %. This advantage becomes strinkingly clear in a comparison between the volume occupied by the UAl_4 intermetallic compound in a 48 wt % U-Al alloy and that occupied by the UC_2 compound when dispersed in aluminum, both containing identical quantities of U^{235}. In the alloy system the brittle intermetallic compound occupies 51% of the volume, but in the uranium dicarbide—aluminum dispersion system the compound occupies merely 20%. In addition, the 0.0045-barn thermal neutron absorption cross section of the combining element, carbon, is very low and the thermal conductivity of 0.06/0.08 cal/sec·cm °C (ref 2,3) is relatively high compared to other favorable fuel compounds.

Because of the lack of processing data on uranium carbides, it was necessary at the inception of the investigation to evaluate methods for preparing these compounds. Two methods were selected for study. The first

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[2] A. C. Secrest, E. L. Foster, and R. F. Dickerson, Preparation and Properties of Uranium Monocarbide Castings, BMI-1309 (January 2, 1959).

[3] Katz and Rabinowitch, The Chemistry of Uranium, p 220, McGraw-Hill, New York (1951).

involved inert-atmosphere arc-melting uranium and carbon in the proper proportion to obtain the desired carbide. The second was based on the reaction between uranium oxide and carbon. The massive compounds produced by each method were then comminuted to the desired particle size.

Because it was desirable to adhere as closely as possible to procedures established for manufacturing alloy-type fuel elements, the chemical compatibility of uranium carbides with aluminum in the temperature range of 580 to 620°C was studied. Since any incompatibility between the fissile compound and the matrix can minifest itself in gross distortion and warpage of composite fuel plates, it is important to establish the fact that fuel plates have dimensional stability, particularly during the 605°C brazing treatment. Investigations were conducted to determine the extent of reaction of uranium carbides with aluminum in cold-pressed compacts during heat treatment at 580 and 620°C, and to evaluate the dimensional stability of full-size composite fuel plates containing dispersions of uranium carbides in aluminum after extended heat treatment at 605°C.

In addition, reliable procedures were devised for preparation of UC_2-Al compacts by powder-metallurgy methods, roll cladding the compacts into composite plates, and brazing the plates into fuel assemblies. Testing of deliberately defected composites to establish corrosion behavior in room temperature and in 60°C water was conducted. Specimens were also prepared for evaluating the irradiation behavior of composite plates after burnups ranging from 20 to 80% of the U^{235} atoms.

CONCLUSIONS

1. Small quantities (~ 200 g) of uranium carbides with varying but controlled stoichiometry can be conveniently prepared by nonconsumable-electrode arc-melting mixtures of uranium and carbon in an inert atmosphere.

2. Either arc-melted UC_2 or UC_2 prepared by the reaction of UO_2 with graphite can be readily comminuted by conventional milling methods under an inert atmosphere into sizes suitable for dispersion-type fuel elements.

3. Uranium dicarbide particles produced by the milling of massive castings are characterized by angular shapes and are not prone to stringering or fragmentation during roll bonding.

4. Since finely divided uranium carbides are pyrophoric, reasonable caution should be exercised during handling to preclude the possibility of accidental ignition. These compounds also tend to hydrolyze and storage for extended periods should be under a protective atmosphere.

5. Uranium dicarbide does not react significantly with aluminum below 620°C.

6. Uranium monocarbide reacts catastrophically with aluminum at 620°C with gross volume increases resulting from the formation of uranium-aluminum intermetallic compounds.

7. Arc-melted UC_2 and UC_2 prepared by the reaction of UO_2 with carbon are equally stable in the presence of aluminum at 620°C.

8. The presence of H_2 in UC_2-Al bodies appears to reduce UC_2 to UC at 620°C.

9. Severe blistering, which occurs on Al-clad plates containing dispersions of UC_2 in aluminum, is attributed to gaseous hydrocarbons produced by the hydrolysis of the carbide, and can be virtually eliminated by vacuum degassification of fuel cores at 600°C prior to assembling into billets for roll bonding.

10. Composite fuel plates containing dispersions of UC in aluminum exhibit negligible dimensional increases during the high-temperature (~ 600°C) operations conventionally specified in manufacturing plate-type aluminum fuel elements. After extensive heat treatment, significant elongation of fuel plates occurs.

11. With the exception of vacuum degassification of the core compacts, fuel elements containing dispersions of UC_2 in aluminum can be manufactured using conventional powder metallurgy, roll bonding, and brazing procedures.

12. Uranium dicarbide—aluminum dispersions corrode catastrophically in room temperature and in 60°C water.

PROPERTIES AND PREPARATION OF URANIUM CARBIDES

The uranium-carbon constitution diagram[4] is shown in Fig. 1. The system contains three refractory carbides: UC, U_2C_3, and UC_2, whose pertinent physical properties are summarized in Table I.

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[4] M. W. Mallet, A. F. Gerds, and H. R. Nelson, "The Uranium-Carbon System," TRANS. Electrochem. Soc. 99, pp 197 - 204 (1952).

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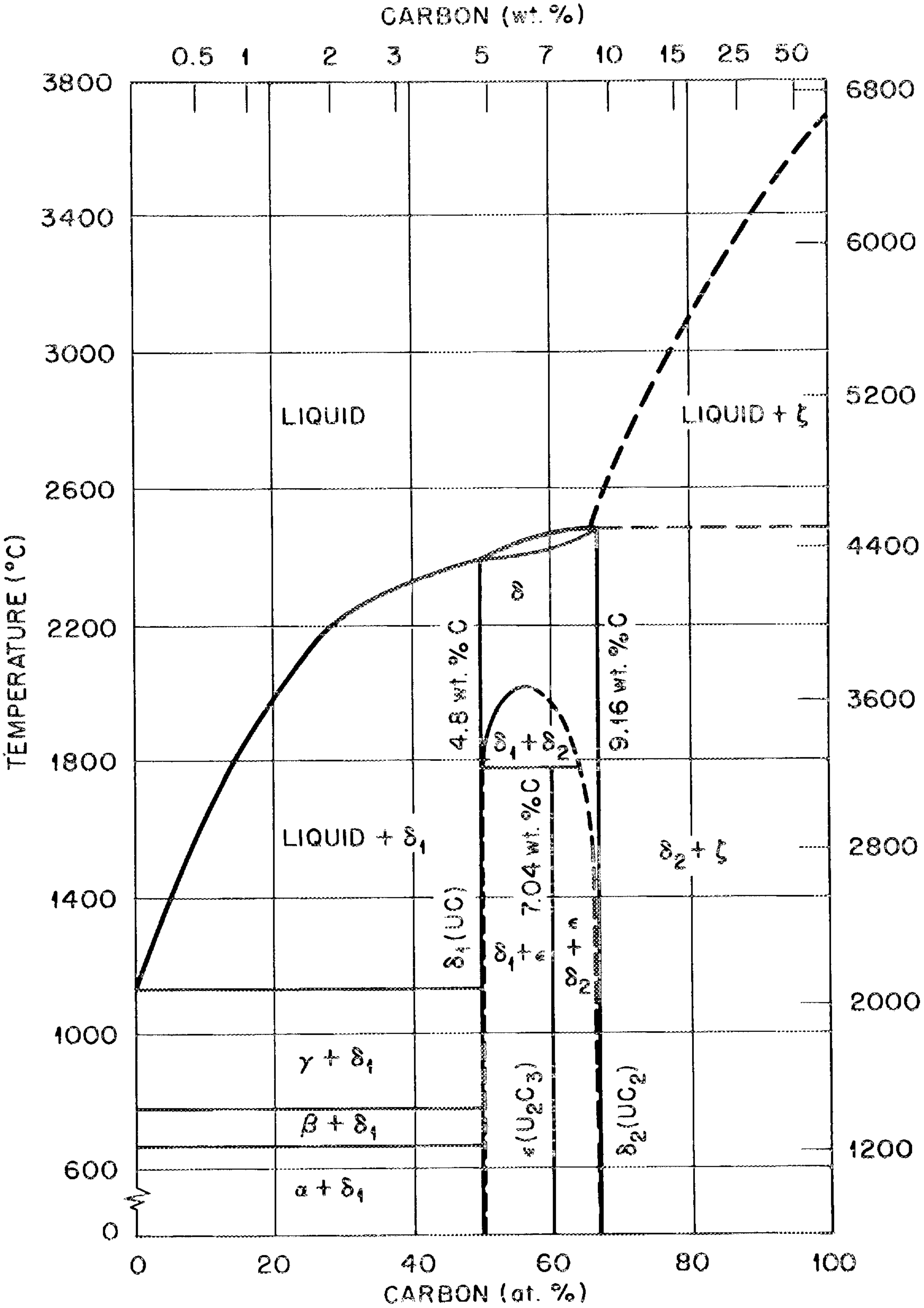

Fig. 1 Uranium-Carbon Constitution Diagram

TABLE I

PROPERTIES OF URANIUM CARBIDES

Compound	Description	Carbon Content Wt %	Crystal Structure	Lattice Constants		Melting Point °C	Pyrophoric Nature	Density* g/cc
				a_o Å	c_o Å			
UC	Uranium Monocarbide	4.80	fcc	4.961	-----	2590	yes	13.63
U_2C_3	Uranium Sesquicarbide	7.04	cubic	8.088	-----	2350 – 2400	?	12.88
UC_2	Uranium Dicarbide	9.16	bct	3.524	5.999	2350 – 2400	yes	11.68

*From lattice parameters

Uranium sequicarbide can be obtained only by special preparation techniques involving some stress,[5] while the monocarbide and the dicarbide, on the other hand, can be readily prepared by a variety of methods. A summary of the preparation techniques reported for all three compounds is included in Table II. Due to the difficulty of preparation of reasonably pure U_2C_3, this compound was not evaluated in the present study. Uranium monocarbide, uranium dicarbide, and mixtures of intermediate carbon contents, however, were extensively studied.

TABLE II*

PREPARATION OF URANIUM CARBIDES

Compound	Method of Preparation	Temperature (°C)
UC	1. $U + UC_2$	> 1800
	2. $U + CH_4$	625 – 900
	3. $U + C$	2100
	4. $U_3O_8 + C$	1800
U_2C_3	1. $UC + UC_2$ (with stressing)	1250 – 1800
UC_2	1. $UO_2 + C$	> 2400
	2. $U + C$	2400
	3. $U_3O_8 + C$	2400

*Reactor Handbook 3(1) p 120, AECD-364 (March 1955) Unclassified.

It has been reported that the most satisfactory method for obtaining bulk UC is to react stoichiometric quantities of UC_2 and U at 1800 – 1900°C.[6]

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[5] M. W. Mallett, A. F. Gerds, and D. A. Vaughan, Uranium Sequicarbide, AECD-3060 (January 1950).

[6] Sylvania Electric Products, Inc., Monthly Progress Report, SEP-242 (December 1956). CLASSIFIED.

Uranium dicarbide can be readily prepared in quantity by the reaction of UO_2 and carbon at 2400 – 2500°C.[7]

In the present investigation, limited quantities of carbides varying from sub-stoichiometric UC to super-stoichiometric UC_2 were required for compatibility studies. Inert-gas nonconsumable-electrode arc-melting provided an ideal method for compound preparation. This technique, which is more fully discussed by R. J. Gray et al.,[8] involved melting under argon the appropriate mixtures of uranium and carbon on a water-cooled copper hearth using a tungsten-tipped electrode. The resultant buttons were then commimuted to -100 mesh by hand under an argon atmosphere to prevent ignition of the pyrophoric carbides. Table III lists the arc-melted uranium-carbon compounds prepared for subsequent compatibility studies. The carbon content of each button was determined by averaging three samples. An x-ray spectrometer trace of the crushed material was also obtained to predict the relative amount of each phase present. It is interesting to note that in no case was U_2C_3 detected even in button UC-3 which had a composition equivalent to stoichiometric uranium sesquicarbide. In almost every case, reasonable homogeneity and high-carbon recovery were realized by careful control of the melting process. Microstructures of massive UC (button UC-6), an intermediate carbide containing 7.98 wt % C (button UC-2), and UC_2 (button UC-7) are shown in Figs. 2, 3, and 4, respectively.

In addition to the arc-melted carbides, larger batches of UC_2 required for compatibility studies and composite fuel plate fabrication were prepared by the reaction of carbon with UO_2. A summary of pertinent information obtained for several lots of UC_2 obtained by this process is included as Table IV. The data indicate that in every case some contamination of UC_2 with UC and/or C occurred, emphasizing the difficulty of obtaining stoichiometric UC_2 in quantity. The massive UC_2 prepared by this process was comminuted to a -100 mesh particle size by ball-milling under argon. Although the uranium dicarbide was milled under argon, it was handled in air with little

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[7] A. H. Daane, F. H. Spedding, and H. A. Wilhelan, Preparation of Carbides of Uranium, ISC -11 (December 1947). DECLASSIFIED.

[8] R. J. Gray et al., Preparation and Metallurgy of Arc-Melted Uranium Carbides, ORNL-2446 (December 1957).

TABLE III

CONSTITUTION OF ARC-MELTED URANIUM CARBIDES

Button	Intended Carbon Content (Wt %)	Analyzed Carbon Content (Wt %) End 1	Center	End 2	Avg.	Relative Amounts of Phases Present from X-Ray Intensities (%) UC_2	UC	C	U
UC-7	9.66	9.02	9.04	9.18	9.20	95	5	-	-
UC-1	9.16	8.35	8.17	8.19	8.24	90	10	-	-
UC-2	8.00	7.96	8.02	7.96	7.98	80	20	-	-
UC-3	7.04	6.94	6.96	6.98	6.96	50	50	-	-
UC-4	6.00	5.64	5.74	5.86	5.75	10	90	-	-
UC-5	4.80	4.82	4.88	4.88	4.86	10	90	-	-
UC-6	4.30	4.61	4.56	4.22	4.46	--	99	-	1

TABLE IV

CONSTITUTION OF UC_2 HEATS PREPARED BY REACTION OF CARBON WITH UO_2

Lot	Chemical Assay (Wt %) U	C	X-Ray Spectrometer Trace (%) UC	UC_2	C	Batch Size (g)
UC_2-B	90.26	9.36	12	78	10	1900
UC_2-C	-----	9.19	13	80	7	1580
UC_2-D	90.03	9.85	10	75	15	1670
UC_2-F	89.65	9.17	20	80	--	5610
UC_2-G	90.53	9.44	10	80	10	1000
UC_2-H	89.96	9.30	11	74	15	670
UC_2-I	90.28	9.31	10	74	16	1940
UC_2-J	90.20	9.20	9	69	22	2420

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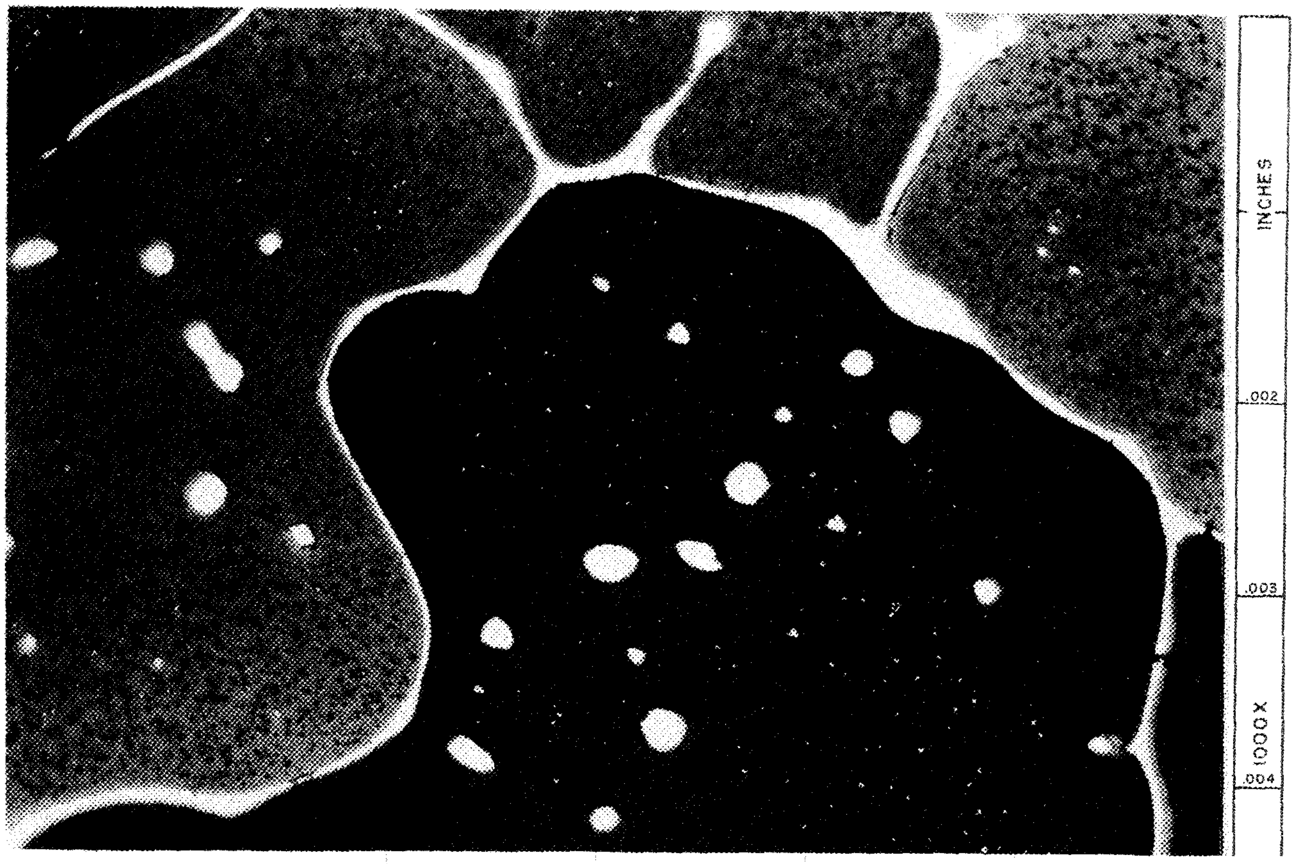

Fig. 2 Arc-Melted Uranium Monocarbide Containing 4.46 wt % C by Chemical Analysis. Microstructure is composed of massive UC with globules and intergranular film of α-uranium.
Etchant: 1/3 HNO_3, 1/3 CH_3COOH, 1/3 H_2O. 1000 X.

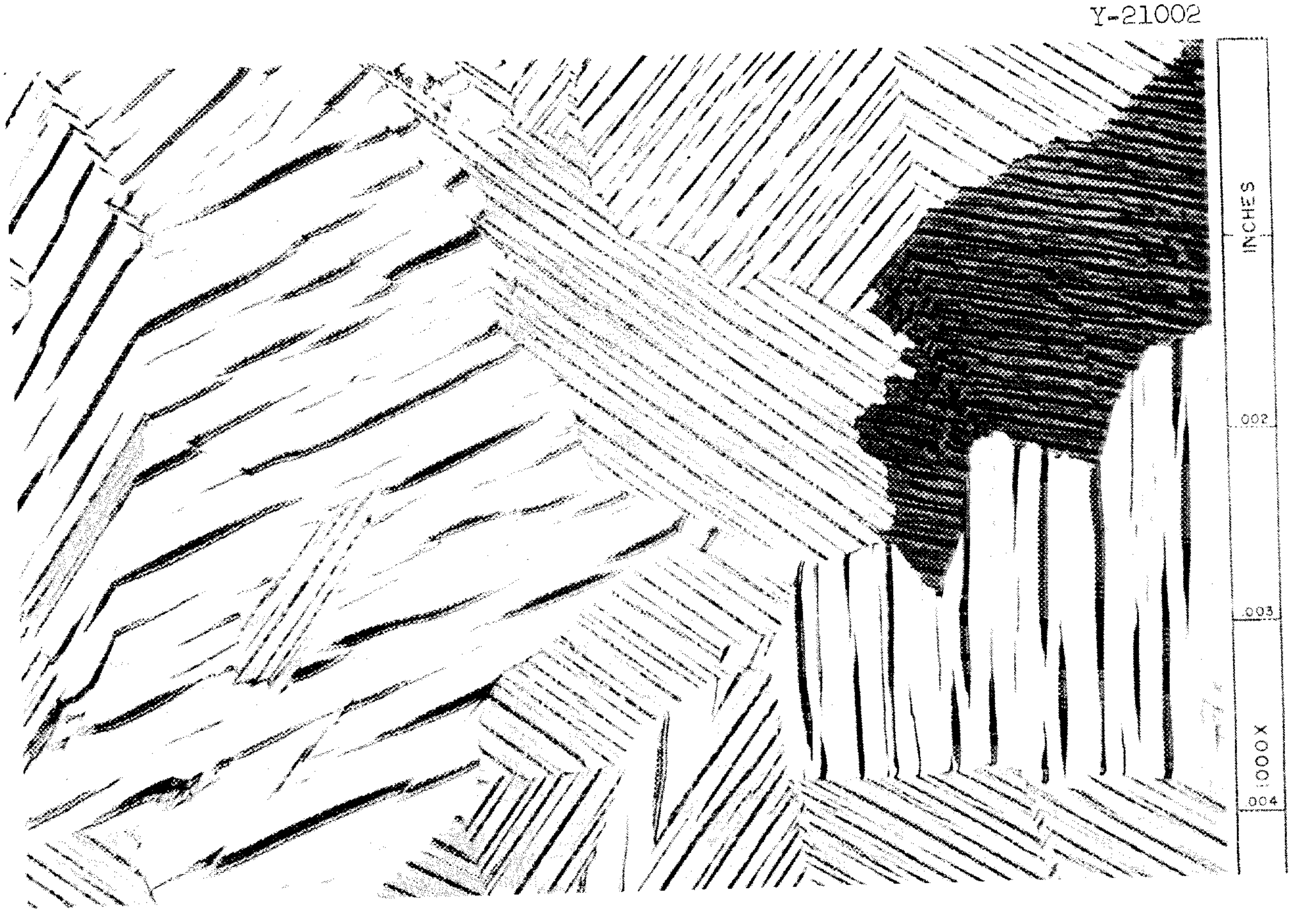

Fig. 3 Arc-Melted Uranium Carbide (Button UC-2) Intermediate in Composition Between UC and UC_2 Containing 7.98 wt % C. Matrix is UC_2 with a UC precipitate occurring along preferred crystallographic planes. Etchant: 1/3 HNO_3, 1/3 CH_3COOH, 1/3 H_2O. 1000 X.

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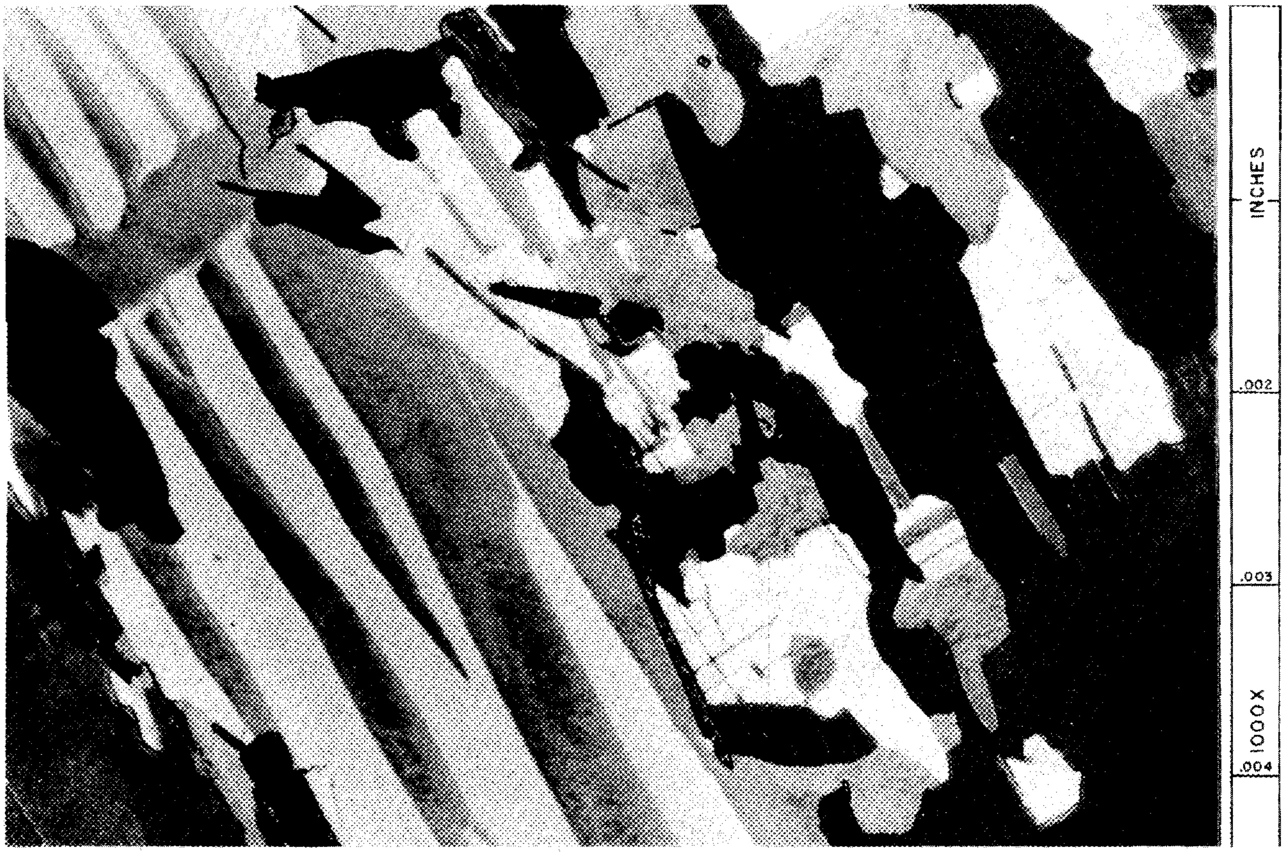

Fig. 4 Arc-Melted UC_2 (Button UC-7) Containing 9.20% C by Chemical Analysis. The matrix is UC_2 with some scattered graphite flakes present. Etchant: 1/3 HNO_3, 1/3 CH_3COOH, 1/3 H_2O. 1000 X.

difficulty and could be stored in Mason jars. For long-time storage of the finely divided material, a protective atmosphere should probably be provided to minimize hydrolysis of the carbide by atmospheric moisture. A typical screen analysis for the ball-milled UC_2 (lot UC_2-G) is:

-100 + 170 = 20.12%
-170 + 325 = 31.25%
-325 = 47.49%
Screening loss = 1.14%

COMPATIBILITY STUDIES

The compatibility studies were designed to meet the following objectives:

1. To substantiate the preliminary studies of Picklesimer[9] regarding the stability of UC_2 and the instability of UC in the presence of aluminum at elevated temperatures.

2. To establish the tolerable limits of UC contamination in UC_2 from the standpoint of designing quality-control specifications for the UC_2, and also from the standpoint of maximizing obtainable fuel loadings, if intermediate compositions proved to be unreactive with aluminum.

3. To determine the role, if any, that gases play in stability considerations. Since both UC_2 (ref 10) and Al react with water to release H_2, it was felt that any hydrogen produced might subsequently influence the uranium carbide—aluminum reaction. Therefore, this possibility received particular attention.

A. Compatibility of Uranium Carbides in Pressed and Heat-Treated Compacts

The compatibility studies primarily employed cylindrical compacts, 0.80 in. in diameter and 1.0 in. in length, containing an equiweight mixture of aluminum and the selected uranium carbide. The carbides evaluated included the arc-melted materials, UC-1 through UC-7 listed in Table III, as well as an additional carbide (UC-9) prepared by the reaction of carbon with UO_2.

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[9] M. L. Picklesimer, The Reaction of UO_2 with Aluminum, ORNL-CF 56-8-135 (August 1956).

[10] L. M. Litz, Uranium Carbides - Their Preparation, Structure, and Hydrolysis, NP-1453, USAEC (1948).

The method selected for preparation of the compacts consisted of weighing 18.00 g of -100 mesh uranium carbide and 18.00 g of -100 mesh aluminum, blending 3 hr on an oblique blender, and pressing in a 0.80-in.-dia die at 33 tsi using a stearic acid-acetone die lubricant.

The "green" compacts were held for various lengths of time at 620°C in a dynamic vacuum generally ranging from 0.10μ to 0.003μ of Hg. In addition to the extensive testing at 620°C, one series of compacts was treated in vacuo at 580°C for 24 hr. The temperatures of 580°C and 620°C essentially bracket the temperatures encountered in composite aluminum fuel plate manufacture.

The extent of reaction of the various carbides with aluminum was determined primarily by micrometric measurements, and was supplemented by visual, metallographic, and x-ray examination. Results of the tests conducted at 620°C are summarized in Table V. Similar data for the limited testing at 580°C are included in Table VI. Several salient features of the reaction of uranium carbides with aluminum can be discerned from the information presented in these two tables. In summarizing these data, it may be stated that:

1. The reaction of uranium carbides with aluminum involved conversion of the carbide to UAl_3 and UAl_4 and was generally accompanied by gross volumetric expansion of the compacts.

2. Carbides which were essentially equivalent to stoichiometric UC_2 (UC-7 and UC-9, Table V) exhibited no growth after 96 hr in vacuo at 620°C. Trace amounts of uranium-aluminum intermetallics, however, were detected by both x-ray and metallographic examination. Figure 5 illustrates the microstructure observed in the compact composed of carbide UC-7 and Al after 96-hr treatment. The greyish, shapless mass is a U-Al intermetallic while the angular, geometrical shapes are the uranium carbide. (This is an atypical field in the microstructure selected to illustrate the U-Al compound.) Metallographic examination of the several thermally treated compacts indicated that reaction occurred primarily in localized areas such as that shown in Fig. 5, rather than by peripheral attack reported for reaction of UO_2 with aluminum.[11] No explanation is apparent for this selective transformation.

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[11] R. C. Waugh and J. E. Cunningham, The Application of Low Enrichment Uranium Dioxide to Aluminum Plate-Type Fuel Elements, ORNL-CF 56-8-128 (July 1957).

TABLE V. REACTION OF EQUIWEIGHT MIXTURES OF VARIOUS URANIUM CARBIDES AND ALUMINUM AT 620°C

Time at 620°C (hr)	Weight Per Cent Carbon in Uranium Carbide														
	9.24% C (UC-9)	9.20% C (UC-7)		8.24% C (UC-1)		7.98% C (UC-2)		6.96% C (UC-3)		5.75% C (UC-4)		4.86% C (UC-5)		4.46% C (UC-6)	
	Run I	Run I	Run II	Run I	Run II	Run I	Run II	Run I	Run II	Run I	Run II	Run I	Run II	Run I	Run II
4			NG		NG		NG	ΔV = 3.9%	NG	ΔV = 90%	NG	ΔV = 99%	NG	ΔV = 110%	ΔV = 1.2%
10			NG		NG		NG		NG		ΔV = 71% ($3\% \ UAl_3$ + $34\% \ UAl_4$)		ΔV = 88% ($8\% \ UAl_3$ + $49\% \ UAl_4$)		Disintegrated ($7\% \ UAl_3$ + $34\% \ UAl_4$)
16			NG (0% UAl)		NG		NG		NG (0% UAl)						
24	NG (0% UAl)	NG	NG (0% UAl)	NG	NG	NG	NG	NG	ΔV = 4.6% ($5\% \ UAl_4$)						
48	NG ($5\% \ UAl_3$)	NG ($5\% \ UAl_3$)	NG	NG ($5\% \ UAl_3$)	NG	ΔV = 0.6% ($1\% \ UAl_3$ + $1\% \ UAl_4$)									
72	NG	NG ($5\% \ UAl_3$)	NG	NG ($5\% \ UAl_3$)	NG										
96	NG ($5\% \ UAl_3$)	NG ($5\% \ UAl_3$)	NG ($4\% \ UAl_4$)	NG ($5\% \ UAl_3$)	NG										

KEY TO TABLE:

NG – No growth of compact
(% UAl) – Amount of U-Al intermetallic compound from x-ray studies
ΔV – Volume expansion

TABLE VI

REACTION OF EQUIWEIGHT MIXTURES OF VARIOUS URANIUM CARBIDES AND ALUMINUM AT 580°C

Time at 580°C (Hr)	Wt % Carbon in Uranium Carbide						
	9.20% C (UC-7)	8.24% C (UC-1)	7.98% C (UC-2)	6.96% C (UC-3)	5.75% C (UC-4)	4.86% C (UC-5)	4.46% C (UC-6)
0	↓	↓	↓	↓	↓	↓	↓
24	NG* (0% UAl)	NG* (0% UAl)	NG* (0% UAl)	NG* (0% UAl)	ΔV = 0.2% (0% UAl)	ΔV = 49% (15% UAl_4)	ΔV = 110% (25% UAl_4) (5% UAl_3)

*No growth of compact

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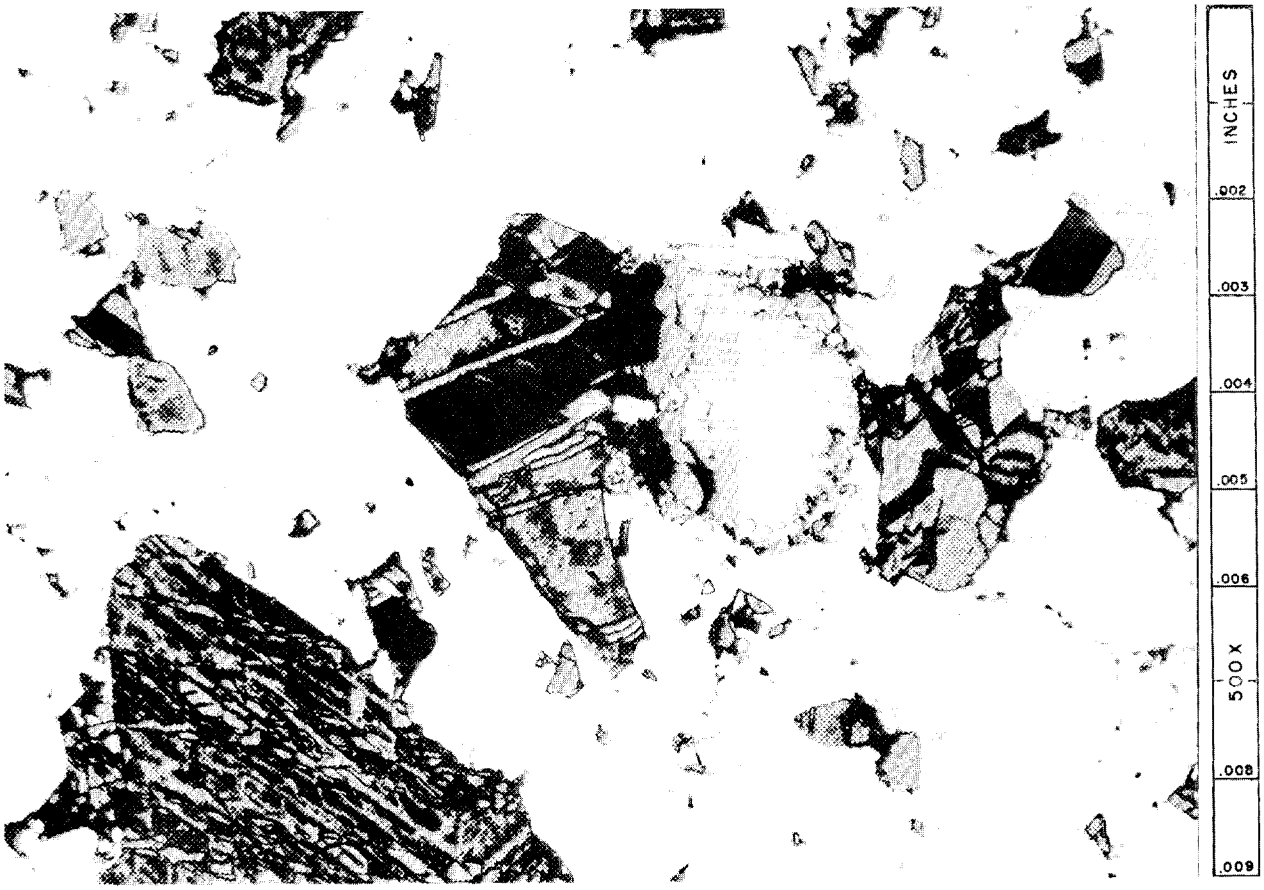

Fig. 5 Microstructure of Compact Containing 50 wt % UC-7 and 50 wt % Al after 96 hr at 620°C. Localized conversion of UC_2 to U–Al intermetallic compound is indicated by greyish reaction product in center of field. Etchant: Water stain. 500X.

3. A carbide which was essentially equivalent to stoichiometric UC (UC-5, Table V) grew catastrophically after only 10 hr at 620°C. The virtual disintegration of this compact is illustrated in Fig. 6.

4. Carbide UC-1, containing 8.24 wt % C, was found to have stability with aluminum equivalent to carbides of higher carbon content, while carbide UC-2, containing 7.98 wt % C, indicated a slight tendency for growth after 48 hr at 620°C. This indicates that the desirable minimum specification on the carbon content of the uranium carbides should be about 8.2%.

5. Comparison of the data for UC-9 and UC-7 in Table V indicated that UC_2, prepared either by arc-melting or by the reaction of C with UO_2, was equally inert to aluminum at 620°C.

6. Comparison of the 620°C data after 24 hr for each carbide listed in Table V with the 580°C data in Table VI revealed that the tendency for reaction was slightly reduced at the lower temperature for the intermediate carbides UC-4 and UC-5. However, reaction between aluminum and uranium carbide of carbon compositions near monocarbide was quite evident at 580°C.

The first experiment directed toward evaluating the role which gases play in the reaction of uranium carbides with aluminum was accomplished by individual encapsulation of aluminum compacts containing 50 wt % UC (UC-5, Table III) and 50 wt % stoichiometric UC_2 in quartz capsules. The capsules were evacuated to less than 0.01μ of Hg prior to sealing and were then heated for 24 hr at 620°C. The gases evolved during the heat treatment were removed through a break-off tip on the quartz bulb and analyzed by mass spectrometry. The nature of the solid-reaction products was subsequently determined by x-ray spectrometry. Results of these measurements are presented in Table VII, while the visual appearance of these two compacts is illustrated in Fig. 7. As expected, the compact containing UC grew catastrophically during the 24-hr thermal treatment, whereas the compact containing UC_2 remained dimensionally stable.

From Table VII, it can be seen that the compact which initially contained UC had completely reacted to form UAl_3 and UAl_4 while the gas in the capsule was essentially H_2. The UC_2–Al compact, on the other hand, contained minor quantities of UC and UAl_3 as solid-reaction products with the gaseous product consisting chiefly of CH_4; no H_2 was detected.

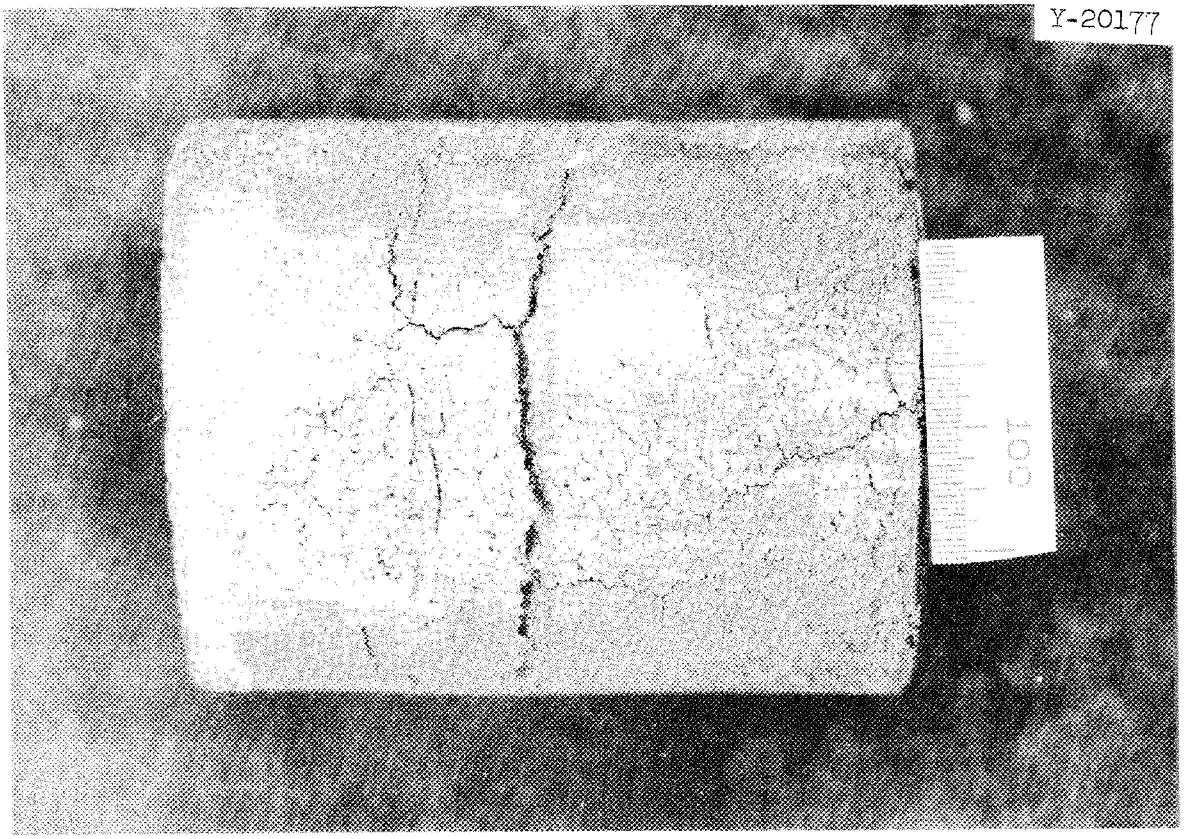

Fig. 6 Appearance of Compact Containing 50 wt % UC (4.86 wt % C) and 50 wt % Aluminum after Treatment in Vacuum for 10 hr at 620°C. 4 X.

TABLE VII

REACTION OF URANIUM CARBIDES WITH ALUMINUM IN VACUO AT 620°C

$$UC_2 + Al \xrightarrow[24\ \text{hr}]{620°C} \begin{cases} \text{Solid} & \left\{ \begin{array}{ll} UC_2 & - 36\% \\ UC & - 16\% \\ Al & - 45\% \\ UAl_3 & - 3\% \end{array} \right. \\ \text{Gas} & \left\{ \begin{array}{ll} CH_4 & - 89.32\% \\ N_2 & - 2.55\% \\ O_2 & - 0.08\% \\ H_2O & - 6.95\% \\ H_2 & - 0\% \\ CO_2 & - 0.79\% \\ H\text{-}C & - 0.31\% \end{array} \right. \end{cases}$$

$$UC\ (+\ 10\%\ UC_2) + Al \xrightarrow[24\ \text{hr}]{620°C} \begin{cases} \text{Solid} & \left\{ \begin{array}{ll} UAl_3 & - 18\% \\ UAl_4 & - 47\% \\ Al & - 35\% \end{array} \right. \\ \text{Gas} & \left\{ \begin{array}{ll} CH_4 & - 0.06\% \\ H_2 & - 99.93\% \\ N_2 & - 0.01\% \end{array} \right. \end{cases}$$

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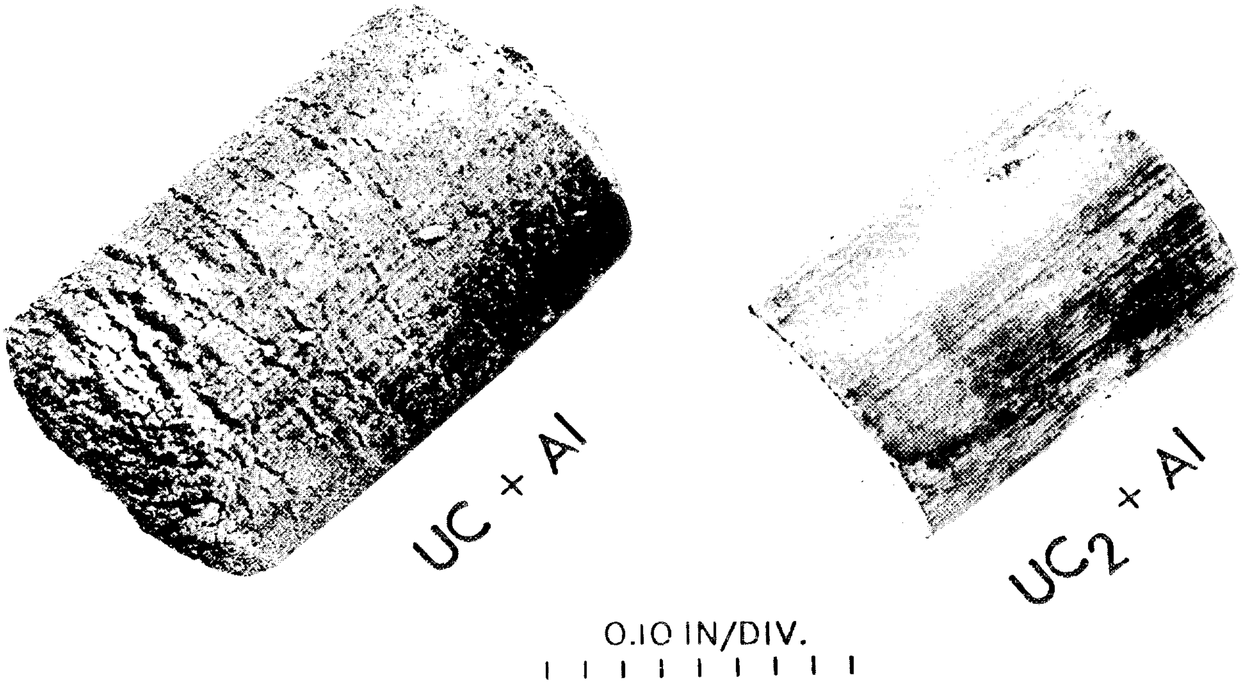
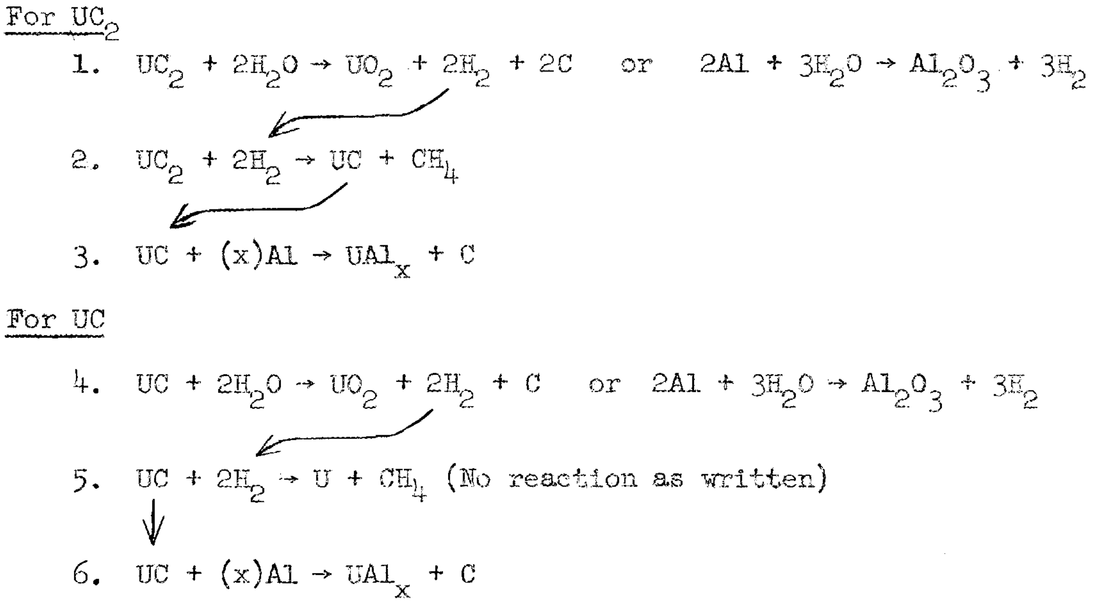


Fig. 7 Appearance of Aluminum-Base Compacts Containing Both UC and UC_2. Marked growth of UC after heat treatment at 620°C obvious.

Growth which might be anticipated from the slight amount of reaction in the UC_2-bearing compact was probably masked by shrinkage from sintering. On the basis of these observations, it was postulated that hydrogen produced from water associated with either/or both components in the compact, reacted with UC_2 to form methane and UC, but H_2 from the same source did not react with the UC. The UC which formed from the UC_2 subsequently reacted with the aluminum to produce UAl_3 or UAl_4. This mechanism can be represented as follows:



For UC_2

1. $UC_2 + 2H_2O \rightarrow UO_2 + 2H_2 + 2C$ or $2Al + 3H_2O \rightarrow Al_2O_3 + 3H_2$
2. $UC_2 + 2H_2 \rightarrow UC + CH_4$
3. $UC + (x)Al \rightarrow UAl_x + C$

For UC

4. $UC + 2H_2O \rightarrow UO_2 + 2H_2 + C$ or $2Al + 3H_2O \rightarrow Al_2O_3 + 3H_2$
5. $UC + 2H_2 \rightarrow U + CH_4$ (No reaction as written)
6. $UC + (x)Al \rightarrow UAl_x + C$

Evidence that reactions 1 and 4 do occur has been well documented in the literature.[12] The thermodymanics of reactions 2 and 5 were examined to determine validity. Calculations[13] revealed that (1) $\Delta F°$ for reaction 2 was -5.7 kcal/mole, indicating that the reaction could occur as written and (2) $\Delta F°$ for equation 5 was +39.2 kcal/mole, indicating that this reaction would not occur in the direction shown. In fact, as described previously in Table II, uranium monocarbide is prepared by the reverse reaction. Experimental evidence of the reaction shown in equation 2 can also be found in the work of Litz.[12]

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[12] L. M. Litz, Uranium Carbides - Their Preparation, Structure, and Hydrolysis, NP-1453, USAEC (1948). Unclassified.

[13] W. C. Thurber and R. J. Beaver, Interim Report on the Application of Uranium Carbides - Aluminum Dispersions in Aluminum Fuel Elements, ORNL-CF 57-8-65 (August 1957). Unclassified.

A supplementary experiment dramatically demonstrated the influence of hydrogen on reaction of uranium dicarbide with aluminum. Identical aluminum compacts containing 50 wt % UC_2 (UC_2—C, Table IV) were fired for 24 hr at 620°C in two different environments. One compact was heated under a dynamic vacuum while the other was exposed to hydrogen. The vacuum-heat-treated compact showed no dimensional changes and no detectable uranium—aluminum intermetallics. On the other hand, the hydrogen-heat-treated compact increased 1.4% in volume and x-ray examination revealed that the specimen contained 23% UAl_4.

On the basis of the foregoing experiments, it is obvious that H_2 does influence the reaction between UC_2 and aluminum at 620°C, probably by reducing UC_2 to UC which subsequently reacts with aluminum to produce UAl_3 and UAl_4. It can also be conjectured that hydrogen promotes the UC_2—Al reaction by reducing protective surface films on the UC_2, and that UC_2 with a properly conditioned surface is actually reactive with aluminum. Since free-energy data are not available for the uranium—aluminum intermetallic compounds, it is not possible to choose between the two postulates.

B. Compatibility of Uranium Carbides in Composite Fuel Plates

The compatibility of UC_2 with aluminum was determined from the results of six full-size plates containing 59 wt % UC_2. All plates except one were fabricated identically. The exception (No. M-128), in addition to containing a vacuum-sintered core, was manufactured from a billet which was evacuated at 600°C prior to the rolling operation. After fabrication, each plate was heat treated at 605°C in 16-hr increments. At the termination of each 16-hr period, the plates were air cooled and length and volume changes accurately measured by micrometer and water-displacement techniques, respectively. After 64 hr (4 cycles) at 605°C, volume increases were less than 0.04%. After 128 hr (8 cycles) at this temperature, however, volume increases varied from negligible to 2.07%. Elongation data of the heat-treated plates are summarized in Fig. 8. Only very small changes in elongation occur during the estimated 3-hr fuel element manufacturing cycle. However, plate elongation increases at an increasing rate with time, and, in general, after 64 hr has progressed to about 0.5%. It may be significant that plate No. M-128, which was evacuated at 600°C prior to rolling, exhibits significantly less elongation than the other

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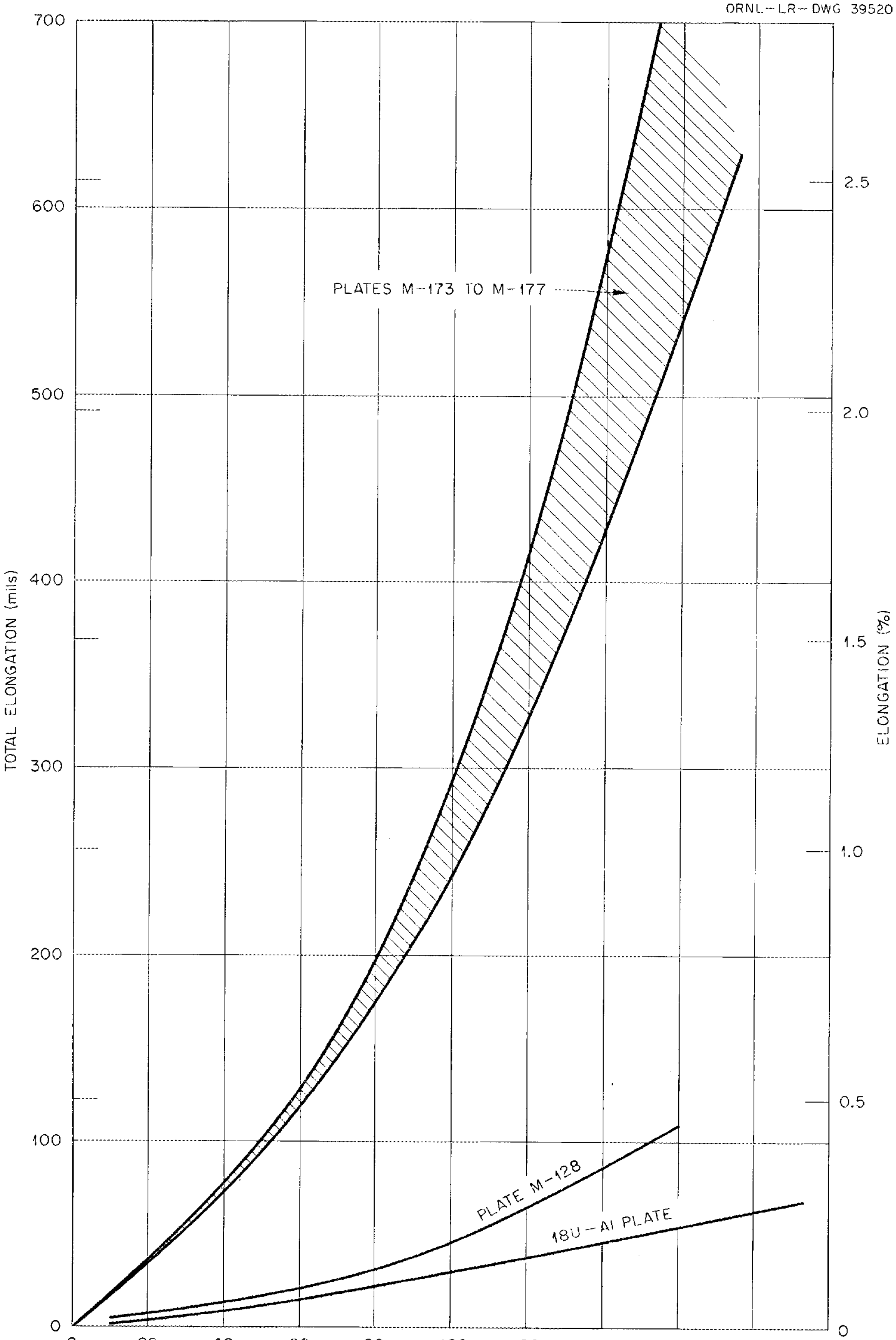

Fig. 8. Growth of Composite 59 wt% UC_2-Al Fuel Plates at 605°C.

carbide-bearing plates. It is also pertinent that the conventional 18% U-Al alloy composite elongated during this study. Table VIII lists the reaction products found in the plates after completion of the thermal cycling. The majority of the plates contained rather large amounts of the intermetallic reaction product. However, plate No. M-128, which had a negligible volume change and a low elongation after 128 hr, contained relatively small quantities of the U-Al intermetallic compounds. In accordance with the previously proposed reaction mechanisms, it appears that the UC impurity in the UC_2 is responsible for a portion of the U-Al intermetallics and that a hydrogen source exists in the fuel compact which contributes additional UC. This, in turn, reacts to form more UAl_3 and UAl_4 intermetallic compounds. It is possible that the low conversion ratio for plate M-128, which was evacuated at 600°C prior to rolling, may be due to lower hydrogen content. Admittedly, the basis for this proposal is from one experiment and, although a trend appears to exist, it needs substantiation. A portion of the deformation is obviously due to creep as indicated by the growth of the U-Al alloy plate.

TABLE VIII

REACTION OF UC_2 WITH AL IN COMPOSITE FUEL PLATES

Plate	Total Time at 605°C (hr)	Relative Amounts of Components from X-Ray Intensities				Conversion Ratio*
		UAl_3	UAl_4	UC	UC_2	
M-128	160	10	10	18	70	0.23
M-173	176	8	53	22	62	0.73
M-174	176	9	65	18	45	1.17
M-175	176	5	68	20	42	1.02
M-176	176	12	78	20	40	1.50
M-177	176	9	72	20	57	1.05

$$^{*}\text{C. R.} = \frac{UAl_3 + UAl_4}{UC + UC_2}$$

FABRICATION STUDIES

On the basis of the compatibility investigations, UC_2 was the carbide selected as the fuel dispersoid in aluminum composite fuel plates. The UC_2 for these fabrication studies was prepared primarily by the reaction of UO_2 and C; a few plates were also manufactured with UC_2 prepared by arc-melting.

To minimize the amount of experimental work required, techniques established for manufacturing composites containing dispersions of UO_2 in aluminum were followed. Basically, the preparation of fuel sandwiches for rolling by these previously developed practices involves: (1) weighing the appropriate charge, (2) blending in oblique blender, (3) pressing into 2.3 x 2.0-in. die cavity at 33 tsi using 0.001-in.-thick Al foils on either side of the compact, (4) pressing the green core compact into an 1100 Al picture frame to eliminate air entrappment around the core perimeter, (5) fusion welding type 1100 Al cover plates to a type 1100 Al frame along the sides parallel to the rolling direction, and (6) preheating the assembled sandwich in air at 600°C and hot rolling to desired thickness using intermediate reheating.

Of sixty fuel plates fabricated by this procedure, slightly more than 50% were rejected because of the formation blisters during the required heat treatments. Figure 9 illustrates the surface appearance of a typical defective fuel plate manufactured by these techniques. The profusion of large blisters is obvious. Figure 10 is a transverse section taken through a blister in a similarly fabricated fuel plate. It can be seen that the blisters are manifestations of defects within the core rather than flaws at the core-clad interface. Gases, present in both blistered and unblistered areas from a typical plate, were collected by vacuum-fusion techniques and identified by mass spectrometry. Results of these analyses are listed below:

Unblistered Sample	
CH_4	0.3%
H_2O	0.5%
N_2	78 %
O_2	20 %
A	1 %
CO_2	0.2%
Other hydrocarbons	0%

Blistered Sample	
CH_4	26%
H_2O	18%
N_2	25%
O_2	1%
A	1%
CO_2	3%
Other hydrocarbons	26%

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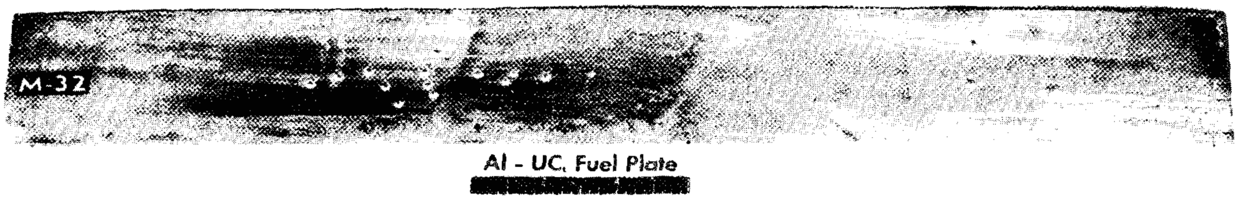

Fig. 9 Blisters on Surface of Composite Fuel Plate Containing Unsintered Core.

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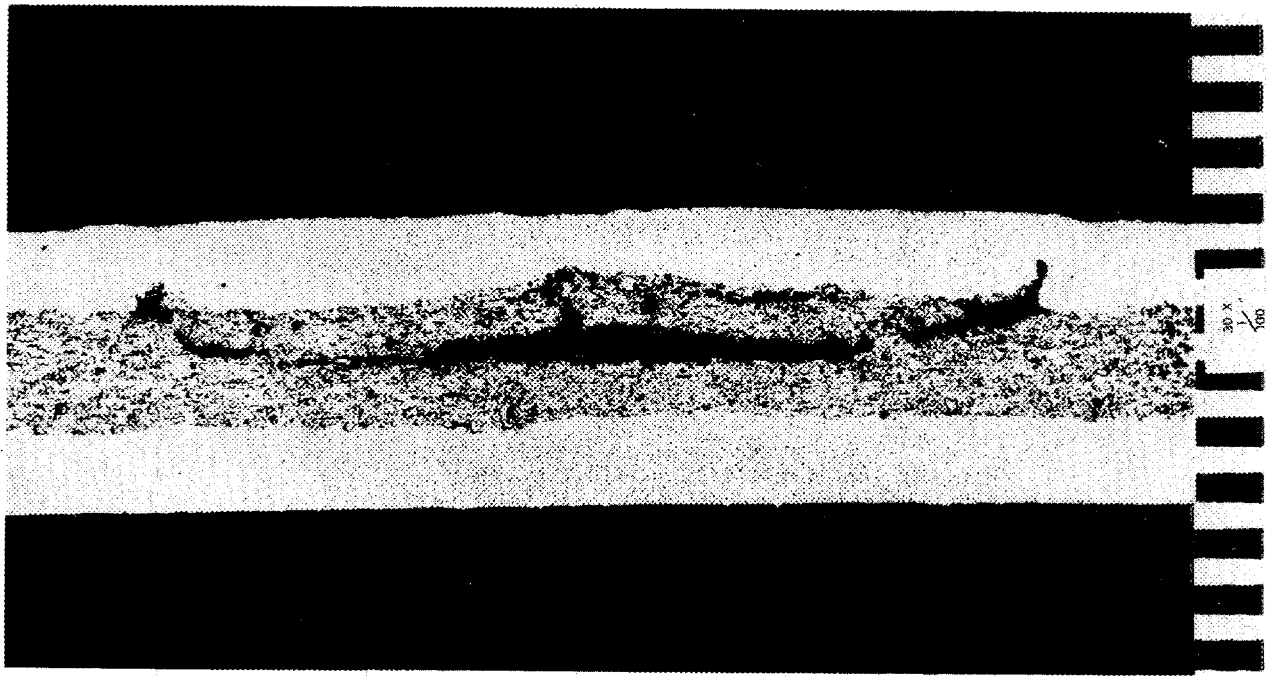

Fig. 10 Transverse Section Through a Blister in a Composite UC_2-Al Fuel Plate (M-77). Origin of blister within core is apparent. As-polished. 30 X.

These analyses reveal that the gases removed from the unblistered material have essentially the same composition as air, whereas gases in the blistered material contain more than 50% hydrocarbons and 18% H_2O. It was suspected from the hydrocarbonaceous quality of the gas that hydrolysis of UC_2 had occurred.

Analyses were conducted on four uranium carbide samples and two aluminum samples to determine the amount of water removed as a function of temperature. The total quantity of water removed after heating to 300°C and to 600°C are listed in Table IX. A maximum and minimum value is presented for each temperature since a minimum level below which no significant changes could be detected existed in the analytical method. From these data it can be seen that the UC_2 contains two to five times more water than aluminum powder. There is also some indication that 600°C is a more effective temperature than 300°C for removal of the water.

TABLE IX

WATER REMOVED FROM CORE MATERIALS AT 300°C AND 600°C

	At 300°C		At 600°C	
Powder	Minimum (Wt %)	Maximum (Wt %)	Minimum (Wt %)	Maximum (Wt %)
UC_2-G	0.29	0.36	0.51	0.72
UC_2-H	0.77	0.82	0.77	0.97
UC_2-I	0.64	0.78	0.64	0.99
UC_2-J	0.43	0.45	0.43	0.48
Al-1	0.14	0.16	0.14	0.22
Al-2	0.19	0.20	0.19	0.23

To effect water removal, billet evacuation was the first process modification selected for study. As shown in Fig. 11, an aluminum evacuation stem was incorporated into the billet assembly and the UC_2–Al compact sealed within the billet by fusion welding around the entire perimeter of the sandwich. For convenience, the first experimental billets were heated only to 300°C and the pressure within the billet was reduced to less than 1μ of Hg. The stem of the billet was then sealed by forge welding, and after preheating at 600°C, the billets were hot rolled into composite plates. Of twenty-four plates prepared by this technique, all but one blistered during heat treatment. Included in this group were eight plates containing arc-melted UC_2 and sixteen plates containing UC_2 produced by the reaction of UO_2 and C, indicating that the method of carbide preparation was not the controlling factor in eliminating blisters.

Since the evacuation for one-half hour at 300°C did not appear sufficiently rigorous to eliminate the blisters, billets were subsequently evacuated at 600°C for 2 hr. Of sixteen plates fabricated by this technique, none were rejected for blisters.

Additional experimentation revealed that billet evacuation could be replaced by vacuum sintering of the fuel cores to minimize blister rejections in the subsequently fabricated fuel composite. The recommended practice is to sinter the core at 600°C for 2 to 3 hr under a dynamic vacuum of less than 1μ of Hg. Of seventy plates fabricated by this procedure, only two were rejected for blister formation during subsequent thermal treatments at 600°C.

No special experiments were conducted to optimize the particle sizes and ratio of particle sizes for matrix and fertile phases. Instead, -100 mesh size fraction of both aluminum and UC_2 was employed in all the fuel plates. However, some effort was made to control the number of UC_2 fines produced in the milling operation in order to minimize the tendency for spontaneous ignition.

To determine the densification on cold compacting of various UC_2–Al mixtures, a series of four compacts containing from 58.8 to 68.2 wt % UC_2 was prepared. These compacts were cold pressed at 33 tsi to 94.7 – 95.7% of theoretical density. Virtually no dimensional changes were noted after vacuum sintering the fuel cores.

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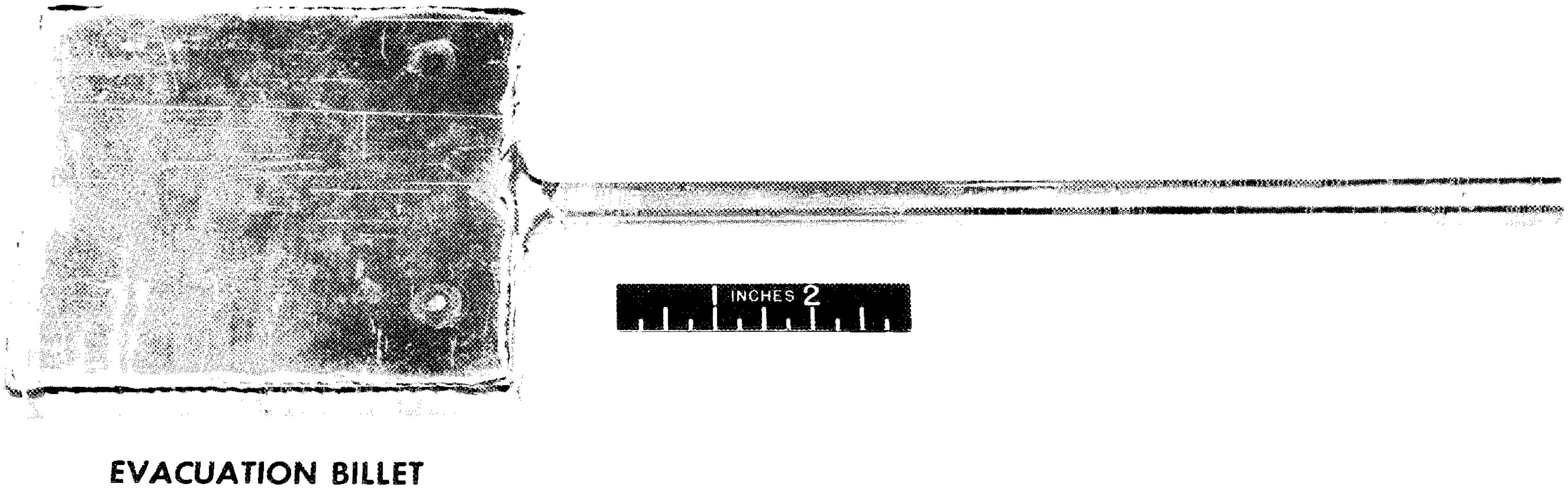

Fig. 11 Typical Evacuation Billet Prior to Sealing.

The longitudinal cross section of a typical fuel plate containing a dispersion of 58.8 wt % UC_2 in aluminum is shown in Fig. 12. It can be observed from this illustration that little or no fragmentation or stringering of the angular UC_2 particles has occurred during the rolling operations.

The feasibility of brazing composite fuel plates containing 60 wt % UC_2 dispersed in aluminum was demonstrated by the manufacture of several MTR-type fuel elements.

CORROSION STUDIES

The corrosion of UC_2—Al dispersions in demineralized water was evaluated at room temperature and at 60°C on two types of samples. The "unclad" type of sample was bare core material, consisting of a dispersion of 61.6 wt % UC_2 in aluminum, prepared by machining the cladding from the fuel section of a composite plate. These specimens were 2 in. long, 1/2 in. wide, 0.017 in. thick with a 1/16-in.-dia hole drilled near one end for suspending the sample in the corrosive medium. The aluminum-clad samples were similar to the unclad samples in composition and size. The fuel dispersion was clad with 0.015-in.-thick 1100 Al on the flat surfaces. However, the fuel was exposed at the edges and at the hanger hole.

Clad and unclad specimens were tested in room-temperature water for 24, 48, 72, 96, and 120 hr. Each type was also tested in 60°C water for 24 and 48 hr. Due to the catastrophic nature of the uranium carbide—water reaction, no long-term tests were conducted.

The extent of corrosion was determined primarily by visual examination and weight-change measurements. The pH of the solution was also monitored and chemical analyses were performed on all of the solid precipitates. Results of these tests are summarized in Table X. It can be seen from the weight change and precipitate weight data that a marked increase in the corrosion rate, or more accurately the hydrolysis rate, occurs in both the clad and the unclad specimens when the temperature is increased from room temperature to 60°C.

Gas evolution was observed when all specimens were placed in water, with the rate of evolution decreasing with time. Although this gas was not quantitatively analyzed, a strong acetylene order was detected, indicating a carbide-hydrolysis reaction.

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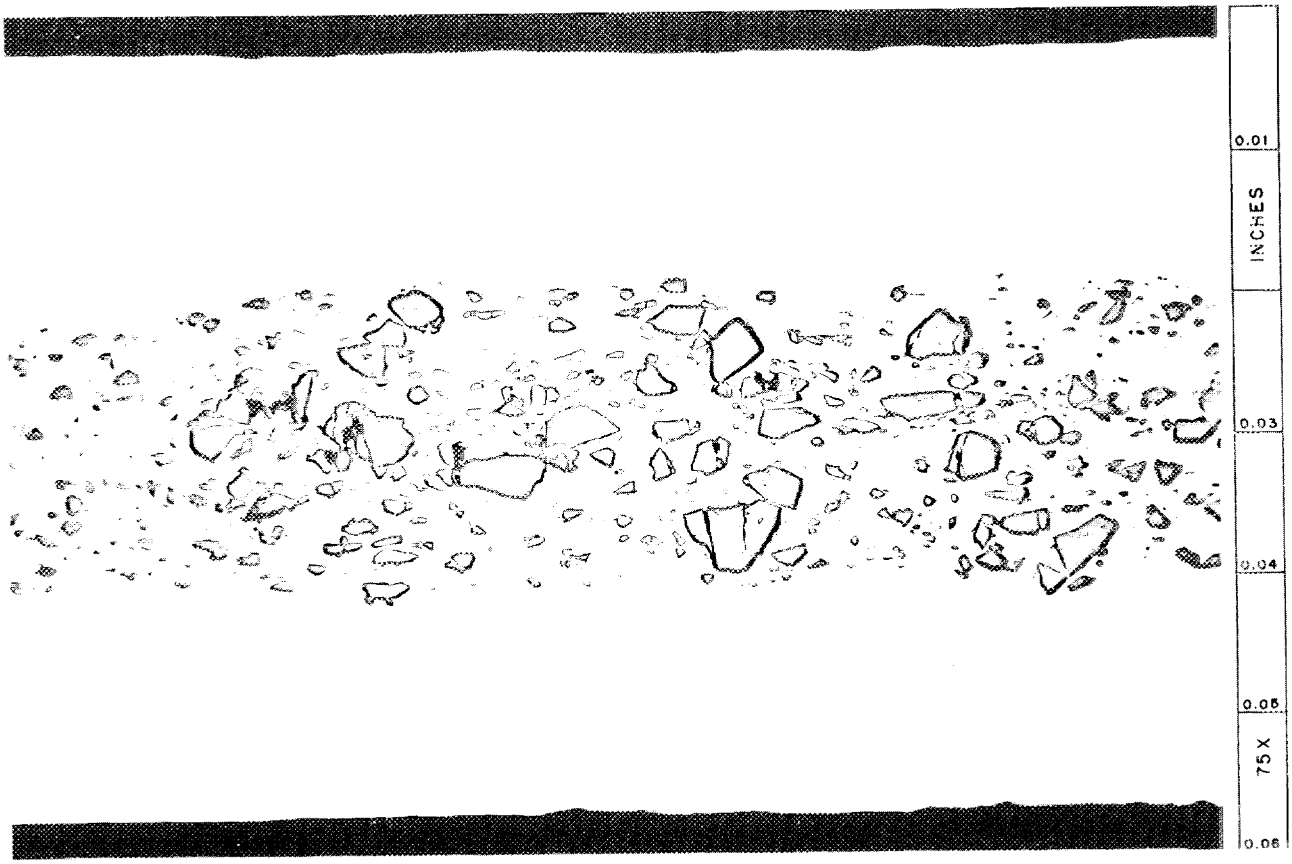

Fig. 12 Longitudinal Cross Section of Composite Aluminum-Clad 59 wt % Al Dispersion Fuel Plate Illustrating Continuity of Core-Clad Bond and Lack of Fragmentation and Stringering of UC_2. As-polished.

TABLE X

CORROSION OF 62 WT % UC_2–AL SPECIMENS IN DEMINERALIZED WATER

Specimen Number	Surface Condition	Time in Test (hr)	Temperature	Weight of Solid Precipitate (mg)	Chemical Analysis of Solid Precipitate		Specimen Weight Gain (mg)	Solution pH*
					(% U)	(% Al)		
300	Clad	24	Room	-	-	-	1.25	6.50
301	Clad	48	Room	-	-	-	.5	6.25
302	Clad	72	Room	-	-	-	1.1	6.30
303	Clad	96	Room	-	-	-	5.2	6.50
304	Clad	120	Room	-	-	-	4.0	6.40
305	Clad	24	60°C	Trace	-	-	126	8.80
306	Clad	48	60°C	13.0	64.1	14.7	299	8.10
310	Unclad	24	Room	-	-	-	20	7.25
311	Unclad	48	Room	26.0	36.1	18.80	97	7.30
312	Unclad	72	Room	10.7	19.5	28.1	90	7.70
313	Unclad	96	Room	10.2	22.4	28.9	86	8.70
314	Unclad	120	Room	58.2	52.4	21.2	**	8.70
315	Unclad	24	60°C	83.0	60.2	7.15	249	7.35
316	Unclad	48	60°C	96.3	76.3	8.6	225	7.30

*Initial pH = 5.6

**Specimen disintegrated, no weight-change data possible.

The unclad specimens were, of course, more drastically attacked with scaling, warpage, and growth occurring simultaneously. One specimen was observed to increase 6% in length. The complete degradation of unclad specimen No. 316 after 48 hr in 60°C water is shown in Fig. 13.

The appearance of a clad specimen (No. 305, Table X) after exposure to 60°C for 24 hr is shown in Fig. 14. Severe attack of the core coupled with peeling back and distortion of the cladding can be noted.

No severe distortions of clad specimens tested in room-temperature water were observed and chemical analysis of several solutions indicated uranium contents of less than 2 ppm. Some attack did, nevertheless, occur in these specimens as witnessed by the water-affected zone around the hanger hole of specimen No. 304 shown in Fig. 15.

The possibility of an inadvertent cladding defect, coupled with the catastrophic nature of the corrosion of UC_2—Al dispersions in 60°C water, renders this materials combination marginal for research reactor applications. However, this does not preclude the potential use of these dispersions in other cooling media, particularly organics.

IRRADIATION TESTING

In order to study the irradiation behavior of UC_2—Al dispersions over a wide range of burnups, a series of ten miniature fuel plates were fabricated. The core in each of these plates was a dispersion consisting of 59.5 wt % UC_2 (enriched 19.99% in the U^{235} isotope) and 40.5 wt % Al. The fabrication procedures were identical to those previously outlined for producing blister-free plates using vacuum-sintered fuel cores. Each plate was 6.0 in. long x 1.0 in. wide x 0.050 in. thick and each core was nominally 5.0 in. long x 0.70 in. wide x 0.020 in. thick. A radiograph of some typical miniature plates is shown in Fig. 16. Pertinent dimensional information and uranium loadings for these plates are included in Table XI.

As a pre-irradiation performance test for cladding defects, the plates were held in demineralized 90°C water for 24 hr. No deleterious effects were noted during the test. Eight of these plates, selected for subsequent irradiation, were inserted in "leaky rabbit" capsules after careful evaluation of external dimensions using the same facilities which will be used for

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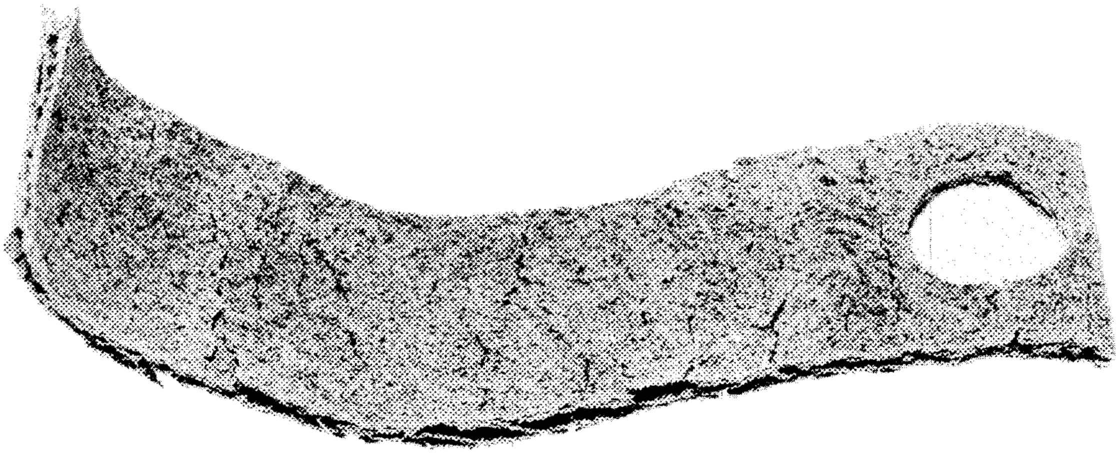

Fig. 13 Unclad UC_2–Al Dispersion Exposed in 60°C Demineralized Water for 48 Hr Showing Catastrophic Attack. 3 X.

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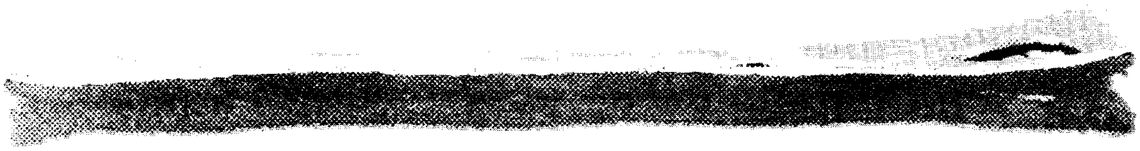

Fig. 14 Aluminum-Clad UC_2—Al Dispersion Exposed in 60°C Demineralized Water for 24 Hr, Showing Core Attack and Cladding Distortion. 3 X.

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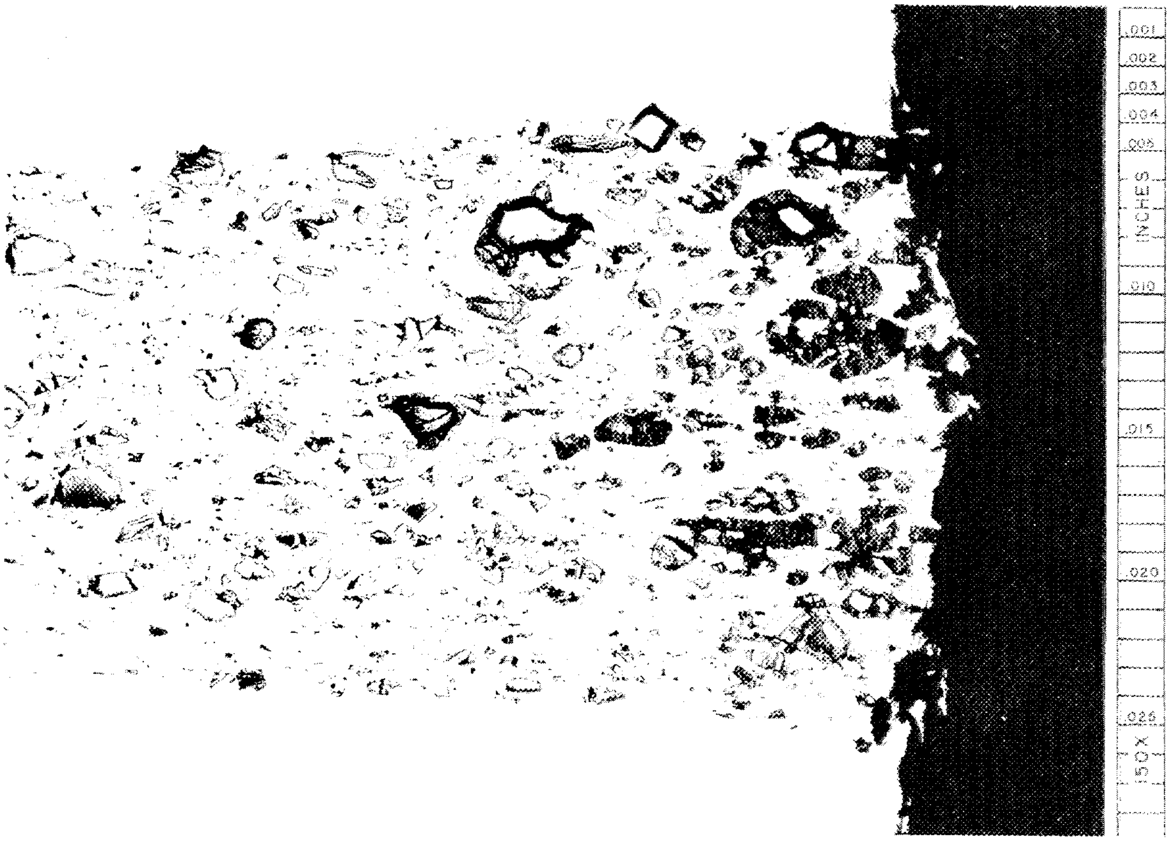

Fig. 15 Cross Section Through Hanger Hole in Aluminum-Clad UC_2—Al Dispersion held at Room Temperature for 120 hr in Demineralized Water Showing Slight Attack at Hole Perimeter. As polished. 50 X.

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Fig. 16 Radiograph of Typical UC_2–Al Miniature Composite Fuel Plates for Irradiation Testing. Actual size.

TABLE XI

DIMENSIONAL AND FUEL-LOADING DATA FOR MINIATURE UC_2—AL FUEL PLATES

Plate Number	Finished Core Length[a] (in.)	Finished Core Width[b] (in.)	Finished Core Area (cm^2)	Core Weight[c] (g)	Total Uranium Content of Core[d] (g)	U^{235} Content of Core[e] (g)	U^{235} Surface Density (g/cm^2)
OR-35-C-1	4.875	0.703	22.11	5.786	3.087	0.617	0.028
OR-35-C-2	4.844	0.703	21.97	5.803	3.096	0.619	0.028
OR-35-C-3	4.906	0.703	22.25	5.669	3.025	0.605	0.027
OR-35-C-4	4.844	0.703	21.97	5.839	3.116	0.623	0.028
OR-35-C-5	4.844	0.703	21.97	5.810	3.100	0.620	0.028
OR-35-C-6	4.812	0.688	21.36	5.588	2.982	0.596	0.028
OR-35-C-7	4.875	0.703	22.11	5.839	3.116	0.623	0.028
OR-35-C-8	4.812	0.703	21.82	5.786	3.087	0.617	0.028
OR-35-C-9	4.781	0.703	21.68	5.761	3.074	0.614	0.028
OR-35-C-10	4.906	0.703	22.25	5.838	3.115	0.632	0.028

[a] ± 0.031 in. (from radiographs)

[b] ± 0.016 in. (from radiographs)

[c] Corrected for weight of aluminum foil (0.022 g)

[d] Based on uranium assay for the UC_2 of 89.65%

[e] Based on an enrichment of 19.99% in U^{235} isotope

post-irradiation inspection. A typical assembly ready for insertion in the MTR is shown in Fig. 17. Pairs of samples are presently under irradiation to scheduled burnups of 20, 40, 60, and 80% of the U^{235} atoms.

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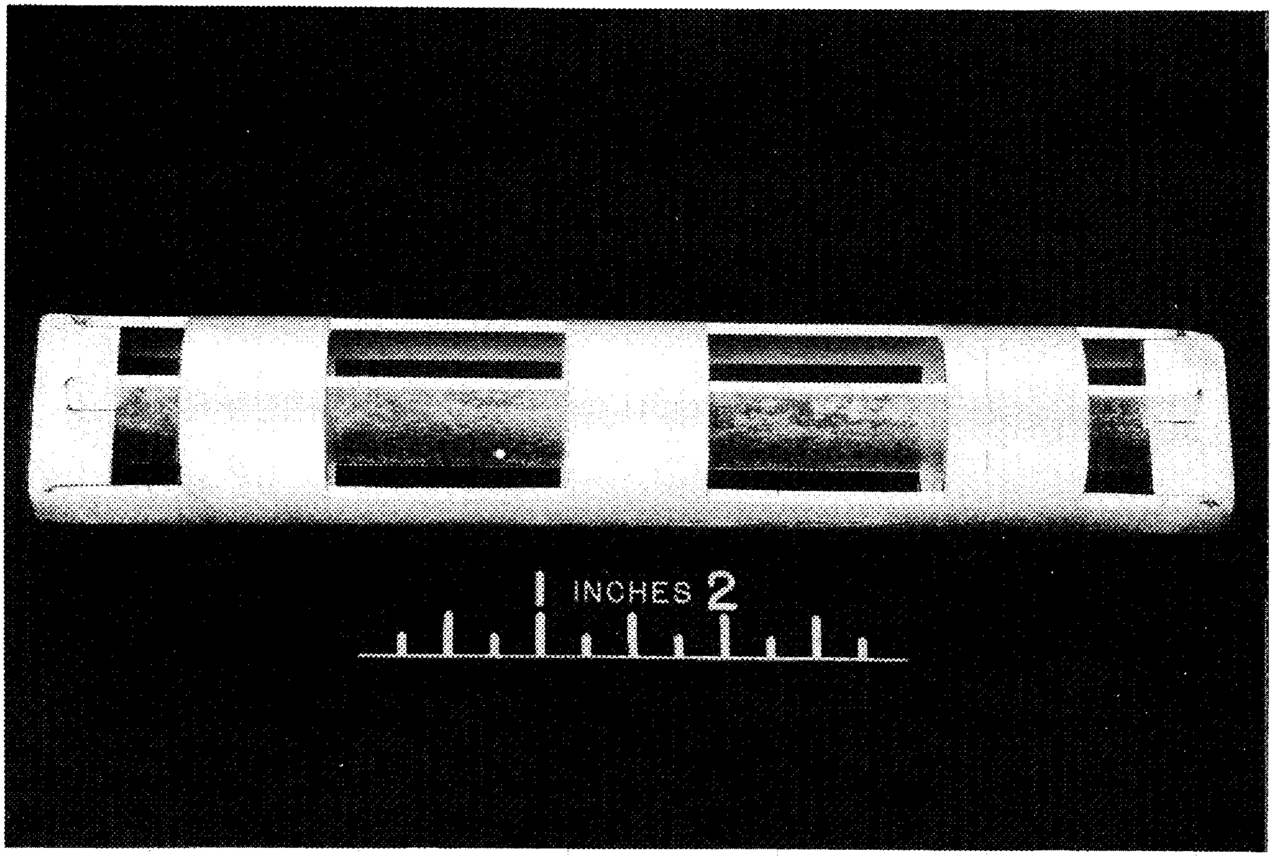

Fig. 17 Typical "Leaky Rabbit" Irradiation Test Assembly Showing UC_2–Al Miniature Fuel Plate in Aluminum Capsule.

APPENDIX A

MANUFACTURING PROCEDURES RECOMMENDED FOR PRODUCING MTR-TYPE FUEL ELEMENTS USING DISPERSIONS OF UC_2 IN ALUMINUM AS THE FUEL MATRIX

The following procedures detail the best practice for producing one 19-plate MTR-type fuel element containing 190 g of U^{235} using composite plates with UC_2-Al powder-metallurgy cores. This practice selects the most satisfactory techniques derived from the previously described experimental program and integrates them in established manufacturing procedures.

I. PRODUCT SPECIFICATIONS - CORE MATERIALS

A. $\underline{UC_2}$ - Use -100 mesh UC_2 produced by the reaction of UO_2 with C.[14] This material should contain no residual UO_2 as determined from x-ray spectrometer patterns and should contain at least 8.2 wt % carbon as determined by chemical analysis. For UC_2 containing 89.65% U and enriched 19.99% in the U^{235} isotope, 1060.01 g of carbide will be required.

B. Aluminum - Use -100 mesh aluminum having a typical analysis of 99.75 wt % Al and 0.25 wt % Si. The amount of aluminum required will be 729.6 g.

II. PROCESS SPECIFICATION - UC_2 PREPARATION

Fracture the massive UC_2 with impact blows and comminute the resultant chunks to -100 mesh by ball-milling in inert atmosphere. The milling time between subsequent screenings should be held to 10 – 15 min to minimize the number of fines, thus curbing the pyrophoric tendencies of the material. The UC_2 should be handled in an inert-gas-filled dry box during removal from the mill, screening, and transfer to storage containers. This minimizes radiological and pyrophoric hazards during processing. The sized material can then be stored in Mason jars in air.

III. PROCESS SPECIFICATION - CORE MANUFACTURE

A. In an appropriately vented glove box, weigh into 4-oz-wide-mouth bottles charges consisting of 55.79 g of UC_2 enriched 19.99% in the U^{235}

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[14] A. H. Daane et al., Preparation of Carbides of Uranium, ISC-11 (December 1947). Unclassified.

isotope and 38.40 g of aluminum. Cap the bottles and remove from the glove box. Nineteen such charges are required for one fuel element.

B. Seal the bottle cap with masking tape and simultaneously blend the individual charges on an oblique blender for 3 hr.

C. Press the blended charge into a fuel core 1.964 in. long and 2.264 in. wide x 0.278 in. thick using a 300,000-lb pressure on a hydraulic press. A 0.001-in.-thick aluminum foil should be pressed onto each side of the core. A mixture of stearic acid and acetone is used as the die lubricant.

D. Sinter the green cores at 600°C for 2 – 3 hr in a dynamic vacuum at less than 1μ of Hg. "Al-Si-Mag" ceramic is a suitable receptacle for the cores in the furnace during sintering.

E. Slightly bevel the edges of each core with a file and remove any free filings by wiping with an acetone-wet tissue.

IV. PRODUCT SPECIFICATION - COMPOSITE FUEL PLATE MATERIALS

A. Frames - Use 1100-grade aluminum - 19 required.

B. Cover Plates - Use 1100-grade aluminum - 38 required.

C. Fuel Cores - Use cores produced by techniques described in Section III - 19 required.

V. PROCESS SPECIFICATIONS - COMPOSITE FUEL PLATE MANUFACTURE

A. Shear from large sheets, picture-frame blanks of 1100 Al, 5 in. long x 4.25 in. wide x 0.250 in. ± 0.002 in. thick. Punch on a blanking press a window 2.30 in. wide x 2 in. long in the center of each frame blank. The long dimension of the window lies parallel to the width dimension of the frame. Degrease frames in vapor degreaser with trichloroethylene. Prepare 19 frames per fuel element.

B. Shear from large sheets, cover plates of 1100 Al, 5 in. long x 4.375 in. wide x 0.198 in. ± 0.001 in. thick. Thirty-four cover plates of this size (for short plates) are required per fuel element. Shear from large sheets, cover plates of 1100 Al, 5 in. x 4.375 in. wide x 0.295 in. ± 0.001 in. thick. Four cover plates of this size (for long plates) are required per fuel element. Degrease all cover plates in the vapor degreaser.

C. Press fuel cores (from Section III) in picture frames using approximately 260,000-lb pressure.

D. Scratch-brush one side of each cover plate.

E. Weld cover plates with scratch-brushed surface inward to each side of framed core by making an edge-fusion weld along the long edges of the frame only. Use no filler rod.

F. Preheat the assembled sandwich one-half hour at 600°C and roll according to the following schedule, reheating 5 min between passes:

1st pass - 15% reduction
2nd pass - 20% reduction
3rd pass - 20% reduction
4th pass - 20% reduction
5th pass - 25% reduction
6th pass - 25% reduction
7th pass - 25% reduction
8th pass - 27% reduction
9th pass - 30% reduction

The billets are turned end for end and reversed after each pass. Hot-finish short plates to a thickness 0.055 in. and long plates to a thickness of 0.070 in.

G. Paint cold plates with slurry of eutectic 190 flux in methyl alcohol. Dry at 180°C for one-half hour in aluminum pans and then flux-anneal for 1 hr at 605°C in the same pans.

H. Wash plates in hot water (60°C) to remove flux, etch in 15% HNO_3–5% HF aqueous solution for 10 min, rinse in hot water, and air dry.

I. Cold-roll short plates to 0.050 in. ± 0.001 in., cold-roll long plates to 0.065 in. ± 0.001 in.

J. Stress-relief anneal plates for 3/4 hr at 500 – 550°C. Reject any blistered plates.

K. Center fuel core in center of fuel plate using template and an x-ray fluoroscope outlining the appropriate area with a scribe. Reject any plates containing flaws in the core.

L. Shear plates to scribed dimensions.

M. Machine plates to required length and width, gang machining the width and length on a universal milling machine. The short plates are 2.796 in. wide and 25-5/8 in. long, while the long plates are 2.792 in. wide and 28-5/8 in. long. Seventeen short plates and two long plates are required per fuel element.

N. Degrease plates in vapor degreaser and remove burrs with a file.

O. Blister-check plates by annealing for 1 hr at 605°C. Reject any blistered plates.

P. Form plates to 5-1/2 in. radius of curvature between appropriate plattens on a hydraulic press.

Q. Degrease plates in vapor degreaser..

R. Acid-etch for 10 min in $HF-HNO_3$ aqueous solution, rinse in hot water, and dry in oven at 180°C.

VI. PRODUCT SPECIFICATION - FUEL ELEMENT COMPONENTS FOR BRAZING

A. Detailed drawing - Phillips Petroleum Company Dwg. MTR-E-2963.

B. Long Plates - processed per Section V - 2 required.

C. Short Plates - processed per Section V - 17 required.

D. Side Plates - 1100 Al braze, clad with Al-Si eutectic - 2 required.

E. Braze Metal - Use Al-Si eutectic composition alloy (11.6 wt % Si).

Thirty-four strips 0.008 in. thick x 0.100 in. wide x 26 in. long required for short plates and 4 strips 0.020 in. thick x 0.100 in. wide x 30 in. long required for long plates.

F. Combs - 1100 Al braze, clad with Al-Si eutectic - 2 required.

VII. PROCESS SPECIFICATION - FUEL ELEMENT ASSEMBLY BY BRAZING

A. Apply one strip of braze metal to each long edge of each fuel plate, anchoring in position by folding strip over end of the plate.

B. Paint side plates with eutectic 190 flux slurry and position in stainless steel assembly jig.

C. Apply flux slurry to the long edge of one short fuel plate by dipping into a small flux-containing trough. Insert fuel plate into groove in side plates.

D. Position first comb in comb holder of stainless steel assembly jig after fluxing.

E. Add remaining sixteen short fuel plates after fluxing as in step C.

F. Add fluxed second comb to assembly.

G. Insert appropriately fluxed long plates.

H. Tighten cap screws of assembly jig and check width dimensions with 3-in. micrometer.

I. Remove stainless steel-comb holder.

J. Place assembly in drying oven at 180°C for 1-1/2 hr.

K. Remove assembly from oven and separate element from stainless steel assembly jig.

L. Place element in ceramic (Al-Si-Mag) brazing jig and transfer to preheat furnace.

M. Hold in preheat furnace at 465°C for 1-1/2 hr.

N. Transfer to forced-air circulation brazing furnace and hold for 3/4 hr at 605°C.

O. Remove from brazing furnace and air-cool 5 min.

P. Remove brazed element from brazing jig - cool to room temperature.

Q. Wash in hot agitated water for 3/4 hr, acid-etch for 10 min, rinse, and dry in air blast.

R. Check critical external dimensions by micrometric measurements and check plate spacings with special measuring probe. Reject out-of-tolerance elements.

S. Attach appropriate end boxes by fusion welding and perform final machining operations to specifications.

T. Degrease and visually inspect element prior to shipment.

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