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The Properties and Irradiation Behavior
of Carbide Fuels:
A Literature Survey

UNITED STATES
ATOMIC ENERGY COMMISSION
CONTRACT W-7405-ENG. 36

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Printed in the United States of America. Available from
Clearinghouse for Federal Scientific and Technical Information
National Bureau of Standards, U. S. Department of Commerce
Springfield, Virginia 22151

Price: Printed Copy \$3.00; Microfiche \$0.65

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of the
University of California
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Report written: October 6, 1967

Report distributed: March 14, 1968

The Properties and Irradiation Behavior
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by

Felix B. Litton

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THE PROPERTIES AND IRRADIATION BEHAVIOUR OF CARBIDE
FUELS: A LITERATURE SURVEY

Felix B. Litton

ABSTRACT

No major deficiencies for fast reactor applications were found in the reported properties of carbide fuels. It appears probable that container compatibility problems can be reduced by using carbide compositions that are strictly stoichiometric, or buffered. Free metal in carbon-deficient fuel reacts with iron- or nickel-base containers to form low-melting alloys. In sodium-bonded configurations, carbon in excess of stoichiometric monocarbide is transferred to cladding materials that contain elements forming carbides more stable than the fuel carbide. No information was found on heat transfer or sodium stability in sodium-bonded fuel elements.

Irradiation data indicate that fuel swelling depends on the carbon content, operating temperature, and burnup. Fuel swelling is a linear function of temperature to 1800°F, after which it is an exponential function. Fission gas release is related to the integrity of the fuel structure, the operating temperature, and burnup. The retention of fission gas appears to be satisfactory in mixed fuels up to about 50,000 MWD/T burnup. Strong containers that are stable to radiation effects will be required for burnups approaching 100,000 MWD/T. Metallurgical advances are anticipated from current carbide development programs; it is recommended that their economic significance be evaluated in comparison to the total processing cycle visualized for other competitive fuel systems. No clear-cut analysis appears in the literature.

INTRODUCTION

This report summarizes information on the properties and behavior of carbide fuels relative to their use in fast reactors. The literature was evaluated to June 1967. For brevity, only information considered to be the most reliable was selected for inclusion. Techniques of preparation and fabrication are not evaluated because the report is directed toward delineating more basic problems.

PHYSICAL AND MECHANICAL PROPERTIES

Uranium Monocarbide

The properties of uranium carbide as reported since 1949 are not consistent, probably because of the difficulty of obtaining pure material. The UC structure is face-centered cubic, NaCl type. The lattice parameter¹ is $4.9605 \pm 0.0002 \text{ \AA}$, and the x-ray density² is 13.63 g/cm^3 . The melting temperature is $2495 \pm 30^\circ\text{C}$.³ The structure is isomorphous

with UO and UN; solid solutions are formed with these compounds.^{4,5,6,7} Although the solubility of uranium in stoichiometric monocarbide is restricted below 500° and above 2100°C, a maximum of 2 at. % of the metal is soluble at 1700 ± 30°C.⁸ Uranium dicarbide is observed in the microstructure of uranium monocarbide fabricated with excess carbon.

The heat of formation of uranium monocarbide is reported⁹ as -23.2 ± 0.5 kcal/mole. The standard free energy of formation is reported as -21.0 ± 1.0 kcal/mole by Farr et al.¹⁰ A value of 11.3 ± 0.5 eu was assigned to the entropy.¹¹ An acceptable value for the specific heat is 0.048 ± 0.003 cal/°C g (at 125°C).^{12,13}

Frost¹⁴ reported the diffusion coefficient for carbon in uranium carbide as $0.02 \exp(-50000/RT)$ cm² sec⁻¹ at 1200°-1940°C and that for uranium in uranium carbide as $0.0012 \exp(-64000/RT)$ cm² sec⁻¹ at 1600°-2120°C. Griffiths¹⁵ reported the activation energies for formation and movement of vacancies as 2.8 ± 0.1 eV and 1.1 ± 0.1 eV, respectively.

Wheeler¹⁶ assumed a specific heat of 0.05 cal/°C g and calculated thermal conductivities (k) at 2300°K of 0.19, 0.15, and 0.12 W/cm-°C for arc-cast alloys containing 49.5, 51, and 52 at. % carbon, respectively. The thermal conductivity of sintered UC (94% density) was found to be 0.16 W/cm-°C. The thermal diffusivity as reported by Wheeler on 49.5 at. % carbon alloy increased nonlinearly from 0.049 cm² sec⁻¹ at 1400°K to 0.067 cm² sec⁻¹ at 2200°K and remained constant for a further temperature rise. Leary et al.¹⁷ reported thermal conductivity at 400°C of 0.054 cal/cm-°C-sec (0.23 W/cm-°C) which is in agreement with the value reported by Secrest et al.¹⁸

The coefficient of linear thermal expansion on heating cast 4.8 wt % carbon alloy varied from 9.5 to $13.0 \times 10^{-6}/^{\circ}\text{C}$ from room temperature to 950°C.¹⁸ Mendez-Peñalosa et al.¹⁹ reported a value of $11.3 \times 10^{-6}/^{\circ}\text{C}$ for UC. The electrical resistivity of cast carbides was found to vary from about 40 μΩ cm for UC to about 90 μΩ cm for UC₂.²⁰

A hardness of 600 kg/mm² is reported for cast uranium monocarbide containing 50 at. % carbon.²¹ A hardness of 935 kg/mm² (consistent with other values in the literature) was reported by Tripler et al.²² The average compressive strength is 51,100 psi, and Young's modulus is 29.0×10^{-6} psi.²¹

Uranium Sesquicarbide

The structure of U₂C₃ is body-centered cubic, space group symmetry 143d, with an x-ray density of 12.88 g/cm³.^{23,24} Although the sesquicarbide is not observed in the microstructure of uranium-carbon alloys of interest to the fuel technologist, it may be observed in the microstructure of carbon-rich uranium-plutonium monocarbide. Imoto et al.²⁵ studied the critical temperature below which the formation of U₂C₃ occurred either by the decomposition of tetragonal UC₂ or by the combination of UC₂ with UC. The temperature was observed to be between 1500° and 1600°C.

Huber et al.¹¹ have reported the thermochemical properties of uranium sesquicarbide. The heat of formation (ΔH_{298}°) is $-49,000 \pm 4,000$ cal/mole; the free energy of formation (ΔF_{298}°) is $-48,000$ cal/mole; the entropy of formation (ΔS_{298}°) is -3 eu; and the entropy (S_{298}°) is 25 eu. Behl et al.²⁶ reported the free energy for the reaction $2 \text{ U}(\beta) + 3 \text{ C}(\text{gr}) = \text{U}_2\text{C}_3(\text{s})$ as $(\Delta F_T^{\circ}) = 43,860 - 7 T$ kcal (± 0.2).

The coefficient of thermal expansion up to 1000°C is $10.49 \times 10^{-6}/^{\circ}\text{C}$ as reported by Norreys et al.²⁷ A hardness of 1100 kg/mm² was reported in reference 21.

Uranium Dicarbide

According to the U-C phase diagram,²⁸ uranium dicarbide exists in two polymorphic forms. The high-temperature form ($> 1820^{\circ}\text{C}$) is face-centered cubic, which undergoes a martensitic type of transformation on cooling to the body-centered tetragonal form.²⁹ The cubic form has the composition UC_{1.86 ± 0.02} and the tetragonal form has the composition UC_{1.96 ± 0.04}.²⁵ Andon et al.³⁰ reported the higher carbide to have the formula UC_{1.91} near room temperature. The lattice parameters were reported by Imoto et al.²⁵ to be $a_0 = 3.517 \text{ \AA}$ and $c_0 = 5.979 \text{ \AA}$ on quenched specimens and $a_0 = 3.523 \text{ \AA}$ and $c_0 = 5.995 \text{ \AA}$ on slowly cooled specimens. Lattice-parameter measurements at elevated temperature showed unusual behavior in the tetragonal modification, presumed to be associated with departure from stoichiometry.²⁴ The x-ray density is 11.68 g/cm³.¹⁴ By x-ray analyses, Litz et al.³¹ determined the structure of UC₂ to be the CaC₂ structure (14/mmm), which was confirmed by neutron diffraction analyses conducted by Atoji et al.³²

The heat content for tetragonal UC_{1.93} was

determined by Levinson as $H_T - H_{310^\circ K}^\circ = 20.21 + 7.978 \times 10^{-4} T + 3.024 \times 10^{-5} T^2$ cal/g with a standard deviation of ± 0.409 cal/g, and that for cubic UC_{1.93} as $H_T - H_{310^\circ K}^\circ = 67.91 - 9.113 \times 10^{-3} T + 2.629 \times 10^{-5} T^2$ cal/g with a standard deviation of ± 0.844 cal/g.³³ A transition temperature of 2043°K was observed with a heat of transformation of 11.0 cal/g.³⁴ The standard heat of formation, ΔH_{298}° , of UC_{1.91} was calculated as -20.2 kcal; the entropy, S_{298}° , as 16.86 eu; and the specific heat, C_{P298}° , as 14.57 cal/mole-°C. A standard free energy, ΔF_T° , of -15,820-8.2 T cal/mole was reported for the reaction $U(\beta) + 2 C(\text{gr}) = UC_2(s)$.²⁶

Plutonium Monocarbide

The presence of four carbide phases was observed in the plutonium-carbon system.³⁵ The zeta phase, Pu₃C₂, on heating to 575°C, decomposed to ϵ -Pu and PuC. Rosen et al. observed limited solubility of carbon in liquid plutonium, which resulted in lowering the melting temperature from $641 \pm 1^\circ$ to the eutectic at $637 \pm 1^\circ$ C.³⁶ The dicarbide is unstable below approximately 1750°C with respect to Pu₂C₃ and C.³⁵ Plutonium monocarbide and sesquicarbide are the primary stable phases of interest for nuclear fuel.

The structure of PuC is face-centered cubic, NaCl type. The compound is formed by a peritectic reaction at 1654°C between plutonium and Pu₂C₃. It is a carbon-deficient structure, having a range of homogeneity at approximately 570°C of from 43.5 at. % carbon at the (ϵ -Pu + PuC)/PuC boundary to 48 at. % carbon at the PuC/(PuC + Pu₂C₃) boundary.³⁶ Kruger reported that the composition containing 46.5 at. % carbon was the closest to a single phase microstructure.³⁷ The lattice parameter of arc-melted alloys varies from 4.9582 ± 0.0003 Å for carbon-deficient PuC to 4.9737 ± 0.0003 Å for excess-carbon-containing PuC.³⁵ The theoretical density is 13.62 g/cm³. The solubility of oxygen in PuC was studied by x-ray and micrographic analyses, delineating phases in the Pu-C-O system.³⁸

The heat of formation of PuC_{0.77} was determined by combustion calorimetry.¹¹ The estimates of the thermodynamic properties follow: $\Delta H_{298}^\circ = 3.7 \pm 3.1$ kcal/mole, $\Delta F_{298}^\circ = 2.5$ kcal/mole, $\Delta S_{298}^\circ = 4$ eu, and $S_{298}^\circ = 17$ eu. The heat capacity of PuC_{0.86} was reported as $C_p = 13.08 \pm 11.44 \times 10^{-4} T - 3.23 \times$

$10^{-5} T^{-2}$ cal/mole-°C between 425° and 1300°K.³⁹

The vapor-transpiration technique was used to measure the vapor pressure of PuC (47.3 at. % carbon) between 1500° and 1565°C by Palfreyman et al.⁴⁰ Although opinions differ regarding the composition of the vaporized species in similar experiments,^{41,42} he assumed the vaporizing species to be PuC and expressed the data by the relationship $\log_{10} p(\text{atm}) = 22.3 - (51,100)/T$.

The hardness of PuC (46.9 and 47.1 at. % carbon) was reported to be 860 ± 25 VHN (200 g load).³⁷ Kruger observed abrupt increases in rate of thermal expansion in performing dilation curves on cast plutonium carbide.³⁷ This anomalous behavior was attributed to retention and subsequent thermal rearrangement of the high temperature phase in the PuC structure.⁴³ The average linear thermal expansion coefficient between 20° and 900°C is $10.6 \times 10^{-6}/^\circ\text{C}$.

Leary et al. reported a thermal conductivity of 0.0318 cal/sec/cm-°C (0.133 W/cm-°C) at 400°C for PuC_{0.87}.¹⁷ The electrical resistivity was 268 $\mu\Omega$ cm at room temperature at the same composition.

Plutonium Sesquicarbide

The structure of Pu₂C₃ is body-centered cubic, space group $\bar{1}43d-T_d^6$. The lattice parameter is 8.129 ± 0.001 Å corresponding to a density of 12.70 g/cm³.⁴⁴ Pu₂C₃ has a homogeneity range (between 610° and 555°C) with the lower limit between PuC_{1.41-1.46} and the upper limit PuC_{1.50}.⁴⁵ The compound is formed by the peritectic reaction between liquid plutonium and PuC₂ at 2050°C.³⁵

Huber et al. measured the heat of formation of Pu₂C₃ by combustion calorimetry and reported the following thermochemical properties for the material: $\Delta H_{298}^\circ = 1.7$ kcal/mole, $\Delta F_{298}^\circ = 2.6$ kcal/mole, $\Delta S_{298}^\circ = -3$ eu, and $S_{298}^\circ = 25$ eu.¹¹ The free energy values at 1000° and 2000°K were -5.6 and -8.6 kcal/mole, respectively.

The hardness of plutonium sesquicarbide was reported⁴⁶ as being 900 kg/mm². Pallmer⁴⁷ determined the linear coefficient of thermal expansion between 20° and 780°C as $\alpha = 14.8 \times 10^{-6}/^\circ\text{C}$. No measurement was found for the thermal conductivity.

Mixed Uranium-Plutonium Monocarbide

The relatively poor thermal conductivity, low melting temperature, and high vapor pressure of plutonium in the plutonium-carbon system indicate the

desirability of modifying these properties by alloying additions. The addition of uranium to form mixed uranium-plutonium carbides is an obvious compositional modification. Plutonium monocarbide forms a complete series of solid solutions with uranium monocarbide.⁴⁸ There is a negative deviation from Vegard's law in the lattice parameters up to approximately 35 at. % plutonium. The microstructure of carbon-deficient compositions shows a second phase identified by x-ray diffraction as either a binary U-Pu alloy or the zeta phase (Pu_3C_2).⁴⁹ The mixed monocarbide dissolves relatively high concentrations of oxygen and/or nitrogen which may result in deviation from carbon stoichiometry as determined by the examination of the microstructure.⁵⁰

A study of the equilibria on (U,Pu)C containing excess carbon was conducted by Dalton.⁵¹ He reported that plutonium stabilized the sesquicarbide by raising the temperature at which the reaction $\text{U}_2\text{C}_3 \rightleftharpoons \text{UC} + \text{UC}_2$ occurred. Pu_2C_3 forms a complete series of solid solutions with U_2C_3 at temperatures below 1760°C.

In practice, the single phase, pure mixed monocarbide is extremely difficult to prepare. The microstructure of carbon-deficient ($\text{U}_{0.8}\text{Pu}_{0.2}$)C consists of the mixed monocarbide plus a binary U-Pu alloy. However, in compositions containing excess carbon, the microstructure consists of monocarbide plus higher carbides which may be either the sesquicarbide or dicarbide depending on the thermal history of the fuel.

The thermal expansion of ($\text{U}_{0.87}\text{Pu}_{0.13}$)C was determined on cast and annealed (800°C) specimens.⁵² The average coefficient of linear expansion between room temperature and 900°C is $11.1 \times 10^{-6}/^\circ\text{C}$. The result agreed with the value of $11.3 \times 10^{-6}/^\circ\text{C}$ determined on sintered ($\text{U}_{0.8}\text{Pu}_{0.2}$)C_{0.95} between 25° and 950°C.⁵³ Leary et al. observed that the thermal conductivity of UC decreased on the addition of PuC from 0.054 to 0.035 cal/sec-cm-°C (0.23 to 0.16 W/cm-°C) at approximately 30 mole % PuC.¹⁷ The thermal conductivity of ($\text{U}_{0.8}\text{Pu}_{0.2}$)C at 400°C is 0.042 cal/sec-cm-°C (0.18 W/cm-°C).

Volatilization of plutonium from U-Pu-C alloys is recognized to be a problem in fuel fabrication, particularly at high plutonium content. Dalton⁵¹ heated ($\text{U}_{0.5}\text{Pu}_{0.5}$)C containing 54.8 at. % carbon for 170 h in vacuum at 1800°C. The plutonium content

decreased in arc-melted material from 46.2 wt % to 42.6 wt %. In arc-melting experiments with ($\text{U}_{0.8}\text{Pu}_{0.2}$)C as a function of time the alloy remained molten. Burnham et al. reported a decrease in the Pu/U ratio by a factor of two after 8 minutes in the arc.⁵⁴ Although the vapor pressure measurements over ($\text{U}_{0.8}\text{Pu}_{0.2}$)C_{0.95} were not sufficiently precise for plotting, Stahl et al. stated that the mass effusion rates should not interfere with the use of the fuel for a high temperature, high power reactor.⁵⁵ Leary stated that no significant processing loss of plutonium occurs from ($\text{U}_{0.8}\text{Pu}_{0.2}$)C at below 1500°C.⁵⁶

The melting temperatures of mixed monocarbide compositions vary linearly from that of UC to that of PuC.⁵¹

PROCESSING REACTIONS

Outgassing of Consolidated Carbides

The outgassing of bulk uranium and plutonium carbides at up to 1000°C was studied by Evered et al.⁵⁷ Cast material, when stored under pure argon (< 20 ppm H₂O and < 50 ppm O₂) for periods up to 6 weeks, evolved a maximum of 50 $\mu\text{l g}^{-1}$, consisting primarily of hydrogen, admixed with small amounts of methane and N₂/CO, presumed to arise from reaction of the carbide with chemisorbed moisture. Larger quantities of gas were evolved from sintered material and/or carbides stored under less favorable conditions.

Reaction with Water and Steam

Consolidated uranium monocarbide is severely corroded at ambient temperatures in moist air and organic liquids containing moisture. The gaseous hydrolysis products from UC were identified as consisting primarily of methane, with some hydrogen and small quantities of higher hydrocarbons.⁵⁸ The residue consisted of hydrated UO₂.⁵⁹ Anselin et al.⁶⁰ observed the complete disintegration of UC, PuC, or ThC in boiling water in about 30 min. In contrast, mixed carbonitride compositions, ($\text{U}_{0.75}\text{Pu}_{0.25}$)C_{0.67}N_{0.33} and U(C_{0.67}N_{0.33}), were much more resistant, although plutonium appeared to enhance carbide reactivity in boiling water.

The oxidation of UC in steam at from 300° to 1000°C approximated a linear rate with a constant of (450 mg wt gain)/h (cm² geometric area).⁶¹ The reaction product consisted of a nonadherent oxide.

Reaction with Hydrogen

The reduction of UC and (U,Pu)C containing excess carbon to single-phase material by hydrogen was studied.⁶² Green-pressed pellets (7-mm o.d. x 4-mm tall) were sintered for 1 h at 1500°C under a hydrogen flow of 500 cm³/min, and the effluent was analyzed for methane. The rate of heating to sintering temperature was varied to obtain homogeneity in the metallographic structure. A residual high carbon core was absent from (U_{0.85}Pu_{0.15})C pellets having an initial carbon content of 5.0 wt % carbon heated at rates up to 500°C/h.

Reaction with Oxygen

The reaction of UC and (U,Pu)C at low partial pressures of oxygen is harmful to compositional control. Shupe⁶³ reported that (U,Pu)C gained weight at room temperature on exposure to argon containing 50 ppm oxygen, and that the oxygen and the water content of inert process boxes needed to be held to less than 5 ppm and 1 ppm, respectively, during the routine preparation of single-phase mixed carbide. An ignition temperature of 0°C was observed⁶⁴ for a ball-milled UC powder in dry air having a surface area of 18,000 cm²/g compared to 180°C for powder with a surface area of 4,800 cm²/g. The ignition products were U₃O₈ and CO₂. The presence of plutonium enhanced the reactivity of the powder. The low temperature oxidation of UC and (U,Pu)C powder results in liberation of carbon and the appearance of higher carbides in the microstructure of sintered pellets. Below 230°C, above which temperature formation of CO₂ occurs, the degree of oxidation (α) was related to the time (t) in seconds at a temperature (T°K) as follows:

$$5.9 \times 10^{10} \exp \left(- \frac{30,000}{RT} \right) = 3.925 - (1-\alpha)^{2/3} - 2.925 (1 + 0.342 \alpha)^{2/3}.$$

The expression applied for partial pressures of oxygen of 40 to 1000 ppm in argon at normal pressure. It was observed that the low temperature (140°-230°C) reaction products were UO_{2.20} and free carbon, which appeared to be controlled by the diffusion of oxygen through the oxide layer.

Reaction with Nitrogen

UC and (U,Pu)C react with nitrogen in nitrogen-contaminated atmospheres to form corresponding

nitrides and free carbon.⁶⁵ The extent of reaction depends on the specific conditions, such as the partial pressure of nitrogen, the temperature, and the surface area.

FUEL-SYSTEM REACTION

The reaction between carbide fuel and cladding material may often be predicted from free energy considerations. The thermodynamic properties of mixed fuel carbides are not precisely known, but the data indicate that plutonium carbide is less stable than the corresponding uranium compound. The effect of imposing a reactor environment on the fuel system may increase chemical reactivity and promote mechanisms in which diffusion is rate-controlling. Unfortunately, there are few instances of experimental work reporting tests of either pure UC or pure (U,Pu)C with potential cladding material. The impurity content of fuel is related to the fabrication method and may be up to 5000 ppm residual oxygen and nitrogen. Although there is an indication that the chemical reactivity of the fuel may decrease with oxygen (and nitrogen) addition, a significant deterioration of uranium oxycarbide was observed⁶⁶ under neutron irradiation at approximately 800°C. This led to the formation of free metal.

Katz studied the high temperature reactions between UC, UC₂, and UN and various metals, including Type 304 stainless steel.⁶⁷ Applying free-energy criteria to the reaction with UC at 1000°C, molybdenum and vanadium were postulated not to react while a reaction was predicted with aluminum, chromium, iron, niobium, nickel, thorium, titanium, and zirconium. Compatibility experiments were conducted showing that all these metals, including Type 304 stainless steel, reacted at 1000°C with both UC and UC₂. The reaction products were identified, and penetration curves were presented for the experimental work.

Thermochemical data for the carbides of molybdenum, niobium, tantalum, titanium, tungsten, uranium, hafnium, and zirconium were reviewed, and the free energy of formation was calculated as a function of temperature by Alexander et al.⁶⁸ On the basis of the calculations, only molybdenum and tungsten were considered to be compatible with uranium monocarbide. Uranium sesquicarbide and dicarbide were predicted to react with these metals between

1000° and 2000°C.

The compatibility of uranium carbide/sodium/Type 304 stainless steel system was investigated by in-pile and out-of-pile tests.⁶⁹ Above 1000°F, uranium dicarbide was attacked by the molten sodium. Carbon was transferred by the sodium to the stainless steel cladding, which acted as a carbon sink. However, carbon-deficient uranium carbide (4.5 to 4.8 wt % carbon) was compatible with Type 304 stainless steel at a fuel surface temperature of 1250°F, center temperature of 2000°F, and a burnup of 25,000 MWD/T.

Subsequent tests by Hahn dealt specifically with fission-gas retention and dimensional stability of uranium carbide fuels, ranging in carbon content from 4.45 to 4.76 wt % at a burnup of 19,200 MWD/T.⁷⁰ Uranium carbide was decarburized in a UC/Na/Type 304 stainless steel system, provided that (a) the fuel temperature was greater than 1300°C, (b) there was insufficient free uranium metal in the metallographic structure to combine with all the metastable carbon at temperature, and (c) a carbon sink existed, such as chromium in a stainless steel container. Radial redistribution of the free uranium phase occurred in varying degrees in the irradiated specimens. In the low carbon specimen (4.45 wt % carbon), the reaction products on the Type 304 stainless steel surfaces were identified as $U_6Fe-UFe_2$, $U_6Fe-UFe_2-UC$, and/or U_6Fe-UC .

The limitations in compatibility of uranium carbides with Type 304 stainless steel, Nb-1 % Zr and a zirconium base alloy were reported by Elkins.⁷¹ Below 1800°F, in either the presence or absence of a sodium bond, neither uranium nor carbon reaction products were observed on the cladding surface. Although no significant contact reaction was observed in the absence of sodium below 1600°F, under practical conditions of intimate contact after long-term irradiations, liquid- or solid-state reactions would be expected between the metallic uranium phase in carbon-deficient UC and the Type 304 stainless steel cladding material. With stainless steel containers, the fuel surface should be restricted to temperatures well below the lowest iron-uranium eutectic temperature of 1340°F.

In 6000-h tests, UC containing excess carbon without a liquid-metal bond was compatible with the three cladding materials to the extent that the ob-

served reactions at 1600°F were localized and limited in depth. However, in a sodium environment, this fuel was not compatible with Type 304 stainless steel. Tests in Nb-1 % Zr and zirconium-base alloy containing carbon-saturated sodium showed satisfactory compatibility below 1600°F. At higher temperatures, 1800° and 2000°F, carburization was excessive with evidence of accelerated effects caused by spalling of the carbide layer.

The reaction of Type 316L stainless steel with mixed uranium-plutonium carbides was studied.⁷² However, impure specimens (20% sesquicarbide and 0.4% oxygen) were used. A maximum carbon penetration through a solid/solid interface of 2.5×10^{-3} in. was observed in the stainless steel. This degree of penetration was assumed not to degrade the properties of the clad. The carbon transfer was independent of the sesquicarbide and oxygen content of the pellet, of the treatment of the stainless steel, of the temperature (650° to 800°C), and of the period (700 to 8,400 h) of test. The source of carbon was suspected to be the low-temperature oxidation of the pellet surface prior to assembly of the test specimens.

A sodium bond resulted in a catastrophic carburization of the stainless steel. Using pellets containing 4.53 wt % carbon, 0.37 wt % oxygen and 0.05 wt % nitrogen at 650°C and test periods of from 700 to 4,200 h, a maximum depth of carbon penetration of 5×10^{-3} in. was observed. French et al. concluded that the rate of carburization was less for mixed uranium-plutonium carbides than for uranium carbide for equivalent carbon analysis owing to the presence of larger quantities of the sesquicarbide phase in the latter material.⁷³ The structure of the mixed carbides ($U_{0.85}Pu_{0.15}$)C was depleted in the dicarbide phase, while no depletion of the sesquicarbide phase occurred, during up to 3 months of tests at 700° and 800°C.

In addition to the compatibility study of sodium-bonded mixed carbides and Type 316L stainless steel, tests were conducted also by the latter investigator on the reaction of mixed carbides in 50% nickel cermet. At 600° within 14 days a reaction was observed which yielded Pu-Ni compounds, primarily $PuNi_5$. At higher temperature (800°-900°C), the reaction products (consisting of (U,Pu)-Ni compounds and free carbon) were observed in shorter periods (1 week).

A proposed method for improving the compatibility of sodium-bonded uranium-plutonium carbide is currently under investigation at Westinghouse Electric Corporation. The structure of mixed carbide is modified (buffered) by the addition of 120% excess of either iron or Cr_{23}C_6 to combine with the metallic phase during sintering to form high melting phases (UFe_2 and Cr, for example).⁷⁴ Preliminary tests conducted on sodium-bonded, Cr_{23}C_6 -stabilized pellets indicated no extensive interaction with either Type 316L or 304L stainless steels or high strength vanadium alloys (1000- and 1440-h test periods).

IRRADIATION BEHAVIOR

Essentially all the irradiation data up to July, 1966, on uranium carbide were reviewed by Frank et al.⁷⁵ The description of the specimens is shown in Table I, and the irradiation behavior is recorded in Table II. The data indicate that fuel swelling is primarily a function of the carbon content, the fuel burnup, and the temperature of fuel operation. Fuel swelling is a linear function of temperature up to 1800°F, above which it becomes exponential. Below 1800°F, fuel swelling is a function of burnup, varying from 1.5 to 3.75% per 10,000 MWD/T. A minimum swelling occurs on approach towards

Table I
Description of Irradiated Uranium Carbide Specimens
(Reference 75)

Capsule No.	Reference	Composition (wt % C)	Fabrication Method	Structure	Density (% T.D.)	Bond	Clad
BMI-23-5	BMI-1452 NAA-SR-7696	4.6	Cast	MC + M	99.0	NaK	Type 304 SS
BMI-23-6	BMI-1452 NAA-SR-7696	4.8	Cast	MC	99.9	NaK	Type 304 SS
AI-3-1	NAA-SR-7696	4.4 to 4.7	Cast				
AI-3-12 (1)	NAA-SR-TDR-11334	4.4	Cast	MC + M	99.0	Na	Type 304 SS
(2)		4.68	"	MC + M	99.0	Na	Type 304 SS
(3)		4.44	"	MC + M	99.0	Na	Type 304 SS
(4)		4.60	"	MC + M	99.0	Na	Type 304 SS
(5)		4.47	"	MC + M	99.0	Na	Type 304 SS
(6)		4.62	"	MC + M	99.0	Na	Type 304 SS
NAA-81-3 (1)	NAA-SR-11140	4.6	Cast	MC + M	99.0	Na	Type 304 SS
(2)		4.8	"	MC	99.0	Na	Type 304 SS
(3)		4.8	"	MC	99.0	Na	Type 304 SS
(4)		4.9	"	MC + MC_2	99.0	Na	Type 304 SS
(5)		4.9	"	MC + MC_2	99.0	Na	Type 304 SS
(6)		4.6	"	MC + M	99.0	Na	Type 304 SS
(7)		4.5	"	MC + M	99.0	Na	Type 304 SS
(8)		4.5	"	MC + M	99.0	Na	Type 304 SS
NAA-81-4 (1)	NAA-SR-TDR-11182	4.68	Cast	MC + M	99.0	Na	Type 304 SS
(2)		4.46	"	MC + M	99.0	Na	Type 304 SS
(3)		4.47	"	MC + M	99.0	Na	Type 304 SS
(4)		4.45	"	MC + M	99.0	Na	Type 304 SS
(5)		4.61	"	MC + M	99.0	Na	Type 304 SS
(6)		4.70	"	MC + M	99.0	Na	Type 304 SS
NRX-85-1	NAA-SR-11134	4.45 to 4.77	Cast	MC + M	98.0	Na	Type 304 SS
AI-3-11 (1)	NAA-SR-11440	4.53	Cast	MC + M	99.0	Na	Type 304 SS
(2)		4.55	"	MC + M	99.0	Na	Type 304 SS
(3)		4.53	"	MC + M	99.0	Na	Type 304 SS
(4)		4.50	"	MC + M	99.0	Na	Type 304 SS
(5)		4.53	"	MC + M	99.0	Na	Type 304 SS
(6)		4.59	"	MC + M	99.0	Na	Type 304 SS
(7)		4.59	"	MC + M	99.0	Na	Type 304 SS

Table I (Continued)

Capsule No.	Reference	Composition (wt % C)	Fabrication Method	Structure	Density (% T.D.)	Bond	Clad
UNC-1-1 (1)	UNC-5080	4.45	Cast	MC + M	99.1	Na	Nb-1 wt % Zr
(2)		4.31	Sintered	MC + M	97.6	Na	Nb-1 wt % Zr
(3)		4.70	Cast	MC + M	99.3	Na	Nb-1 wt % Zr
(4)		4.86	Sintered	MC + MC ₂	93.5	Na	Nb-1 wt % Zr
(5)		5.21	Cast	MC + MC ₂	98.9	Na	Nb-1 wt % Zr
UNC-1-2 (1)	UNC-5080	4.45	Cast	MC + M	99.2	Na	Nb-1 wt % Zr
(2)		4.31	Sintered	MC + M	97.6	Na	Nb-1 wt % Zr
(3)		4.71	Cast	MC + M	99.1	Na	Nb-1 wt % Zr
(4)		4.86	Sintered	MC + MC ₂	93.6	Na	Nb-1 wt % Zr
(5)		5.21	Cast	MC + MC ₂	99.0	Na	Nb-1 wt % Zr
UNC-1-3 (1)	UNC-5080	4.45	Cast	MC + M	99.1	He	Nb-1 wt % Zr
(2)		4.31	Sintered	MC + M	97.6	He	Nb-1 wt % Zr
(3)		4.71	Cast	MC + M	98.8	He	Nb-1 wt % Zr
(4)		4.86	Sintered	MC + MC ₂	93.7	He	Nb-1 wt % Zr
(5)		5.21	Cast	MC + MC ₂	99.0	He	Nb-1 wt % Zr

Table II
Irradiation Performance of Uranium Carbide
(Specimens Described in Table I)
(Reference 75)

Capsule No.	Burnup (MWD/T)	Fuel Temp. (°F)		Fission Gas Release (% of Theoretical)	Fuel Diameter Change (%/10000 MWD/T)	Fuel Density Change	Comments
		Surface	Center				
BMI-23-5	8600	882	1326	0.04	0.92	2.75	Intact. No cladding-dimension change.
BMI-23-6	6500	780	1300	0.04	0.74	1.77	Intact. No cladding-dimension change.
AI-3-1	6500	1250	1800	4.6			Intact. Maximum temperature (1600°F surface) experienced at end of run. Postirradiation analyses of clad 0.16 wt % C.
AI-3-12 (1)	18000	1000	1640	Lost	--	--	Intact. No cladding-dimension change. Postirradiation analyses of clad from 0.12 to 0.28 wt % C.
	(2)	19000	1050		--	--	
	(3)	20000	1100		1.45	3.6	
	(4)	19800	1080		0.61	2.27	
	(5)	19000	1140		0.37	2.64	
	(6)	17000	960		--	2.0	
NAA-81-3 (1)	7200	660	1040	0.25	--	2.36	Intact. No cladding-dimension change.
	(2)	7800	780		--	2.30	
	(3)	8800	880		--	2.50	
	(4)	9300	900		Negligible	1.83	
	(5)	10400	975		--	2.02	
	(6)	9400	920		--	1.92	
	(7)	8200	880		--	3.29	
	(8)	7000	750		--	2.58	
NAA-81-4 (1)	19200	--	--	3.3	3.44	7.6	Failed. Peak temperature believed to be 3000-4000°F. Failure due to pinhole leak at end plug weld.
	(2)	21000	--		4.0	8.8	
	(3)	21600	--		--	6.5	
	(4)	21600	--		--	3.75	
	(5)	21000	900		1.19	2.52	
	(6)	19200	850		0.68	1.47	
NRX-85-1	18400	1580	2510	18.7	--	--	Failed.

Table II (Continued)

Capsule No.	Burnup (MWD/T)	Fuel Temp. (°F)		Fission Gas Release (% of Theoretical)	Fuel Diameter Change (%/10000 MWD/T)	Fuel Density Change (MWD/T)	Comments
		Surface	Center				
AI-3-11	(1)	19500	760	1225	--	2.92	Intact. No cladding-dimension change. Postirradiation analyses of clad from 0.08 to 0.20 wt % C.
	(2)	26000	950	1550	--	2.42	
	(3)	30000	1020	1650	--	2.86	
	(4)	29000	1020	1700	--	2.76	
	(5)	28000	1040	1800	--	2.72	
	(6)	26300	1040	1700	--	2.48	
	(7)	22500	980	1600	--	2.31	
UNC-1-1	(1)	37900	1300	2040	5	2.7	All intact. No cladding-dimension change. Temperature may have been higher than estimated as there was no centerline thermocouple. Data comparable within themselves.
	(2)	40100	1340	2050	36	2.6	
	(3)	34800	1300	1880	31	3.6	
	(4)	34500	1325	1900	14	1.8	
	(5)	36000	1270	1830	5	1.6	
UNC-1-2	(1)	~22,900	--	--	30	4.9	All intact. No cladding-dimension change. Temperature may have been higher than estimated as there was no centerline thermocouple. Data comparable within themselves.
	(2)		1150	1800	45	5.0	
	(3)		1330	2050	7	--	
	(4)		1330	2050	14	--	
	(5)		1340	2075	4	--	
UNC-1-3	(1)	~14,300	1600	2210	43	0.5	All intact. No cladding-dimension change. Temperature may have been higher than estimated as there was no centerline thermocouple. Data comparable within themselves.
	(2)		1610	2150	50	0.7	
	(3)		1510	2000	5	1.8	
	(4)		1580	2100	16	0.8	
	(5)		1610	2150	1	2.0	

stoichiometric carbon compositions. Brinkman calculated a minimum swelling of 1.8% per 10,000 MWD/T due to the accumulation of fission products in the uranium carbide lattice.⁷⁶

The irradiation behavior of vibratorily compacted uranium-20 wt % plutonium carbide is under study at Argonne National Laboratory.⁷⁷ Irradiation-test results have been reported on fuel prepared from mechanically blended UC and PuC powders, powders prepared from arc-melted compositions, and pellets prepared by sintering the powder. The data^{77,78} are summarized in Tables III and IV.

The specimens shown in Table III were 70 mm overall with a 50-mm-long fuel section (86% smear density) and a small, 13-mm helium-containing gas plenum above the fuel. The fuel diameters were 3.96 and 6.53 mm. The jackets were either Nb-1 wt % Zr or Type 304 stainless steel, 0.23- and 0.30-mm thick. The carbon content of the unenriched uranium carbide (20/40 and -325 mesh) was 4.8 wt %; that of the plutonium carbide (120/240 mesh) ranged from 3.4 to 6.4 wt %. Free plutonium metal was present in the low carbon material, and plutonium sesquicarbide was

present in the high carbon batches.

The specimens operated at clad-surface temperatures from 570 to 650°C at linear heat ratings of approximately 6 kW/ft. The dimensional changes on the Nb-1 wt % Zr capsules were negligible, while the stainless-steel capsules had diameter increases up to 4.7% and volume increases up to 1.3%. Fission-gas release was related to the carbon content of the PuC with the low-carbon samples releasing up to 40% compared to approximately 1% of theoretical for the high-carbon material.

The PuC was reported to have reacted slightly with the stainless steel and occasionally with the Nb-1 wt % Zr containers.⁷⁹ Two capsules containing PuC fractured during irradiation. The cause of rupture in specimen F-8 was presumed to be high internal gas pressure (4.5 kg/mm²), while an examination of specimen F-6 indicated plutonium melting.

An electron-microscopic study of the structural changes in specimens C-72 and C-70 was reported by Yoon et al.⁸⁰ An average burnup of 6.8 at. % corresponded to burnups of 34 at. % (C-70) and 29 at. % (C-72) of the plutonium atoms present. The greater

Table III
Irradiation Tests on Blended Vipac PuC and UC-PuC Fuel Powders
(Reference 77)

Specimen No.	Fuel	Jacket Material	Carbon in PuC (wt %)	Burnup		Linear Heat Rating (W/cm)		Jacket Surface Temp. (°C)		Fuel Center Temp. (°C) (a)		Fuel Swelling Percent Change			Gas Release, % Theoretical
				at. %	Fiss/cc (Units of 10^{20})	Av.	Max.	Av.	Max.	Av.	Max.	Diam	Length	Volume	
F-7	PuC	304 SS	4.24	1.95	5.1	285	310	450	495	1000	1095	1.0	0.1	1.1	35.2
F-6	PuC	304 SS	6.22	5.43	13.1	390	505	550	625	1305	1610	0.8(b)	Specimen Melted		
F-8	PuC	304 SS	4.24	6.0(c)	16.0	195	210	605	675	980	1080	14.3	2.0	Jacket Ruptured	
C-44	PuC	Nb-1Zr	3.40	2.18	5.3	295	335	515	625	1070	1255	0.5	Neg.	0.3	45.9
C-43	PuC	Nb-1Zr	4.24	5.3(c)	14.0	165	170	510	550	820	870	0.1	Neg.	0.3	31.0
F-18	UC-20PuC	304 SS	6.42	1.35	3.4	190	200	455	505	820	890	0.3	Neg.	0.6	8.8
F-21	UC-20PuC	304 SS	3.40	1.40	3.5	195	220	395	430	770	820	0.5	Neg.	0.2	25.7
F-19	UC-20PuC	304 SS	6.22	5.8(c)	15.0	170	180	525	570	855	915	2.8	0.3	3.5	<1(d)
F-20	UC-20PuC	304 SS	4.24	6.8(c)	18.0	195	205	600	645	975	1040	4.7	1.3	7.3	40.0
C-69	UC-20PuC	Nb-1Zr	6.42	1.21	3.1	170	185	370	405	690	750	0.5	Neg.	0.3	5.7
C-74	UC-20PuC	Nb-1Zr	3.40	1.57	4.1	225	260	400	455	820	945	0.3	Neg.	0.5	21.5
C-72	UC-20PuC	Nb-1Zr	6.22	6.8(c)	18.0	190	195	575	635	925	1000	0.2	Neg.	0.4	<1(d)
C-70	UC-20PuC	Nb-1Zr	4.24	6.8(c)	18.0	195	200	600	650	965	1025	0.4	Neg.	0.5	20.0

(a) Calculated assuming a fuel-jacket interface conductance of $0.136 \text{ cal/sec-cm}^2\text{-}^\circ\text{C}$ ($1000 \text{ Btu/h-ft}^2\text{-}^\circ\text{F}$).

(b) Measured on remaining pieces.

(c) Burnup estimated from heat-generation rates.

(d) Based only on xenon release.

Table IV
Irradiation Behavior of $(U_{0.8}Pu_{0.2})C$ and UC-20 wt % PuC Fuel Pins
(Reference 78)

Specimen No.	Fuel	Form	Effective Density (%)	Jacket Material	Burnup (at. %)	Linear Heat Rating (kW/ft)	Jacket Surface Temp (°C)	Change in Jacket Dimension	Gas Release, % Theoretical
SMP-1	$(U_{0.8}Pu_{0.2})C$	Pressed and Sintered	81.4	316 SS	2.1	18.9	600	Insignificant	4.1
VMV-1	$(U_{0.8}Pu_{0.2})C$	Vibratorily Compacted	85.9	Vanadium	3.02	29.5	680	$\Delta d=0.83\%$	8.6
SMV-1	UC-20 wt % PuC	Vibratorily Compacted	80.0	316 SS	2.59	25.3	675	Insignificant	13.4

fission-gas release in the $PuC_{0.89}$ was attributed to the fact that fission-gas bubbles in the $PuC_{0.89}$ were generally larger and more interconnected than in the $PuC_{1.33}$. The maxima of the size distribution of fission-gas bubbles were approximately 3 and 12 microns diameter, respectively. Spherical, second-phase particles of various sizes presumed to be associated with a high concentration of solid fission products were observed in both high burnup $PuC_{1.33}$ and $PuC_{0.89}$. There were more of the second-phase particles in the $PuC_{1.33}$ than in the $PuC_{0.89}$. This difference in the distribution of the second-phase particles may account for the differences in the size and distribution of the fission-gas bubbles. UC_2 platelets, originally in the Widmanstätten structure in the UC_{1+x} sample disappeared from the UC matrix in areas within the range of fission recoil from adjoining PuC. This indicated a recoil effect, or that some solid-solution formation had occurred across the UC_2 -PuC phase boundaries. However, the depletion of the UC_2 platelets at the free surface of the UC_{1+x} particles, and also adjacent to internal cracks, indicated that at least part of the depletion may have occurred by the oxygen-carbon reaction.

Recent irradiation data on specimens prepared by Argonne National Laboratory are recorded in Table IV. The specimens were helium-bonded to their respective claddings. No information was reported on a mixed-carbide/clad reaction. However, the data indicate a superiority of fission-gas retention for pressed and sintered pellets in comparison to either pre-alloyed, vibratorily compacted, or blended mixed-carbide compositions.

Irradiation tests reported by United Nuclear Corporation up to 1965 on sintered mixed uranium-plutonium carbides are summarized in Table V.^{81,82} The pellets (with or without 0.1% added nickel) were prepared by sintering the reaction product of carbon-reduced mixed oxides to 92% and 96% of the theoretical density.⁸³ After the fuel had been irradiated up to an average of 37.6×10^{20} fissions/cm³ (114,000 MWD/T) and 150 to 330 kW/kg (U + Pu), the microstructure was stable, and the diameter increase was 0.6% per at. % burnup. The compatibility of the fuel with the clad was excellent up to a maximum of 870°C. Referring to Table V, and the performance of six capsules, failure on one resulted from fuel melting, while the failure on four was attributed to clad embrittlement. Although fission-gas retention was satisfactory up to a burnup of 35,000 MWD/T, about 30% of the fission-gas was released at a burnup of 114,000 MWD/T.

Recent fast-reactor irradiation tests performed by United Nuclear Corporation was reported in Table VI.⁸⁴ The irradiation was conducted in EBR-II on three 14-in. fueled capsules made from V-20Ti, Type 316 stainless steel, and vanadium metal. Post-irradiation examination showed a surface reaction (described as very slight) with V-20Ti, which also showed a crack in the closure weld. There was no interaction between the fuel pellets and either the Type 316 stainless steel or the vanadium capsule material. These tests showed a maximum fuel-density decrease of 4.6% and a maximum fission-gas release of 0.5% for a burnup of 8.7×10^{20} fissions/cm³ (25,000 MWD/T).

There are two series of irradiation tests on

Table V
Irradiation Tests on Sintered Mixed Uranium-Plutonium Carbide
(Reference 81)

Specimen No.	Composition (a)	Clad (a)	MWD, T	Fission Gas Release (%)	Clad Diameter Increase (%/at.%)	Fuel Diameter Increase (%/at.%)	Fuel Density Decrease (%/at.%)	Heat Flux (w/cm ²)		Specimen Temp (°C) Average		Hours of Operation	Condition of Clad
								Av.	Max.	Clad Surface	Fuel Center		
UNC-61T	(U _{0.8} Pu _{0.2})C + 0.1 Ni	Nb	21,700	0.37	--	0.6	1.0	165	210	625	1030	3267	Intact
61B	(U _{0.8} Pu _{0.2})C	Nb	24,200	0.25	--	0.5	2.4	165	205	615	1015	3267	Intact
UNC-62T	(U _{0.8} Pu _{0.2})C + 0.1 Ni	Nb	31,300	1.45	--	--	0.4	155	210	640	1005	4879	Intact
62B	(U _{0.8} Pu _{0.2})C	Nb	35,100	0.94	--	--	--	155	190	580	935	4879	Intact
UNC-63T	(U _{0.8} Pu _{0.2})C + 0.1 Ni	Nb	112,700	27.9(b)	0.3	0.6	1.2	204	257	555	925	9198	Intact
63B	(U _{0.8} Pu _{0.2})C	Nb	114,000	<1 (sample error)	0.2	--	1.1	154	227	460	790	9198	Intact
UNC-64T	(U _{0.8} Pu _{0.2})C	316 SS	23,400	0.75	--	--	--	180	220	655	1125	2189	Intact
64B	(U _{0.8} Pu _{0.2})C + 0.1 Ni	316 SS	22,800	12.7	--	0.6	1.4	185	240	690	1175	2189	Localized failure
UNC-65T	(U _{0.8} Pu _{0.2})C + 0.1 Ni	316 SS	37,700	Lost	0.8	--	1.1	220	355	445	1025	3723	Longitudinal brittle cracks
65B	(U _{0.8} Pu _{0.2})C	316 SS	49,300		0.6	--	--	250	325	473	1090	3723	
UNC-66T	(U _{0.8} Pu _{0.2})C + 0.1 Ni	316 SS	101,000		0.6	--	1.7	183	282	575	1030	8729	
66B	(U _{0.8} Pu _{0.2})C	316 SS	100,500	30.8(b)	0.5	--	1.6	184	275	455	885	8729	

(a) All specimens had a hyperstoichiometric structure and were helium-contact-bonced.

(b) Preliminary. Clad failure was attributed to a combination of causes:

(a) transmutation of ¹⁰B and subsequent agglomeration of helium at the high operating temperature, (b) the γ,n reaction from Fe, Ni, Cr, and N, and (c) the combination of thermal and mechanical stresses on the clad.

Table VI
Recent United Nuclear Corporation Irradiation Tests
(Reference 84)

	Rod 78	Rod 79	Rod 80
Rod Power (W/cm)	920	625	920
Clad	V-20 wt % Ti	Type 316	Vanadium
Fuel	(U _{0.8} Pu _{0.2})C	(U _{0.8} Pu _{0.2})C	(U _{0.8} Pu _{0.2})C
Smear Density (% T.D.)	94	93	93
Average Burnup ^(a) (fiss/cm ³)	8.7 x 10 ²⁰	5.9 x 10 ²⁰	8.7 x 10 ²⁰
(MWD/T)	(25,000)	(17,000)	(25,000)
Clad Appearance	Excellent	Excellent	Excellent
Clad Integrity	Intact except one thin crack near weld	Intact	Intact
Clad o.d. Increase (av. %)	0.4	0.4	0.7
Fission Gas Release ^(b) (% of theoretical)	0.20	0.45	0.50
Condition of Pellets	Surface reaction with most pellets recovered whole	Surface excellent, all pellets cracked	Surface excellent, all pellets cracked
Pellet Diameter Increase (av. %)	2.5	---	---
Fuel Density Change by Immersion (av. %(-))	4.4	3.4	4.6
Estimated Max. Fuel Temper- ature at Startup (°C)	1625	1530	1760
Estimated Clad Surface Temperature (°C)	620	540	620
Fuel o.d. (in.)	0.251	0.252	0.251
Fuel-Clad Gap, Cold, at Startup (in.)	0.005	0.007	0.006
Clad Thickness (in.)	0.021	0.023	0.022

(a) Estimated.

(b) Based on estimated burnup.

the performance of uranium-plutonium carbides reported by Bradbury et al.^{85,86} In the fast-reactor experiment, recorded in Tables VII and VIII, the fuel was clad in 0.030-in.-thick Nimonic 90 alloy cans with a 0.006-in.-thick co-drawn tantalum liner. The radial clearance between fuel and can was about 0.0003 in., which was filled with reactor-grade helium. Heat was conducted from the specimen cans through a close-fitting, steel-matrix block fitted to the outer capsule sheath. The capsules were irradiated in the Dounreay Fast Reactor. In these experiments, only two cans out of the eleven survived irradiation without failure. Serious compatibility problems were encountered with specimens contain-

ing free metal.

The specimens for the thermal-reactor experiment, shown in Table IX, were canned in Type 316L stainless steel, 0.020-in. wall thickness, with an average helium gap between fuel and can of < 0.001 in. Specimen temperature was controlled by a 3-kW heater in an aluminum heater block. The assembly was encased in a stainless-steel capsule. Twelve specimens, 1-in. long and 0.1-in. diameter, were irradiated in the DiDo reactor at Harwell in three capsules, each containing four specimens. Postirradiation examination was not complete at the time of reporting; however, radiography indicated that all 12 specimen containers were intact.

Table VII

Description of Uranium-Plutonium Carbide Specimens in the Fast Reactor Experiment
(Reference 85)

Can No.	Fuel Type	Specimen Size		Composition (wt %)					Equivalent Carbon (wt %)
		Length (in.)	Diam (in.)	C	O	N	Pu	U	
1	Sintered	1.024	0.32	4.76	0.24	0.04	13.0	82.0	4.97
2	Sintered	1.018	0.29	5.32	0.07	0.02	12.6	81.9	5.38
3	Sintered	1.015	0.27	4.73	0.09	0.07	13.2	81.9	4.86
4	Sintered	1.013	0.26	5.32	0.07	0.02	12.6	81.9	5.38
5	Arc Cast	1.068	0.25	4.76	--	--	--	--	--
6	Sintered	1.024	0.25	4.76	0.24	0.04	13.0	82.0	4.97
7	Powder	1.023	0.25	5.32	0.07	0.02	12.6	82.0	5.39
8	Powder	1.016	0.26	5.32	0.07	0.02	12.6	82.0	5.39
9	Arc Cast	1.017	0.27	4.75	--	--	14.2	--	--
10	Sintered	1.022	0.29	4.00	0.07	0.10	14.3	81.5	4.14
11	Arc Cast	1.016	0.32	4.80	--	--	--	--	--

Table VIII

Irradiation of Uranium-Plutonium Carbide Specimens in the
Fast Reactor Experiment
(Reference 85)

Can No.	Microstructure	Bulk Density	Theoretical Density (g/cm ³)	Specimen Rating (W/g)	Estimated Surface Temp (°C)	Estimated Center Temp (°C)	Estimated Burnup (% heavy atoms)	Estimated Fission Gas Release (%)
1	MC + M ₂ C ₃	12.9	13.4	144	1040	1610	3.95	--
2	MC + M ₂ C ₃	12.8	13.3	162	940	1440	4.45	--
3	MC + M ₂ C ₃	13.0	13.4	180	950	1460	4.95	--
4	MC + M ₂ C ₃	12.6	13.3	190	930	1430	5.20	--
5	MC + M	--	--	199	940	1420	5.40	--
6	MC + M ₂ C ₃	12.9	13.4	201	950	1430	5.50	--
7	--	60% T.D.	13.3	199	800	1270	5.40	0.2 - 0.5
8	--	60% T.D.	13.3	190	800	1250	5.20	1.25 - 2.5
9	MC + M	13.4	13.6	180	930	1440	4.95	--
10	MC + M	13.5	14.2	162	990	1530	4.45	--
11	MC + M	13.5	13.6	144	1020	1610	3.95	--

Table IX
Irradiation of Uranium-Plutonium Carbide in the
Thermal Reactor Experiment
(Reference 86)

Can No.	Sheath No.	Fuel Type	Equivalent Carbon Content	Micro-structure	Bulk Density (g/cm ³)	Specimen Rating	Estimated Specimen Surface Temp (°C)	Estimated Specimen Center Temp (°C)	Estimated Burnup (% heavy atoms)	Gas Release (%)
2	4A	Sintered	4.72	MC + trace of free metal	13.2	283	625	825	5.4	0.96
	3C	Powder	4.65	MC in metal matrix	~ 60% T.D.	400			7.4	6.2
	3B	Powder	4.40	MC in metal matrix	~ 60% T.D.	430			8.1	18.8
	3A	Sintered	4.72	MC + trace of free metal	13.2	358			6.7	2.12
3	2D	Arc Cast	--	MC + free metal	--	472	650	850	8.8	2.02
	2C	Arc Cast	--	MC + free metal	--					0.64
	2B	Arc Cast	--	MC + M ₂ C ₃	--					0.70
	2A	Sintered	4.72	MC + trace of free metal	13.2					0.56
4	1D	Sintered	4.45	MC + free metal	13.3	568	650	850	10.6	2.5
	1C	Sintered	4.95	MC + M ₂ C ₃	13.1					0.23
	1B	Sintered	4.72	MC + trace of free metal	13.2					0.32
	1A	Sintered	4.72	MC + trace of free metal	13.2					0.19

An irradiation program is in progress on (U,Pu)C, (U,Pu)(CN), and (U,Pu)N at Fontenay-aux-Roses.⁸⁷ The pellets are clad in Type 316L stainless steel, and the diametral clearance is filled with helium. A preliminary postirradiation examination on three cans after a burnup of 19,200 MWD/T in a thermal flux indicated a clad failure in all specimens which, in two of the cans, was associated with a drawing defect in the stainless steel. The maximum carbide swelling was 4% (fuel surface temperature was estimated at 1000°C).

CONCLUSIONS AND RECOMMENDATIONS

The objective of the current Los Alamos Scientific Laboratory carbide program is the development

of single-phase, mixed uranium-20 at. % plutonium carbide, sodium-bonded to Type 316L stainless steel as a clad-fuel configuration in the liquid metal fast breeder reactor system. Since stainless steel decreases in strength above 700°C, high strength vanadium alloy has been selected as the alternate cladding material.

This review indicates that considerable information has been generated by research on carbide fuels. However, much of the information is not statistically correlated toward the confident selection of an optimum fuel-clad configuration for fast reactor application. Emphasis has been placed on performance of carbide fuels in a thermal flux as a

function of burnup. The tests have been conducted at relatively high temperatures, using a number of experimental variables, such as cladding material, carbon stoichiometry, impurity content, medium for heat transfer, and rate of heat generation. Although the majority of irradiation tests have been conducted on uranium carbides, the results shown in this survey do not indicate that the behavior of mixed uranium-plutonium carbide is different. However, it should be emphasized that the behavior of single-phase, mixed-carbide fuel has not been evaluated in a fast reactor environment.

The irradiation tests indicate that the minimum decrease in fuel density occurs as the fuel approaches the stoichiometric composition. In tests conducted by the United Nuclear Corporation with uranium carbide containing free uranium (4.4 wt % carbon), a decrease in density of 8.8% per 10,000 MWD/T was measured compared to about 3.5% per 10,000 MWD/T in stoichiometric material (approximately 4.8 wt % carbon) and that containing excess carbon (approximately 5.2 wt % carbon), respectively. A minimum swelling of 1.8% per 10,000 MWD/T was calculated to result from fission product accumulations. When the uranium carbide is deficient in carbon, fission gas diffuses to the free uranium phase segregated at the grain boundaries, resulting in excessive swelling of the fuel.

In addition, free uranium or a uranium-plutonium alloy reacts with either iron- or nickel-base cladding to form low-melting alloys. This was experienced by Atomics International using sodium-bonded stainless steel specimens. Complex compositions were formed on the stainless steel surface in contact with carbon-deficient uranium carbide (4.45 wt % carbon). These reaction products were identified as $U_6Fe-UFe_2$, $U_6Fe-UFe_2-UC$, and/or U_6Fe-UC . Melting temperatures are as low as 725°C (1337°F) and 740°C (1364°F), respectively, in the uranium-iron and uranium-nickel systems. These temperatures are further reduced by plutonium addition.

Although long-term (9600 h), out-of-pile tests indicate that no carburization of the cladding occurs when the carbon content of uranium carbide is 4.8 wt % and below, extensive carburization of sodium-bonded uranium carbide occurs at 1000°F and above when the carbon content exceeds stoichiometric composition.⁸⁸ The in-pile tests conducted on car-

bon-deficient uranium carbides by Atomics International indicate that carbon transfer probably resulted from eutectic formation between the free uranium and cladding material which brazed particulate carbide to the clad surface. The development of fundamental data on the mechanism and kinetics of the transfer reaction as function of compositional changes of both the sodium and fuel carbides should aid in the solution of the problem. It is anticipated that the preparation of strictly stoichiometric mixed uranium-plutonium carbide by the Los Alamos Scientific Laboratory and the introduction of buffered compositions by the Westinghouse Electric Corporation will alleviate the carbon-transfer problem.

This review does not point to major deficiencies in the use of mixed uranium-plutonium carbide fuel. The addition of plutonium decreases the thermal conductivity and probably the thermodynamic stability of the carbide fuel. The fission-gas release from mixed carbide appears to be a function of the integrity of the structure, the operating temperature, and the fuel burnup. Fission-gas retention is good up to a burnup of about 50,000 MWD/T and 1800°F. It appears that a relatively strong and irradiation-resistant clad will be required to contain the fuel in a fast reactor environment at burnups of about 100,000 MWD/T. A sodium bond should decrease the fuel temperature. However, information on the in-reactor thermal conductivity and integrity of the sodium bond is lacking.

Concurrent with continued research on the use of mixed carbides, it is recommended that potential metallurgical advances in the total processing cycle be evaluated in comparison with other fuel systems in terms of reactor physics, reactor operation, and economics of power production. There appears to be no such analysis in the literature to define problem areas and potential economic rewards in the technical development of carbide fuels.

ACKNOWLEDGMENTS

This review was made as part of the program at the Los Alamos Scientific Laboratory for the development of single-phase, mixed uranium-plutonium carbide for fast reactor application. Appreciation is expressed to personnel in the Documentary and Reactor Divisions of the Laboratory who aided in the prepa-

ration of the review. Particular thanks are expressed to Dr. B. J. Thamer with whom the information was discussed, and to Dr. J. A. Leary for his comments on the review.

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