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FABRICATION OF DISPERSED URANIUM FUEL
ELEMENTS USING POWDER-METALLURGY TECHNIQUES

by

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FABRICATION OF DISPERSED URANIUM FUEL
ELEMENTS USING POWDER-METALLURGY TECHNIQUES

Stan J. Paprocki, Donald L. Keller, and George W. Cunningham

Abstract

Fabrication techniques for producing dispersion fuel elements with cores of 30 volume per cent of UC, U_2Ti , U_3Si , or U_6Ni dispersed in Zircaloy 2 and 30 volume per cent of UC or UN dispersed in Type 18-8 stainless steel have been investigated.

Roll-clad plate-type elements of all these compositions may be fabricated by powder-metallurgy methods in such a manner that good core-to-cladding bonds and cores with uniform dispersions of discrete uranium-compound particles are obtained.

From the standpoint of fabricability, elements containing UC in Zircaloy 2, UC in stainless steel, and UN in stainless steel are the most promising.

The UN in stainless steel has the best corrosion resistance in 680 F degassed water; however, UC in stainless steel has the best resistance to corrosion in 700 F NaK.

INTRODUCTION

Two important factors concerned with the development of fuel elements for service at moderate to high temperatures are the resistance to catastrophic failure due to pinholes in the cladding and the resistance to failure by radiation damage. Both of these problems may be at least partially solved by the use of powder-metallurgy techniques to produce dispersed uranium fuel-element cores.

In these fuel elements discrete particles of fuel material are placed in a continuous corrosion-resistant matrix. Thus the corrosion resistance of the core is increased and the radiation damage may be confined to local areas in the core. In addition, powder-metallurgy techniques allow close control of grain size and the use of fabrication temperatures low enough to prevent many reactions which would occur with normal fabrication procedures.

The selection of fuel and matrix materials to be used involves consideration of several factors. Some of these factors are weight percentage of uranium in the fuel, density of the uranium compound, compatibility of the fuel with the matrix at both fabricating and operating temperatures, and corrosion resistance of the matrix. On this basis, the following fuel-matrix combinations were selected as the most favorable for fabrication studies:

- (1) UN and UC dispersions in Type 18-8 stainless steel
- (2) UC, U_2Ti , U_3Si , and U_6Ni dispersions in Zircaloy 2.

The use of these uranium compounds allows a high fuel loading in a minimum volume, and for these studies a relatively high loading of 30 volume per cent of fuel material in

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the core was selected. With this loading, uranium-compound content varies from 42.5 to 53.6 w/o of the core and uranium content varies from 41.3 to 51.5 w/o of the core.

In the preparation of fuel elements by powder-metallurgy techniques, one of the most important variables is that of powder quality from batch to batch and process to process. Gas content (oxygen, nitrogen, and hydrogen) may vary greatly, and the size and shape of powder particles may be entirely different, depending upon the method used to produce the powder. Close control of these and other variables must be maintained if reproducible results are to be obtained. Therefore, considerable effort has been devoted to powder production.

The scope of this investigation has been broad and no attempt has been made to fully investigate physical properties, corrosion resistance, or resistance to irradiation. This investigation has been primarily concerned with the fabricability of the various dispersion elements. Preliminary corrosion tests have been completed and an investigation of resistance to irradiation is planned in a future correlated program.

POWDER PREPARATION

In the program as outlined in this report many combinations of fuel and matrix materials have been considered for use in dispersion fuel elements.

Initially, Type 18-8 stainless steel, Zircaloy 2, zirconium, and titanium were considered as matrix materials, and UC, UO₂, UN, UAl₂, UFe₂, USi₂, U₃Si, UNi₅, U₆Ni, UCu₅, U₂Ti, UC₂, U₅Sn₄, U₆Mn, U₆CO, and U₆Fe were considered for fuel materials. Of these materials, only U₃Si, UC, UN, U₆Ni, and U₂Ti, and Zircaloy 2 and Type 18-8 stainless steel, were selected for fabrication studies. Therefore, the preparation of these seven powders is discussed in more detail below.

Zircaloy 2

Zircaloy 2 powder of good quality may be prepared by hydriding chips, crushing them to the desired size, and dehydriding. However, gas contamination may be quite severe if the process is not carefully controlled.

The method for preparing Zircaloy 2 powder is similar to a hydride method previously developed⁽¹⁾ with the exception of the hydrogen source. The Battelle procedure uses the highly purified hydrogen obtained by the dissociation of UH₃. Thus, no contamination (within the range of accuracy for vacuum-fusion gas analysis) occurs during the hydriding operation. However, gas contamination does occur during the grinding and degassing operations.

The general procedure for the preparation of powders by the hydride method is illustrated in Figure 1. By the use of two UH₃ reaction tubes, a constant supply of

(1) References at end.

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FIGURE 1. SCHEMATIC DIAGRAM OF APPARATUS FOR HYDRIDING AND DEHYDRIDING METAL POWDERS

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hydrogen is assured. Uranium metal in one tube can be reacted with tank hydrogen to form UH_3 , while at the same time a previously evacuated tube of UH_3 is dissociated to form hydrogen which reacts with the material being hydrided. The rate of reaction can be followed by changes in hydrogen pressure.

In the case of Zircaloy 2, a series of experiments was run to establish optimum hydriding temperature and time, crushing procedure, and dehydriding temperature and time. In addition, tests were conducted to establish a satisfactory crushing procedure, while other tests were run to determine the effect of particle size on gas contamination.

Starting material consisted of a Zircaloy 2 ingot which had met WAPD specifications as to alloy content. The ingot was rolled to sheet of either 0.015 or 0.030 in. thickness and sheared to 1/2-in. squares. Before the squares were placed in the hydriding unit, they were pickled in a 46 volume per cent HNO_3 - 46 volume per cent H_2O - 8 volume per cent HF solution, rinsed in water, rinsed in acetone, and air dried. Gas analyses of the squares showed contents of 0.11 w/o oxygen, 23 ppm hydrogen, and 30 ppm nitrogen. This material was used for all Zircaloy 2-powder production.

The effect of process variables is shown in Table 1. The material listed as Lot 1 was hydrided by heating in a vacuum to 500 C and admitting hydrogen obtained from the decomposition of UH_3 . The hydride was crushed in an argon-filled dry box and then dehydrided at 800 C for 10 hr to a vacuum of 5×10^{-5} mm of mercury pressure. The second lot was used to determine contamination from grinding operations and was hydrided exactly as Lot 1, but was not dehydrided.

TABLE 1. EFFECT OF PROCESS VARIABLES ON GAS
CONTAMINATION OF ZIRCALOY 2 POWDER

Material	Vacuum-Fusion Analysis		
	Sample Weight, g	Oxygen, w/o	Hydrogen, w/o
<u>Lot 1</u>			
Wrought Zircaloy 2	0.470	0.11	0.0023
Massive hydride	0.132	0.098	1.6
Degassed powder	0.112	0.71	0.0047
<u>Lot 2</u>			
Massive hydride	0.107	0.12	-
Massive hydride	0.102	-	2.0
Ball-milled hydride	0.100	0.56	-
Ball-milled hydride	0.011	-	1.9
Hydride ground in steel mortar (-325 mesh)	0.100	0.21	-
Hydride ground in steel mortar (+325 mesh)	0.100	0.10	-

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From the examination of this table, it may be seen that oxygen contamination occurred both in the crushing and the degassing operations. A fivefold increase in oxygen occurred when the hydride was ball milled, even though a rubber-lined ball mill sealed in an argon atmosphere was used; however, contamination was greatly reduced when the hydride was crushed in an argon-filled dry box. A sevenfold increase in oxygen content occurred in the crushing and degassing of Zircaloy 2 hydride, even though the material was crushed in a steel mortar in an argon-filled dry box.

The available surface area appears to be an important factor concerning gas contamination. As shown in Table 1, hydride of plus 325-mesh size was not contaminated during the crushing procedure, while oxygen content of minus 325-mesh powder doubled. It is indicated in Table 2 that oxygen pickup during the dehydriding procedure is also dependent upon particle size.

TABLE 2. EFFECT OF PARTICLE SIZE ON GAS CONTAMINATION

Mesh Size	Degassing Conditions			Vacuum-Fusion Analysis		
	Time, hr	Temperature, C	Final Pressure, μ	Sample Weight, g	Oxygen, w/o	Hydrogen, w/o
-100 +200	6	700	3.1	0.101	0.24	0.024
-200 +325	6	700	0.5	0.106	0.52	0.008
-325	6	700	0.5	0.102	1.0	0.012

The two most troublesome variables encountered in the preparation of Zircaloy 2 powder are the time and temperature requirements for degassing. Tests in which these factors were varied gave the results shown in Table 3. The gas analyses reported indicate that less oxygen contamination occurs at the low temperature of 500 C, but that a higher temperature is necessary to dehydride the powder. As the time is increased, oxygen contamination increases. From the information given in Table 3, the decision was made to use the 700 C temperature and sufficient time to remove the major portion of hydrogen without obtaining high concentrations of oxygen. When the operation was scaled up from 30 to 100 g lots, it was found necessary to increase the time to 6 hr at 700 C.

Based on the information given above, the following procedure was used to prepare Zircaloy 2 powder for use in fabrication studies:

- (1) Roll Zircaloy 2 to 0.030-in. sheet and cut into 1/2-in. squares.
- (2) Pickle sheet in a 45 part H_2O -45 part HNO_3 -10 part HF solution, rinse in cold water, rinse in acetone, and air dry.
- (3) Heat in vacuum to 500 C.

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- (4) React with high-purity hydrogen obtained from thermal decomposition of UH_3 (the reaction is followed by means of an open-end mercury manometer).
- (5) Remove the hydride from the system under argon and place in argon-filled dry box.
- (6) Crush hydride in dry box to desired particle size by use of a steel mortar.
- (7) Degas hydride 6 hr at 700 C.
- (8) Screen to desired size fraction in argon atmosphere.

The procedure as outlined above results in powder of the gas content shown in Table 2, with nitrogen contents being less than 0.020 w/o. The analyzed alloy content was 1.35 w/o tin, 0.12 w/o iron, 0.06 w/o chromium, and 0.05 w/o nickel.

TABLE 3. EFFECT OF DEGASSING AND TEMPERATURE
ON GAS CONTAMINATION

Degassing Conditions				Vacuum-Fusion Analysis		
Mesh Size	Time, hr	Temperature, C	Final Pressure, μ	Sample Weight, g	Oxygen, w/o	Hydrogen, w/o
-200	1	500	>5.0	0.123	0.086	>0.9
-200	4	500	1.4	0.101	0.16	>0.9
-200	1	700	>5.0	0.102	0.085	0.37
-200	4	700	1.4	0.107	0.32	0.014
-200	4	700	1.4	0.100	0.24	0.011
-200	10	800	0.05	0.112	0.71	0.0047

A research group at KAPL working independently has reported the following information⁽²⁾ on Battelle Zircaloy 2 powder of minus 100 plus 200-mesh size. The powder was cold pressed at 46 tsi to a density of 77 per cent of theoretical and had excellent green strength. This material was extruded at 700 C and a pressure of 40 tons to a 12.7-to-1 reduction. The density of the extruded material was 6.54 g per cm^3 , and the ultimate tensile strength was reported as 109,000 psi, the reduction in area as 28.7 per cent, and the Vickers hardness as 267.2. The rate of corrosion after 168 hr in 680 F degassed water was reported as 12 mg per dm^2 .

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Stainless Steel

Materials used in the preparation of the stainless steel matrices were obtained from commercial sources. Powder of the Type 302B composition (18 w/o chromium-8 w/o nickel-1 to 2 w/o silicon-balance iron) was used in all cases where prealloyed stainless steel powder was required. The powder has good green strength and may be obtained in fine mesh sizes. For mixing a stainless steel matrix from elemental powders, minus 325-mesh electrolytic-iron powder, minus 325-mesh electrolytic-chromium powder, and minus 325-mesh carbonyl-nickel powder were used. All of the above powders were obtained from Charles Hardy, Inc., and were of the highest purity available.

Titanium

At the present time, titanium powder of good quality and in a limited range of particle sizes can be obtained from commercial sources. In addition, powder for use in this investigation was prepared by a hydride process similar to that used for the preparation of Zircaloy 2 powder.

Titanium sponge of high purity obtained from Du Pont was used as starting material. The sponge was rinsed in ether, air dried, and degassed 1 hr at 800 C in a vacuum of less than 10^{-4} mm of mercury. The sponge was then hydrided at 500 C. The optimum degassing conditions were found to be 5 hr at 600 C. By use of the procedure outlined above, titanium powder with 0.11 to 0.21 w/o oxygen, and 0.018 to 0.047 w/o hydrogen can be consistently produced.

Uranium-Titanium Compound (U_2Ti)

U_2Ti was prepared from Fernald high-purity uranium and Du Pont sponge titanium. Stoichiometric ratios of the two metals were melted in an argon atmosphere in an arc furnace using a tungsten electrode. Buttons were flipped and remelted three times to improve homogeneity.

Early work with U_2Ti involved the use of powder crushed from the as-cast structure. However, due to reactions with the Zircaloy 2 matrix, it became necessary to use a more homogeneous powder. Therefore, the cast button was annealed 24 hr at 800 C before comminution. The difference in structure is illustrated in Figure 2.

Due to the small quantities of powder required, all comminution was done in a steel mortar. The mortar and the U_2Ti were cooled in a dry ice-acetone mixture or in liquid nitrogen in order to facilitate crushing.

Some experimental work was also done with U_2Ti prepared by hydriding the alloy. However, the powder produced was in general smaller than the desired size and no extensive development program was carried out to improve the hydride-process powder. The hydride can be prepared by heating U_2Ti chips to 650 C in a vacuum, adding hydrogen obtained from thermal decomposition of UH_3 , and alternately cooling and heating to 650 C until a maximum amount of hydrogen is absorbed. The powder is degassed at 360 C for a period of 10 hr.

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100X

Etchant: Chromic Acid 1:8

N37133

As-Cast Ingot

250X

Etchant: Lactic Acid-HNO₃-HF

N37134

U₂Ti Powder Heat Treated 24 Hr at 800 C

FIGURE 2. EFFECT OF ANNEALING ON URANIUM-TITANIUM COMPOUND

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Effects of various procedures on the gas contamination of U_2Ti are illustrated in Table 4. Titanium contents varied from 8.2 w/o to 8.8 w/o (30.8 to 32.4 a/o) in the U_2Ti used in this investigation. Phase diagrams available at the time⁽³⁾ indicated a wide range for the U_2Ti intermetallic; therefore, no attempt was made to obtain alloys within an extremely close composition range. However, as shown in Figure 2, there are actually two phases present in the homogenized powder, even though the secondary phase is slight in amount.

TABLE 4. EFFECT OF VARIATIONS IN COMMINUTION
ON GAS CONTENT OF U_2Ti

Specimen	Analysis, ppm by weight			Remarks
	Oxygen	Hydrogen	Nitrogen	
Cast U_2Ti	190	10	30	--
Powder U_2Ti -1	800	45	-	Crushed in steel mortar in argon atmosphere
Powder U_2Ti -2	6300	130	100	Hydride method U_2Ti
Powder U_2Ti -3(a)	373	38	30	Crushed in air, mortar and powder cooled in liquid nitrogen
Powder U_2Ti -4(a)	64	0.5	-	From same U_2Ti button as Specimen U_2Ti -3, but crushed in argon atmosphere

(a) These specimens were homogenized 24 hr at 800 C before crushing to minus 100 plus 200-mesh powder.

Uranium Monocarbide (UC)

Several methods for the production of UC have been discussed in previously published reports. (4, 5, 6) The method selected for preparation of UC for use in this investigation consisted of mixing weighed amounts of uranium (Fernald melting stock) and high-purity graphite and melting this mixture in an argon-filled arc furnace by use of a graphite electrode. The graphite electrode was weighed before and after melting to record the carbon pickup by the melt. Tungsten electrodes consistently result in tungsten-contaminated UC.

This arc-melting procedure allows compounds of carefully controlled carbon content to be produced. In order to prevent reaction of carbon with either the Zircaloy 2 or stainless steel matrix, it is desirable to obtain an alloy with little or no free carbon. Uranium carbide used in this investigation contained <0.05 w/o free carbon, 4.5 w/o total carbon, 240 ppm oxygen, 8 ppm hydrogen, and 60 ppm nitrogen.

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Uranium monocarbide is a brittle compound and the powder is pyrophoric. Therefore, all UC powder was prepared by crushing the compound in a steel mortar in an argon-filled dry box. After this crushing procedure, the oxygen content increased to 895 ppm and the hydrogen content increased to 26 ppm.

Uranium Mononitride (UN)

The general procedure used for the preparation of UN was to nitride either uranium hydride or uranium metal to form U_2N_3 , which was later decomposed in a vacuum to form the stable UN powder. If the uranium were hydrided before the nitriding operation, a very fine UN powder would be obtained as a final product (minus 325-mesh size). For preparation of dispersion-type fuel elements, the uranium metal was nitrified directly in order to obtain a coarse powder (minus 100 plus 325-mesh size).

The exact procedure for preparation of UN used in this investigation is as follows:

- (1) Nibbled uranium (Fernald melting stock) approximately 1/4 by 1/4 by 1/8 in. was pickled in a 1:1 HNO₃-H₂O mixture, rinsed in water, rinsed in cp-grade acetone, and air dried.
- (2) Cleaned material was placed in a Vycor reaction tube, heated to 850 C in a vacuum, and nitrogen added to produce the U₂N₃ (the reaction is followed by means of an open-end manometer).
- (3) The U₂N₃ was transferred under an argon atmosphere to a BeO crucible.
- (4) The material was heated 1/2 hr at 1300 C in a vacuum.
- (5) This heating decomposed U₂N₃ to UN, with a final pressure of less than 10μ being obtained.

Use of the above procedure yielded UN with a typical analysis of 91.8 w/o uranium, 5.5 w/o nitrogen, and 0.150 w/o oxygen.

Uranium-Silicon Compound (U_3Si)

The process used to prepare a homogeneous U_3Si alloy has been discussed in a previous report. (7) The alloy was prepared by an induction-melting process. The material was held at the pouring temperature 5 min and after pouring it was slow cooled to prevent cracking. The alloy was then heat treated 1 week at 700 C to obtain a homogeneous epsilon structure.

The material was brittle at room temperature, but was too ductile to readily crush to a minus 100-mesh powder. Crushing in liquid nitrogen, as described in the section on U_2Ti , was found to give satisfactory yields for experimental purposes.

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STRAIGHT TO THE POINT

Figure 3 is a photomicrograph of the U_3Si -ingot material showing the single-phase structure which was present throughout the entire specimen. Analyses of the U_3Si gave values of >94.7 w/o uranium, 3.65 w/o silicon, 60 ppm nitrogen, 290 ppm oxygen, and 8 ppm hydrogen.

Uranium-Nickel Compound (U_6Ni)

Techniques for preparing U_6Ni are very similar to those used in the preparation of U_2Ti . Fernald melting-stock uranium and high-purity nickel were combined in stoichiometric ratio and arc melted under an argon atmosphere by means of a tungsten electrode. The button was flipped and remelted three times in order to improve the homogeneity.

The alloy was heat treated at 730 C for 24 hr in a vacuum to obtain a single-phase structure, and then crushed in a steel mortar which had been cooled in liquid nitrogen. Typical analyses of the crushed powder are as follows: >94.6 w/o uranium, 3.68 w/o nickel, 80 ppm nitrogen, 320 ppm oxygen, and 14 ppm hydrogen. The microstructure of the heat-treated alloy is shown in Figure 4.

Other Uranium Compounds

Other compounds which were investigated as possible fuel materials included UO_2 , UAl_2 , UFe_2 , USi_2 , UNi_5 , UCu_5 , UC_2 , and U_5Sn_4 . Many other compounds were considered but were not actually prepared because unfavorable physical properties such as low melting points or low decomposition temperatures limit their usefulness. Of the materials listed above, very little work was done with UO_2 because of extensive programs being conducted at Battelle and the major AEC installations. Although powders were prepared and some dispersion-type structures were prepared with the above-mentioned compounds, no extensive fabrication studies were made because of the low percentage of uranium present in these alloys as compared with UO_2 .

All of these materials other than UO_2 may be prepared by arc-melting techniques. U_5Sn_4 is very difficult to work with as it oxidizes readily in air even in the ingot form. UC_2 , UFe_2 , UAl_2 , and USi_2 are very brittle and may be easily reduced to powder; the USi_2 is extremely brittle. UNi_5 and UCu_5 are ductile and, consequently, are difficult to crush. For example, by means of a steel mortar in an argon-filled dry box, only 9 per cent of the UCu_5 was reduced to minus 150-mesh powder after 15-hr crushing time.

SELECTION OF FUEL-MATRIX COMBINATIONS

There are many possible fuel-matrix combinations that could be used for preparation of dispersion fuel elements. Commonly used matrix and cladding materials such as stainless steel or Zircaloy 2 appear to be the most feasible, and there are many uranium alloys and compounds which could be used advantageously as fuel in this type of structure.

500X

N37135

FIGURE 3. URANIUM-SILICON COMPOUND (U_3Si) HEAT TREATED 1 WEEK AT 800 C

500X

N37136

FIGURE 4. U_3Ni URANIUM-NICKEL COMPOUND HEAT TREATED 24 HR AT 730 C

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Materials initially considered for use as fuel materials included UC, UC₂, U₅Sn₄, UO₂, UN, UAl₂, UFe₂, USi₂, UNi₅, UCu₅, U₂Ti, U₃Si, U₆Ni, U₆Mn, U₆Co, and U₆Fe. Matrix materials were to include Zircaloy 2, titanium, and Type 18-8 stainless steel. Because of the increasing interest in reactor materials for service at high temperatures, a series of specimens was sintered at 2280 F and examined for compatibility. Specimens sintered were of the following combinations: UC, UO₂, UAl₂, USi₂, UFe₂, UNi₅, UCu₅, U₂Ti, UC₂, and UN dispersions in titanium; UC, UO₂, UAl₂, USi₂, U₂Ti, UC₂, and UN dispersions in Zircaloy 2; UC, UO₂, UAl₂, USi₂, UFe₂, UNi₅, UCu₅, and U₂Ti dispersions in stainless steel. After sintering, the materials were placed in protective steel jackets and hot rolled; Zircaloy 2- and titanium-matrix compacts at 1475 F, and stainless steel-matrix compacts at 2200 F. Metallographic examination of these specimens indicated that in most cases reaction occurred between matrix and fuel materials, exceptions being the UN-stainless steel system, and the UO₂-stainless steel system. However, almost all of the specimens were successfully hot rolled at temperatures indicated above. These tests indicated that high-temperature sintering could not be used as a method for producing dense Zircaloy 2- and titanium-base materials; however, it appeared feasible to fabricate some of these materials at lower temperatures, i. e., 1475 F.

Because of the necessity to limit the number of fuel-matrix combinations to be investigated, only the two most common matrix materials, Zircaloy 2 and stainless steel, were selected for further study. The fuel materials were selected on the basis of high density, high uranium content per unit volume, stability, melting temperatures, compatibility with matrix materials, and other physical and chemical properties. UO₂ was not considered for further study because of extensive programs in progress at Battelle and other major AEC installations. Table 5 lists some of the information used as a basis for fuel selection.

None of the compounds listed above UO₂ in Table 5 was considered for detailed fabrication studies due to the large volume of compound required to provide a constant weight of uranium. Of the compounds listed below UO₂, UC was selected as the most desirable uranium-carbon compound; UN, U₂Ti, and U₃Si were all selected for further study; and U₆Ni was selected as being representative of the compounds U₆Ni, U₆Co, U₆Fe, and U₆Mn. Because of low decomposition temperatures, no attempt was made to fabricate U₂Ti, U₃Si, or U₆Ni in stainless steel. UC was combined with both stainless steel and Zircaloy 2, while UN was used only with stainless steel.

GENERAL FABRICATION PROCEDURES

There are many possible shapes and forms in which dispersion-type fuel elements may be fabricated; however, this investigation has been directed toward the development of basic fabrication techniques for fuel elements of various combinations of materials. Therefore, a basic shape, the flat plate, was selected for all fabrication studies.

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TABLE 5. SOME PROPERTIES CONSIDERED IN THE SELECTION OF FUEL COMPOUNDS^(a)

Compound	Uranium, w/o	Density, g per cm ³	Uranium, g per cm ³	Uranium Increase, per cent per given volume	Crystal Structure	Melting Point, C	Thermal Neutron Absorption Cross Section of Second Element, barns per atom ⁽⁸⁾
UCu ₅	42.8	10.60	4.54	-52.7	FCC ⁽³⁾	1052 ⁽³⁾	3.69
UNi ₅	44.77	11.31	5.07	-47.1	FCC ⁽⁴⁾	1300 ⁽⁴⁾	4.6
UAl ₂	81.52	8.14 ⁽⁴⁾	6.64	-30.9	FCC ⁽⁴⁾	~ 1590 ⁽⁴⁾	0.230
USi ₂ (alpha)	80.91	8.98 ⁽⁴⁾	7.27	-24.2	BCT ⁽⁴⁾	~ 1600 ⁽⁴⁾	0.13
USi ₂ (beta)	80.91	9.25 ⁽⁴⁾	7.48	-22.1	Hex ⁽⁴⁾	~ 1600 ⁽⁴⁾	0.13
UFe ₂	68.0	13.21 ⁽⁴⁾	8.98	- 6.5	FCC ⁽⁴⁾	1235 ⁽⁴⁾	2.53
UO ₂	88.1	10.96 ⁽³⁾	9.6	0	Cubic ⁽³⁾	2875 ⁽³⁾	<0.0002
UC ₂	90.83	11.68 ⁽⁴⁾	10.61	+10.4	BCT ⁽⁴⁾	2450 - 2500 ⁽⁴⁾	0.0032
UC	95.19	13.63 ⁽⁴⁾	12.97	+35.4	FCC ⁽⁴⁾	2350 - 2400 ⁽⁴⁾	0.0032
UN	94.44	14.32 ⁽⁴⁾	13.52	+40.5	FCC ⁽⁴⁾	2650 ± 100 ⁽⁴⁾	1.88
U ₂ Ti	90.8	15.22	13.8	+43.7	Hex ⁽³⁾	~ 890 (decomposition) ⁽³⁾	5.6
U ₃ Si	96.21	15.58 ⁽⁴⁾	14.99	+56.2	BCT ⁽⁴⁾	930 (decomposition) ⁽⁴⁾	0.13
U ₆ Ni	96.05	17.6 ⁽⁴⁾	16.90	+76.0	BCT ⁽⁴⁾	790 (decomposition) ⁽⁴⁾	4.6
U ₆ CO	96.1	17.7 ⁽³⁾	17.00	+77.2	BCT ⁽⁸⁾	~ 830 (decomposition) ⁽³⁾	37.0
U ₆ Fe	96.2	17.7 ⁽⁴⁾	17.02	+77.5	BCT ⁽⁴⁾	815 (decomposition) ⁽⁴⁾	2.53
U ₆ Mn	96.2	17.8 ⁽³⁾	17.12	+78.7	BCT ⁽³⁾	~ 726 (decomposition) ⁽³⁾	13.2

(a) Superscript numbers are references listed at the end of the report.

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Core Fabrication

Preparation and comminution of metal powders has been discussed in the preceding section. All operations involving the handling of pyrophoric powders or powders which have a tendency to oxidize (most uranium compounds, Zircaloy 2, and titanium) were carried out insofar as possible in an argon-filled dry box (Figure 5).

In the dry box, powders were screened to the desired size, weighed to give the correct composition, and placed in glass containers. It was found necessary in some cases because of large differences in density of the two powders being mixed to add a dispersant or binder to insure a homogeneous mixture. The use of a dispersant resulted in a more uniform dispersion in all cases. An addition of less than 0.5 w/o of a 25 w/o camphor in methanol solution was found satisfactory for mixing all of the core materials used in this investigation. After the addition of the dispersant, the container was sealed in the argon atmosphere, removed from the dry box, and placed in a mixer.

The powders were mixed from 2 to 48 hr. The usual mixing time was 2 hr if the Kelly V-type blender was used, and 15 hr if a ball-mill type of mixer was used. When properly mixed, the powders were transferred to the dry box and weighed to the correct quantities for pressing. These preweighed units of powder were then transferred to a die constructed with interlocking split inserts to allow easy removal of the green pressed compact without ejection.

All cores were mixed to give a 30 volume per cent loading of the uranium compound, and all core mixtures were cold pressed at 50 tsi. The green strength of all cores was good. Variation of the green density for the different mixtures is shown in Table 6.

TABLE 6. COLD-PRESSED DENSITIES OF COMPACTS
PRESSED AT 50 tsi

Fuel	Matrix	Theoretical Density of Mixture, g per cm ³	Cold-Pressed(a) Density, g per cm ³	Per Cent of Theoretical Density
U ₂ Ti	Zircaloy 2	9.15	7.32	80.0
U ₃ Si	Zircaloy 2	9.25	7.30	79.0
U ₆ Ni	Zircaloy 2	9.85	7.60	77.2
UC	Zircaloy 2	8.67	6.80	78.5
UC	Type 302B stainless steel	9.62	7.56	78.6
UN	Type 302B stainless steel	9.82	7.63	77.7
UN	Type 18-8 elemental powder stainless steel	9.82	7.77	79.2

(a) Average values.

Green cores were placed in picture-frame packs for roll cladding, or were sintered or hot pressed before roll cladding.

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N28994

FIGURE 5. DRY BOX

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Roll-Cladding Techniques

The procedure used in roll-cladding studies involved placing the core in a picture-frame pack, welding the pack, evacuating and sealing the pack, hot rolling to the desired thickness, and cold rolling to improve the surface finish, if desired.

The two types of picture-frame packs used are shown in Figure 6. The double pack was used for Zircaloy 2-clad plates, and the single pack was used for stainless steel-clad plates. The packs were carefully cleaned before assembly according to the following procedure:

- (1) Remove grease, dirt, and rust by scrubbing parts in acetone with stiff brush or steel wool (for stainless steel).
- (2) Rinse in cold water.
- (3) Brush parts in warm soapy solution.
- (4) Rinse in cold water.
- (5) Rinse in warm water.
- (6) Rinse in cold water.
- (7) Dry with absorbent paper.

The assembled packs were arc welded in an argon atmosphere. The packs were then evacuated on a vacuum system which uses both a two-stage diffusion pump and a forepump. The packs were heated to 600 F to insure removal of any water, dispersant, or absorbed gases from the core. At a vacuum of less than 1×10^{-3} mm of mercury pressure the stems were heated and forged closed to seal the pack.

Packs of the size used shown in Figure 6 were preheated 30 to 45 min and heated 15 min between passes. In order to reduce end effects, the pack was flipped and reversed every pass. Satisfactory core-to-cladding bonds were achieved in most cases by rolling the packs to a 4-to-1 reduction and by using a heavy reduction of 30 per cent on the first pass and a 10 to 20 per cent reduction in thickness on the remaining passes.

After the rolling procedure, the packs were radiographed, and, in the case of Zircaloy 2 plates, the outside packs were removed.

SPECIAL FABRICATION TECHNIQUES

Because of differences in physical properties of both fuel materials and matrix materials, no exact fabrication procedure can be listed which would be satisfactory for producing all the dispersion-type fuel plates investigated. Many fuel and matrix combinations were considered, but only six combinations were selected for detailed fabrication studies. The results of these investigations are discussed in the following sections.

Stainless Steel

N38033

Zircaloy 2 Inserts, Mild Steel Outer Pack

N38032

FIGURE 6. PICTURE-FRAME PACKS FOR ROLL CLADDING ZIRCALOY 2- AND STAINLESS STEEL-MATRIX FUEL ELEMENTS

U₂Ti Dispersions in Zircaloy 2

There are several factors that increase the difficulty of fabricating U₂Ti-Zircaloy 2 dispersion fuel elements. At normal fabrication temperatures (1500 F), there is a partial reaction of the U₂Ti with Zircaloy 2. At lower temperatures (1300-1400 F), a good core-to-cladding bond cannot be readily attained by normal fabrication procedures. Furthermore, U₂Ti particles, even though brittle at room temperature, are fairly ductile at elevated temperatures and tend to elongate even at fabrication temperatures of 1300 F.

The first problem encountered was that of the reaction between U₂Ti and Zircaloy 2. It has been found that three factors which affect the reactivity of U₂Ti with Zircaloy 2 are particle size, method of U₂Ti-powder preparation, and fabrication temperatures. In specimens which contained U₂Ti of minus 150 plus 325-mesh size, the reaction was severe enough at 1500 F to prevent good core-to-cladding bonds in most cases. When a larger size U₂Ti powder was used a reaction occurred, but diffusion was hindered and the reaction was slowed down due to the smaller surface area of the U₂Ti. Examples of such structures are shown in Figure 7.

An attempt was made to decrease the reaction by improving the structure of the U₂Ti particles. The U₂Ti in the specimens discussed above was crushed from the as-cast condition. In an effort to improve the shape of the particle and reduce the gas contamination due to crushing, U₂Ti powder was prepared from as-cast material by the hydride method. As shown in Figure 8, little improvement was noted in the core structure; however, the core-to-cladding bond was improved. Also, by lowering the rolling temperature to 1400 F, the somewhat improved core structure of Figure 9 was obtained.

A series of experiments was then run using U₂Ti which had been homogenized by annealing 24 hr at 1470 F. Four U₂Ti-Zircaloy 2 compacts were pressed at 50 tsi, then two of the compacts were sintered for 2 hr; one each at 1500 and 1400 F. The remaining two compacts were sintered 4 hr; one each at 1300 and 1200 F. Microstructures shown in Figure 10 indicate that reaction occurs as low as 1300 F. However, by roll cladding green compacts, the time at temperature was reduced sufficiently to produce only a slight reaction at 1400 F (Figure 11) and 1350 F (Figure 12) and no reaction at 1300 F (Figure 16). The effect of time at temperature and particle size may be seen if Figures 13 and 14 are compared to Figure 11. Comparison of Figure 15 with Figure 16 also indicates that the particle size can be reduced without excessively increasing the U₂Ti-Zircaloy 2 reaction zone at 1300 F.

Even though the core structure was improved at lower temperatures, the core-to-cladding bond was usually very poor when standard roll-cladding procedures were used. At 1400 F, if the reaction was decreased by the use of homogenized U₂Ti of large particle size (minus 80 plus 100), as shown in Figure 11, the standard roll-cladding technique using green-core compacts was sufficient to produce plates with a good core-to-cladding bond; however, special techniques were required to bond the cladding to the core at 1300 F.

FIGURE 7. 49.5 w/o U₂Ti DISPERSED IN ZIRCALOY 2 AND ROLL GLAD AT 1500 F

FIGURE 8. 49.5 w/o U₂Ti DISPERSED IN ZIRCALOY 2 AND ROLL CLAD AT 1500 F

U₂Ti powder prepared by hydride method to minus 150 plus 325-mesh size.

FIGURE 9. 49.5 w/o U₂Ti DISPERSED IN ZIRCALOY 2 AND ROLL CLAD AT 1400 F

U₂Ti prepared by hydride method to minus 150 plus 325-mesh size.

250X

Etchant: Lactic Acid-HF-HNO₃

N37143

Sintered 2 Hr at 1200 F

250X

Etchant: Lactic Acid-HF-HNO₃

N37144

Sintered 2 Hr at 1300 F

100X

Etchant: Lactic Acid-HF-HNO₃

N37145

Sintered 2 Hr at 1400 F

500X

Etchant: Lactic Acid-HF-HNO₃

N37146

Sintered 4 Hr at 1500 F

FIGURE 10. 49.5 w/o U₂Ti DISPERSED IN ZIRCALOY 2 AND VACUUM SINTERED

U₂Ti hand crushed from homogenized ingot to minus 80 plus 100 mesh.

FIGURE 11. U₂Ti-ZIRCALOY 2 DISPERSION

Minus 80 plus 100-mesh U₂Ti
homogenized powder roll clad
at 1400 F.

FIGURE 12. U₂Ti-ZIRCALOY 2 DISPERSION

Minus 100 plus 200-mesh U₂Ti
homogenized powder roll clad
at 1350 F.

FIGURE 13. U₂Ti-ZIRCALOY 2 DISPERSION

Minus 60 plus 140-mesh U₂Ti
homogenized powder roll clad
at 1400 F after presintering
1 hr at 1400 F.

FIGURE 14. U₂Ti-ZIRCALOY 2 DISPERSION

Minus 200 plus 325-mesh U₂Ti
homogenized powder roll clad
at 1400 F.

FIGURE 15. U₂Ti-ZIRCALOY 2 DISPERSION

Minus 60 plus 140-mesh U₂Ti
homogenized powder roll clad
at 1300 F after presintering
1 hr at 1300 F.

FIGURE 16. U₂Ti-ZIRCALOY 2 DISPERSION

Minus 100 plus 200-mesh U₂Ti
homogenized powder roll clad
at 1300 F.

Three techniques which show very promising results are (1) use of a green-powder core, a green-powder frame, and wrought cover plates; (2) use of a green-powder core, a green-powder frame, and green-powder cover plates; and (3) hot pressing Zircaloy 2 and core components of (1) or (2) and hot rolling to the desired final thickness. The methods for fabricating by these three techniques are illustrated in Figure 17.

The composite core shown in Figure 17 part C was prepared by the following procedure: (1) the U_2Ti -Zircaloy 2 mixture was pressed in a 0.333 by 0.610-in. die at approximately 10 tsi, (2) this compact was then located in the center of a 1-in.-square die, (3) loose Zircaloy 2 powder was tapped to fill the area around the core, and (4) the composite compact was then pressed at 50 tsi.

For preparation of fuel plates by either Methods (1) or (2), the composite compact can be placed in a picture frame with powder or wrought-Zircaloy 2 cover plates and hot rolled at 1300 F. The process described as Method (3) above has not been thoroughly investigated. However, a composite core with green-powder cover plates may be hot pressed at 20 tsi and 1100 F to 95 per cent of theoretical density. This hot-pressed pack can then be rolled either bare or in a picture-frame pack at 1300 F or even as low as 1100 F to a 4-to-1 reduction.

Of the three methods described, Method (1), which uses a powder frame and wrought cover plates, has been most thoroughly investigated. A good core-to-cladding bond can be obtained and a structure similar to that shown in Figure 18 consistently produced. Methods (2) and (3) also appear promising, as shown by microstructures in Figures 19 and 20.

U_3Si Dispersions in Zircaloy 2

Problems encountered in the roll cladding of U_3Si -Zircaloy 2 cores were similar to those described in the section on U_2Ti -Zircaloy 2 dispersions. There was some reaction between U_3Si and Zircaloy 2 at 1500 F and U_3Si was ductile even at temperatures as low as 1300 F.

All U_3Si used in this investigation was crushed to the desired size in a steel mortar. After mixing and pressing at 50 tsi, green compacts were placed in picture-frame packs and roll clad at 1500, 1400, and 1300 F. Typical core structures are shown in Figure 21. At 1500 F there was indication of a U_3Si -Zircaloy 2 reaction zone, while at 1400 F little or no reaction occurred. However, it should be noted that in both cases the ductility of the U_3Si was increased enough to result in elongated and partially interconnected U_3Si particles. At 1300 F, the structure was somewhat improved, but the U_3Si particles were elongated and interconnected in some cases.

Normal fabrication procedures, using Zircaloy 2 inserts and mild steel outside picture frames, were used for roll cladding at 1500 and 1400 F, but Zircaloy 2 powder frames and wrought cover plates were used in roll cladding at 1300 F. At all three temperatures good core-to-cladding bonds were obtained.

A-22974

FIGURE 17. COMPONENT PARTS FOR ROLL CLADDING ZIRCALOY-MATRIX FUEL ELEMENTS

Part A may be either green-powder compacts or wrought Zircaloy 2 sheet. Part C is a composite pack with center section of uranium fuel dispersed in Zircaloy 2 and an outer frame of Zircaloy 2. Parts A and C may be assembled as green compacts in the mild steel frame (B, D) or may be hot pressed into a single unit before assembly.

FIGURE 18. U₂Ti-ZIRCALOY 2 DISPERSION ROLL CLAD AT 1300 F
Minus 100 plus 200-mesh U₂Ti homogenized powder.

FIGURE 19. 50 w/o U₂Ti DISPERSED IN ZIRCALOY 2 AND ROLL CLAD AT 1300 F
WITH ZIRCALOY 2 POWDER FRAME AND COVER PLATES

FIGURE 20. U₂Ti-ZIRCALOY 2 DISPERSION

Minus 100 plus 200-mesh U₂Ti.
Core, frame, and cladding
components were pressed from
powder, hot pressed into one
compact at 1100 F and rolled
at 1100 F.

100X

Etchant: Lactic Acid-HF-HNO₃

N37155

a. Minus 150 Plus 270-Mesh U₃Si
Roll Clad at 1500 F.

100X

Etchant: Lactic Acid-HF-HNO₃

N37156

b. Minus 150 Plus 270-Mesh U₃Si
Roll Clad at 1400 F.

100X

Etchant: Lactic Acid-HF-HNO₃

N37157

c. Minus 100 Plus 200-Mesh U₃Si Roll Clad at 1300 F

FIGURE 21. 50.4 w/o U₃Si IN ZIRCALOY 2 DISPERSIONS

One cladding procedure which gave a good core-to-cladding bond and a uniform dispersion of discrete U_3Si particles was that of hot pressing a composite-powder compact of a U_3Si -Zircaloy 2 core, a Zircaloy 2 frame, and a Zircaloy 2 cladding at 1150 F and 20 tsi. The resultant structure is illustrated in Figure 22. The procedure used for fabricating this compact and that shown in Figure 21c is described in the section on U_2Ti -Zircaloy 2 dispersions.

U_6Ni Dispersions in Zircaloy 2

U_6Ni -Zircaloy 2 cores may be fabricated in the same manner as U_2Ti -Zircaloy 2 and U_3Si -Zircaloy 2 cores. However, U_6Ni decomposes at 1454 F and there is a eutectic between U_6Ni and U_7Ni_9 at 1364 F; therefore, reactions between Zircaloy 2 and U_6Ni or nickel-rich U_6Ni are possible at quite low temperatures.

The U_6Ni actually used contained 3.68 w/o nickel. Therefore no liquid phase was present below 1454 F. However, there was evidence of a reaction between Zircaloy 2 and U_6Ni in specimens roll clad at 1300 F. Specimens were roll clad at 1300 F by the composite-powder frame and core method using wrought cover plates and green-powder frames as described in the section on U_2Ti -Zircaloy 2 structures. The resultant core structure is shown in Figure 23. A good core-to-cladding bond was obtained. Lower fabrication temperatures can be used by hot pressing a green composite core and green cover plates at 1150 F, placing in a picture-frame pack, and rolling at 1100 F. A microstructure of a specimen fabricated in this manner is also shown in Figure 23.

UC Dispersions in Zircaloy 2

Similar problems were encountered in the preparation of UC-bearing dispersion fuel elements regardless of whether the matrix material was stainless steel or Zircaloy 2. At higher temperatures there was a reaction or diffusion of carbon into the matrix. At lower temperatures where no reaction and little diffusion takes place, the matrix was not ductile enough to prevent fracturing of the brittle UC particles during the roll-cladding operation.

UC-Zircaloy 2 compacts have been roll clad at 1500, 1400, and 1300 F. A sound core-to-cladding bond was obtained at all three temperatures; however, there was a difference in the core structure. At 1500 F, as illustrated by Figure 24, fracturing of the particles was not severe and because the green compact was used as a core, the time at temperature was short and there was apparently little or no carbon diffusion. On the other hand, at 1300 F, nearly all UC particles fractured.

Also shown in Figure 24 is a specimen prepared by hot pressing the core and frame at 1300 F, then hot rolling at 1300 F. This method has not been thoroughly investigated, but indications are that this procedure could be used at higher temperatures with good results.

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100X

Etchant: Lactic Acid-HF-HNO₃

N37158

Composite Core and Cladding as Hot Pressed at 1150 F

100X

Etchant: Lactic Acid-HF-HNO₃

N37159

Composite Core and Cladding After Rolling at 1100 F to 4-to-1 Reduction

FIGURE 22. 50.4 w/o U₃Si IN ZIRCALOY 2 DISPERSION FABRICATED BY HOT-PRESSING TECHNIQUES

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100X

Etchant: Lactic Acid-HF-HNO₃

N37160

Minus 100 Plus 200-Mesh U₆Ni Homogenized Powder
Roll Clad at 1300 F

100X

Etchant: Lactic Acid-HF-HNO₃

N37161

Minus 100 Plus 200-Mesh U₆Ni Homogenized Powder
Hot Pressed at 1150 F, and Rolled at 1100 F

FIGURE 23. 53.6 w/o U₆Ni DISPERSED IN ZIRCALOY 2

100X

Etchant: Lactic Acid-HF-HNO₃

N37162

Minus 140 Plus 270-Mesh UC Green Compact
Roll Clad at 1500 F

100X

As Polished

N37163

Minus 100 Plus 200-Mesh UC Green Compact
Roll Clad at 1400 F

100X

Etchant: Lactic Acid-HF-HNO₃

N37164

Minus 100 Plus 200-Mesh UC Green Compact
Roll Clad at 1300 F

100X

Etchant: Lactic Acid-HF-HNO₃

N37165

Minus 100 Plus 200-Mesh UC Compact Hot Pressed
at 1300 F and Roll Clad at 1300 F

FIGURE 24. 47.2 w/o UC DISPERSED IN ZIRCALOY 2

Figure 25 shows the type of structures which can be obtained by hot rolling green compacts at 1500 F, if a closely controlled particle size of UC is used. From these results it appears that a temperature of 1500 F is sufficiently high to prevent excessive fracturing of UC, and that the use of green compacts permits short enough times at temperature that little carbon diffusion can take place.

100X Etchant: Lactic Acid — HF-HNO₃ N37166

FIGURE 25. 47.2 w/o UC DISPERSED IN ZIRCALOY 2 WITH ZIRCALOY 2 POWDER FRAMES AND CORE

Compact hot rolled at 1500 F.

UC Dispersions in Stainless Steel

Two problems have been encountered in the preparation of UC-stainless steel dispersion cores: (1) at temperatures of 1900 F and above there was a reaction between the UC and the Type 302B stainless steel matrix, (2) the Type 302B stainless steel matrix was not ductile enough to prevent heavy fracture of UC particles at rolling temperatures of 1900 F and below.

UC-stainless steel dispersions have been roll clad at temperatures of 1700, 1800, 1900, and 2000 F. Typical structures are shown in Figure 26. A slight reaction occurred at 1900 F, which was greatly increased at 2000 F. There was no sign of a reaction at 1700 or 1800 F; however, fracturing of UC particles was increased and a good core-to-cladding bond could not be obtained at 1700 F. At 1800 F, the temperature selected as most favorable, a series of specimens was run to determine the effect of UC particle size on the structure. As shown in Figure 27, UC of a minus 100 plus 150-mesh size gives an improved structure.

100X

As Polished

N37167

Minus 100 Plus 200-Mesh UC Roll Clad at 1700 F

100X

As Polished

N37168

Minus 100 Plus 150-Mesh UC Roll Clad at 1800 F

100X

As Polished

N37169

Minus 100 Plus 150-Mesh UC Roll Clad at 1900 F

100X

Marbles Etch

N37170

Minus 100 Plus 150-Mesh UC Roll Clad at 2000 F

FIGURE 26. 42.5 w/o UC DISPERSED IN TYPE 302B STAINLESS STEEL

All cores in green-pressed condition before roll cladding.

100X

As Polished

N37168

Minus 100 Plus 150-Mesh UC

100X

As Polished

N37171

Minus 100 Plus 325-Mesh UC

100X

As Polished

N37172

Minus 200 Plus 270-Mesh UC

100X

As Polished

N37173

Minus 270 Plus 325-Mesh UC

FIGURE 27. 42.5 w/o UC DISPERSED IN STAINLESS STEEL AND ROLL CLAD AT 1800 F

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UN Dispersions in Stainless Steel

Uranium mononitride may be readily incorporated in a stainless steel matrix as discrete particles (30 volume per cent loading) and a good core-to-cladding bond obtained.

In the fabrication of UN-stainless steel cores, it was essential that the UN be properly prepared. If a partially reduced U_2N_3 is used, a reaction may occur at the fabrication temperatures (2000 to 2200 F). A stainless steel matrix of either prealloyed powder (Type 302B) or an iron-18 w/o chromium-8 w/o nickel mixture can be used. Particles of UN were discrete and were not severely fractured in either case; however, use of elemental-powder stainless steel appeared to give a more uniform dispersion. Figure 28 shows typical core structures of UN dispersed in a Type 302B stainless steel matrix and an elemental-powder stainless steel matrix. The packs were rolled at 2000 F.

CORROSION RESISTANCE

In general, bare cores of the six combinations studied had poor corrosion resistance to 680 F degassed water, but had good corrosion resistance in 680 F NaK.

Corrosion in 680 F Degassed Water

Specimens were run both as cladding-defected specimens and as specimens having two edges of the core exposed.

Little work has been done on the corrosion resistance of the uranium fuels used in this investigation. UC decomposes in water at 83 C⁽⁴⁾ and U_3Si corrodes at a rate of $-1.00 \text{ mg}/(\text{cm}^2)(\text{hr})$ in 650 F degassed water⁽⁴⁾. From work done in this investigation, it appears that UN is the most stable compound investigated and that U_2Ti and U_6Ni have poorer resistance to 680 F water than U_3Si .

A series of specimens containing cores of 30 volume per cent U_2Ti , U_3Si , UC, or U_6Ni dispersed in Zircaloy 2 and 30 volume per cent UN or UC dispersed in stainless steel was placed on test on 680 F water with two core surfaces exposed. After 7 days, all specimens except UN in stainless steel were removed from test. The cores of all specimens removed from test were badly ruptured; however, the core-to-cladding bond was intact. The worst specimen was that of U_2Ti in Zircaloy 2. The core completely disintegrated. The best specimen was UN in the stainless steel, and, although small cracks developed in the core, no bulging or swelling was evident. After 155 days on test, there was no further change in appearance regardless of whether elemental or prealloyed stainless steel powder had been used as a matrix.

In the specimens which were completely clad except for a 0.040-in.-diameter drilled cladding defect, an examination after 12 hr indicated that all specimens except

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Type 302B Prealloyed Stainless Steel Powder Used as Matrix

Iron-18 w/o Chromium-8 w/o Nickel Elemental-Powder Mixture
Used as Matrix

FIGURE 28. 43.7 w/o UN DISPERSED IN STAINLESS STEEL AND ROLL CLAD AT 2000 F

Minus 100 plus 325-mesh UN.

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UN in stainless steel had been attacked. Blisters could be seen on all specimens except UN in stainless steel, which remained on test in 680 F water for 3 weeks without evidence of attack.

Corrosion in 700 F NaK

Specimens of the six combinations mentioned in the preceding section were completely immersed in NaK and held at a temperature of 700 F for 500 hr. Little or no change was noted in outward appearance of the specimens even though two core surfaces were exposed.

Metallographic examination indicated that the UC-stainless steel structure was most resistant, with little or no attack on the UC particles. UN particles were only slightly attacked, but the NaK penetrated to a depth of 0.005 in. into the core with a prealloyed stainless steel matrix and slightly more with an elemental-powder stainless steel matrix. The U_2Ti -Zircaloy 2 core had the poorest corrosion resistance, with the core being cracked and ruptured to a depth of approximately 0.010 in. However, there was little change in the outward appearance.

DISCUSSION OF RESULTS

By comparing each of the six types of elements fabricated, the one element which appears to be the most promising is that of UN in stainless steel, both from ease of fabrication and corrosion resistance. Although the exposed core does show microcracks when exposed to 680 F water, the resistance to 700 F NaK is good, and the UN-stainless steel dispersion is uniform, with discrete UN particles and no evidence of stringing. The appearance of the core is better than UO_2 in stainless, regardless of whether prealloyed- or elemental-powder stainless steel is used for a matrix.

From appearance of the fabricated core and its corrosion resistance to NaK, the UC dispersed in either stainless steel or Zircaloy 2 should be considered as promising. The U_2Ti , U_6Ni , and U_3Si dispersions are more difficult to fabricate and have poor resistance to 680 F water. However, because this investigation has not been directed toward the development of a fuel element for a particular reactor, none of these six types is being dropped from consideration for future development studies regardless of the fact that most of these elements have poor corrosion resistance to 680 F water.

In order to obtain reproducible results, powder of similar characteristics must be used for fabrication studies. By using a closely controlled powder-production method, by obtaining chemical analyses on each batch of powder, and by determining screen analyses, it has been possible to reduce any variables which might normally be due to the use of different batches of powder. However, even when good-quality powder is used, the method in which Zircaloy 2 is fabricated may affect both the physical properties and the corrosion resistance.

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For example, even though a U_3Si -Zircaloy 2 core compacted by hot pressing at 1150 F had better corrosion resistance than cores compacted by rolling at 1300 to 1500 F, the Zircaloy 2 matrix and cladding contained oxides in the grain boundaries. Thus, low-temperature techniques used to improve the core dispersions may be decreasing mechanical strength and corrosion resistance by preventing complete diffusion between the Zircaloy 2-powder particles.

On the other hand, it appears feasible to improve the strength of the hot-pressed compacts by hot rolling or heat treatments at low temperatures (1100 F).

CONCLUSIONS

- (1) Roll-clad plate-type fuel elements may be fabricated by powder-metallurgy techniques to contain up to 30 volume per cent of the following uranium compounds dispersed in a matrix as indicated:

<u>Fuel</u>	<u>Matrix</u>
U_3Si	Zircaloy 2
U_6Ni	Zircaloy 2
U_2Ti	Zircaloy 2
UC	Zircaloy 2
UC	Type 302B stainless steel
UN	Type 302B stainless steel or elemental-powder mixtures

- (2) Fabrication methods can be controlled such that discrete particles of the above-named uranium compounds are dispersed homogeneously in the matrix and such that a good core-to-cladding bond is obtained.
- (3) UC dispersed in Type 302B stainless steel has the best corrosion resistance to NaK of all specimens tested. None of the other combinations was severely attacked.
- (4) At a 30 volume per cent loading, UN in stainless steel is the only core material investigated that appears to have favorable resistance to 680 F degassed water.
- (5) When a high loading (30 volume per cent) of a uranium compound, such as U_2Ti , with poor corrosion resistance in 680 F water is dispersed in Zircaloy 2, little or no change in corrosion resistance occurs with a change in fabrication procedures.

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- (6) It is important to note that only high loadings of dispersed particles were investigated in this program. Other related work with dispersed fuel materials has clearly demonstrated that a substantial improvement in corrosion resistance would be obtained as the volume loading of the highly corrosive fuel dispersant was reduced.
- (7) Good-quality Zircaloy 2 powder may be prepared by the hydride method.

FUTURE WORK

Future work already planned includes irradiation of clad specimens prepared by the fabrication methods described in this report.

At the same time, some fabrication work will continue in an effort to improve corrosion resistance in 680 F water. Such work should include heat treatments and variations in the method used to densify compacts in order to improve the corrosion resistance of the matrix.

In addition, reduction of uranium loadings should be considered for some of the more corrosive core structures. For example, a common fuel element consists of 30 w/o UO_2 in stainless steel or 23.6 volume per cent UO_2 in the core; the same amount of fuel could be provided by use of 29.1 w/o U_2Ti in Zircaloy 2, which uses only 14.9 per cent of the core volume. At the same uranium loading, required volume percentages of other uranium compounds are as follows: 18.3 volume per cent UC or 17.6 volume per cent UN in stainless steel; 15.5 volume per cent UC, 13.7 volume per cent U_3Si , or 12.3 volume per cent U_6Ni in Zircaloy 2. Thus, it may be feasible to produce an element with a useful loading and good corrosion resistance, even though corrosion resistance to 680 F water as reported herein is poor.

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