CHEMICAL COMPATIBILITY BETWEEN CLADDING ALLOYS AND ADVANCED FUELS

by

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May 1975

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CHEMICAL COMPATIBILITY BETWEEN CLADDING ALLOYS AND ADVANCED FUELS

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ABSTRACT

The National Advanced Fuels Program requires chemical, mechanical, and thermophysical properties data for cladding alloys. The compatibility behavior of cladding alloys with advanced fuels is critically reviewed. In carbide fuel pins, the principal compatibility problem is cladding carburization, diffusion of carbon into the cladding matrix accompanied by carbide precipitation. Carburization changes the mechanical properties of the cladding alloy. The extent of carburization increases in sodium (versus gas) bonded fuels. The depth of carburization increases with increasing sesquicarbide (M_2C_3) content of the fuel. In nitride fuel pins, the principal compatibility problem is cladding nitriding, diffusion of nitrogen into the cladding matrix accompanied by nitride precipitation. Nitriding changes the mechanical properties of the cladding alloy. In both carbide and nitride fuel pins, fission products do not migrate appreciably to the cladding and do not appear to contribute to cladding attack.

INTRODUCTION

The Advanced Fuels Program has as one of its major goals the review and evaluation of chemical, mechanical, and thermophysical properties data of potential fast reactor fuel candidates. Initial effort in this program led to the issuance of report ANL/CEN/AF-100, a detailed bibliography of properties data on carbides, nitrides, carbonitrides, oxycarbides, and oxynitrides of uranium and plutonium and mixed uranium-plutonium compounds. This effort continued with the issuance of ANL-AFP-2, an interim report on recommended values for equation of state for advanced fuels, and ANL-AFP-3, an interim report on recommended values for thermal conductivity for advanced fuels. This present report will endeavor to review the compatibility behavior of cladding alloys with advanced fuels.

CLADDING ALLOYS COMPATIBILITY WITH ADVANCED FUELS

The chemical compatibility between fuel and cladding can be one of the main life-limiting factors of a fast reactor fuel pin. The formation of new phases at the fuel-cladding interface or other changes in the alloy chemistry of the cladding material can significantly affect the load-bearing capacity of the cladding. In addition, a detailed understanding of the changes that occur near the fuel-cladding interface can be used as a guide in the selection of cladding materials. The cladding materials included in this assessment are given in Table 1.

Uranium Carbide

Uranium carbide-cladding compatibility problems arise mainly from the narrow homogeneity range of single-phase (stoichiometric) UC below 1000°C.^{1,2} This is shown in Fig. 1. A slight change in the C/U ratio leads into adjacent two-phase regions, U + UC and UC + U₂C₃. Hypostoichiometric uranium carbide, containing free uranium metal, is excluded from consideration as a fuel because of possible eutectic reaction with components of the cladding. These reactions mainly lead to the formation of intermetallic compounds of the UFe₂ and UNi₅ type.⁴⁻⁸ Hyperstoichiometric uranium carbide, containing the less stable sesquicarbide phase (U₂C₃), leads to greater carburization of the cladding.^{1,2,7,9} Carburization of the cladding involves transfer of carbon from the fuel to the cladding and diffusion of carbon in the austenite matrix accompanied by carbide precipitation¹⁰ according to the general formula^{8,11,12}

The cladding becomes too brittle for use when the carbon content exceeds 0.6 wt %.²

Fig. 1. Phase Diagram of the Uranium-Carbon System³

TABLE 1. Cladding Alloys under Consideration^a

	Composition, % by Weight											
	Fe	Ni	Cr	C	Al	Ti	Mn	Si	Nb	Mo	W	Other
316 SS	65	12	17	0.08			2.0	1.0		2-3		
330 SS	43	35	19	0.05			1.5	1.25				
A286	53	26	15	0.05	0.2	2.1	1.4	0.4		1-1.75		
M813	39	35	18	0.08	1.4	2.25				4.0		
PE-16	43	35.5	16	0.1	1.2	1.2				3.0		
INCONEL 706	40	41.5	16	0.03	0.2	1.75	0.18	0.18	2.9			
INCOLOY 901	45	42.5	12.5	0.05	0.2	2.8	0.45	0.4		5.7		
INCONEL 718	18.5	52.5	19	0.04	0.5	0.9	0.18	0.18	5.0	3.0		
HT-9	85.5	0.5	12	0.20						1.0	0.5	0.3V

^aFor a discussion of the criteria for the selection of cladding alloys, see S. D. Harkness, R. Carlander, and P. R. Okamoto, ANL/MSD-06 (1973).

Attempts have been made to stabilize single-phase uranium carbide by the addition of buffers. The ideal buffering agent forms a carbide that is more stable than U_2C_3 but is less stable than UC. Thus, the buffer acts as a carbon-atom acceptor from U_2C_3 and a carbon-atom donor to free uranium metal. In both cases, UC is formed and the second phase, U_2C_3 or free uranium metal, is minimized or eliminated. The compounds used as buffers are W-UWC₂,^{13,14} V-UVC₂,^{8,15,16} Mo-UMOC₂,¹² Cr-Cr₂₃C₆,^{8,15-18} Fe-UFe₂,^{17,18} and Ti-TiC.^{12,19} Although buffers offer the advantage of stabilizing single-phase uranium carbide, the use of a buffer creates serious problems. The effect of the buffer on the thermophysical properties and the irradiation behavior of UC (melting point, swelling rate, etc.) and the interactions between buffer and fission products have yet been sufficiently defined.

Carburization of cladding alloys is the principal problem in the use of hyperstoichiometric UC fuel.^{1,2,20-22} Fission products do not migrate appreciably to the cladding and do not participate in cladding attack.^{16,20,21} The extent of carburization depends on temperature, carbon activity, and type of bonding agent (gas or sodium).

Temperature. Above 900°C, carburization proceeds at an extremely rapid rate with type 316 stainless steel.²¹ Acceptable carburization levels are observed below 900°C.

Carbon Activity. The rate of carburization increases with increasing carbon activity (increasing carbon content) of the uranium carbide. This is shown in Fig. 2. The depth and degree of carburization can be predicted from the carbon concentration and the carbon activity at the interior surface of the cladding.²⁴ The carbon activity of uranium carbide fuel is unchanged by the inclusion of fission products.^{16,25,26} Thus, the carbon activity in irradiated UC fuel depends on the C/U ratio.²⁷ The variation of the C/U ratio as a function of burnup may be calculated^{25,28,29} if the stoichiometries of the fission product carbides and intermetallic compounds are known. The stoichiometries can be predicted from phase equilibrium and thermodynamic data. Calculation of the C/U ratio during burnup will be discussed in the review of Phase Equilibria in Advanced Fuel Systems (ANL-AFP-10). This straight-forward approach to determining the extent of carburization may be changed by two factors:

(1) The possibility of radial redistribution of carbon within the fuel rod during burnup. Carbon transfer to the cooler outer regions of the fuel rod-cladding interface via CO/CO₂^{2,21} and H₂/CH₄ mechanisms²² has been proposed. Carbon transfer to the outer regions of the fuel rod would result in a higher carbon activity at the interior surface of the cladding than the carbon activity calculated from the variation of the C/U ratio during burnup. Experimental data to confirm or refute a carbon transfer process are lacking.

(2) The effect of oxygen impurity. The carbon activity is strongly affected by the presence of oxygen impurity in the UC lattice. For example, the carbon activity is decreased by two orders of magnitude when the oxygen content is increased from 150 to roughly 2000 ppm.^{30,31} The carbon activity for varying levels of oxygen impurity can be determined for various C/U ratios. Hence, if the initial oxygen level is known, the effect of the oxygen may be incorporated into the calculation of the carbon activity from the variation of the C/U ratio during burnup. This approach assumes, of course, that any redistribution of oxygen during burnup is known and taken into account.

Fig. 2. Comparison of Transfer of Carbon from Uranium Carbide to the Cladding through Argon and Sodium Bonding²³

Bonding. In all cases, carburization is increased with the use of sodium as a bonding agent.^{2,7,20,32,33} This is shown in Fig. 2. The increased carburization results from the more rapid transfer of carbon to the cladding via the sodium in the annular space between the fuel and cladding. With sodium bonding, diffusion of carbon in the austenite matrix is the rate-limiting step in carburization. In the absence of a sodium bond, transfer of carbon to the cladding is the rate-limiting step.²⁴ While the extent of carburization varies dramatically with type of bond, there is no discernible difference in compatibility behavior between type 316 and type 304 stainless steel. (Type 304 stainless steel is no longer a candidate cladding alloy.) However, of the cladding alloys shown in Table 1, compatibility studies have been undertaken only with type 316 stainless steel.

Plutonium Carbide

By analogy to uranium carbide, problems of plutonium carbide-cladding compatibility are expected to arise mainly from the narrow homogeneity range of single-phase (stoichiometric) PuC. This is shown in Fig. 3. A slight change in the C/Pu ratio leads into adjacent two-phase regions, Pu + PuC and PuC + Pu₂C₃. Hypostoichiometric plutonium carbide is excluded from consideration as a fuel because of possible eutectic reaction with components of the cladding. Hyperstoichiometric plutonium carbide, containing the less stable sesquicarbide phase, (Pu₂C₃), leads to greater carburization of the cladding. Carburization of the cladding involves transfer of carbon from the fuel to the cladding and diffusion of carbon in the austenitic matrix accompanied by carbide precipitation.¹⁰ The cladding becomes too brittle for use when the carbon content exceeds 0.6 wt %.² The limited compatibility data accumulated to date indicate that carburization is the principal compatibility problem.³⁵

Fig. 3. Phase Diagram of the Plutonium-Carbon System³⁴

Uranium-Plutonium Carbide

Uranium-plutonium carbide-cladding compatibility problems arise mainly from the narrow homogeneity range of single phase (stoichiometric) $(U,Pu)C$.^{2, 35-38} This may be seen from an examination of the phase diagrams of UC (Fig. 1) and PuC (Fig. 3), which may be used to elicit information on the expected properties of the analogous pseudobinary $(U,Pu)C$ system. A slight change in the C/M ($M = U + Pu$) ratio leads into adjacent two-phase regions $M + MC$ and $MC + M_2C_3$. Hypostoichiometric mixed (uranium and plutonium) carbide, containing free uranium and plutonium metal, is excluded from consideration as a fuel because of possible eutectic reactions with components of the cladding.^{7, 9} These reactions mainly lead to formation of intermetallic compounds of the $(U,Pu)Fe_2$ and $(U,Pu)Ni_5$ type. Hyperstoichiometric mixed carbide, containing the less stable sesquicarbide phase (M_2C_3), leads to greater carburization of the cladding.^{9, 39-41} Carburization of the cladding involves transfer of carbon from the fuel to the cladding and diffusion of carbon in the austenite matrix accompanied by carbide precipitation¹⁰ according to the general formula¹²

The cladding becomes too brittle for use when the carbon content exceeds 0.6 wt %.²

Attempts have been made to stabilize the single-phase mixed carbide by the addition of buffers. The ideal buffering agent forms a carbide that is more stable than M_2C_3 but is less stable than MC. Thus, the buffer acts as a carbon-atom acceptor in the case of M_2C_3 and a carbon-atom donor in the case of free uranium and plutonium metal. In both cases, MC is formed and the second phase, M_2C_3 or free metal, is minimized or eliminated. The compounds used as buffers are W-(U,Pu)WC₂,¹⁴ Fe-(U,Pu)Fe₂,^{42,43} Cr-Cr₂₃C₆,^{42,43} Mo-(U,Pu)MoC₂,¹² and Ti-TiC.^{12,19} Although buffers offer the advantage of stabilizing single-phase mixed carbide, the use of a buffer creates serious problems. The effect of the buffer on the thermophysical properties and the irradiation behavior (melting point, swelling rate, etc.) of the mixed carbide and the interaction between buffer and fission products have not yet been sufficiently defined.

Carburization of the cladding is the principal problem in hyperstoichiometric mixed carbide fuel observed during in-pile^{35,44-50} and out-of-pile^{9,22,39,41,51-57} tests. No redistribution of uranium or plutonium occurs during irradiation. Fission products do not migrate appreciably to the cladding and do not participate in cladding attack.^{44,48,50} The extent of carburization depends on temperature, carbon activity, and type of bond (helium or sodium).

Temperature. Above 900°C, carburization proceeds at an extremely rapid rate with type 316 stainless steel.¹² Similarly, an upper operational limit of 750°C has been determined for NIMONIC PE-16.⁴⁵ Other than this, there is no discernible difference in compatibility behavior between type 316 stainless steel and NIMONIC PE-16. Acceptable carburization levels are observed below these temperatures. However, of the cladding alloys shown in Table 1, compatibility studies have been undertaken only with type 316 stainless steel and NIMONIC PE-16.

Carbon Activity. The rate of carburization increases with increasing carbon activity. This is shown in Fig. 4 for mixed carbide fuel pins with approximately 5% burnup. The depth and degree of carburization can be predicted from the carbon concentration and the carbon activity at the interior surface of the cladding.²⁴ The carbon activity of uranium carbide fuel is unchanged by the inclusion of fission products.^{16,25,26} Thus, the carbon activity in irradiated UC fuel depends on the C/U ratio.²⁷ Similar results are expected (but not yet experimentally determined) in mixed carbide fuels; that is, the carbon activity in irradiated mixed carbide fuel should depend only on the C/M ratio. The variation of the C/M ratio as a function of burnup may be calculated^{25,28,29} if the stoichiometries of the fission-product and intermetallic compounds are known. The stoichiometries can be predicted from phase equilibrium and thermodynamic data. Calculation of the C/M ratio during burnup will be discussed in the review of Phase Equilibria in Advanced Fuel Systems (ANL-AFP-10). This straightforward approach to determining the extent of carburization may be changed by two factors.

(1) The possibility of radial redistribution of carbon within the fuel rod during burnup. As in the case of uranium carbide, carbon transfer to the cooler outer regions of the fuel rod and to the fuel cladding interface has been proposed via CO/CO₂^{2,21} and H₂/CH₄ mechanisms.^{22,52} Carbon transfer

Fig. 4. Maximum Depth of Carburization of Sodium-Bonded Uranium-Plutonium Carbide Fuel Elements vs. Equivalent Carbon Content⁵⁸

to the cooler regions of the fuel rod would lead to an increased carbon activity at the inner surface of the cladding. The carbon activity calculated solely on the basis of the variation of C/M ratio during burnup (that is, without taking into account carbon transport to the cladding) would be too low. Experimental data to confirm or refute a carbon transfer process is lacking.

(2) The effect of oxygen impurity. Oxygen impurity would affect the estimation of the extent of carburization in much the same way as has been noted in the case of uranium carbide. Although no experimental data on the effect of oxygen on carbon activity are available for the mixed carbide fuel, the effect is expected to approximate that found in the case of uranium carbide, where an increase from 150 to ~2000 ppm oxygen was found to decrease the carbon activity by two orders of magnitude.^{30,31} The carbon activity of the mixed carbide with varying levels of oxygen can be determined for various C/M ratios. Hence if the initial oxygen level is known, the effect of oxygen may be incorporated into the calculation of carbon activity from the variation of the C/M ratio during burnup. This approach assumes, of course, that any redistribution of oxygen during burnup is known and taken into account. In view of the predicted effect of oxygen, it is surprising that out-of-pile compatibility tests show no effect on the carburization of type 316 stainless steel when the oxygen content of the mixed carbide varied between 900-3700 ppm.³⁹

Bonding. As in the case of uranium carbide, the more rapid transfer of carbon to the cladding that results from the use of sodium as a bonding agent increases the rate of carburization.^{38,52} With sodium bonding, diffusion of carbon in the austenite matrix is the rate-limiting step in carburization. In the absence of a sodium bond, transfer of carbon is the

rate-limiting step.²⁴ Sodium-bonded fuel rods exhibit a higher failure rate than helium-bonded fuel rods.⁵⁹ However, this higher failure rate has been attributed to fuel-cladding mechanical interactions rather than increased carburization.

Uranium-Plutonium Nitride

Problems of compatibility between nitride fuels and cladding arise mainly from the narrow homogeneity range of UN and PuN in a manner that is analogous to carbide fuels. Figure 5 indicates that a slight change in the N/U (or N/Pu) ratio leads into adjacent two phase regions U + UN (Pu + PuN) or UN + U₂N₃ (PuN + N₂). Studies with UN,⁶¹ PuN,⁶² and mixed nitrides⁶³⁻⁶⁷ show that hypostoichiometric fuel, containing the free metal, is excluded from consideration as a fuel because of possible eutectic reactions with components of the cladding. These reactions mainly lead to intermetallic compounds of the (U,Pu)Fe₂ and (U,Pu)Ni₅ type. Studies with UN^{64,66,68,69} and mixed nitrides^{64,67} show that hyperstoichiometric fuel, containing the less stable sesquinitride phase (U₂N₃), leads to greater nitriding of the cladding. Nitriding of the cladding involves diffusion of nitrogen in the austenite matrix accompanied by nitride precipitation according to the general formula^{66,68,69}

The consequence of nitriding the cladding is that the strength of the cladding is increased while the ductility is decreased.

Fig. 5. Phase Diagrams of Binary Actinide-Nitrogen Systems⁶⁰

Nitriding the cladding is the principal problem in hyperstoichiometric UN,^{61,63,66,68-75} PuN,^{61-63,66,69} and (U,Pu)N.^{63,65-67,74,76} In-pile studies of UN^{61,75} and (U,Pu)N^{61,65,67,76} show that fission products do not migrate appreciably to the cladding and do not participate in cladding attack. There is, in general, no redistribution of uranium and plutonium in irradiated fuel. However, plutonium redistribution in (U,Pu)N during thermal irradiation has been reported.⁶¹ Out-of-pile studies show no large temperature dependence of nitriding in tests up to 1000°C. Acceptable levels of nitriding were observed below this temperature. No cladding alloy has yet been rejected solely on the basis of compatibility behavior. However, of the cladding alloys shown in Table 1, compatibility studies have been undertaken only with type 316 stainless steel and NIMONIC PE-16.⁶⁹ In general, no difference in nitriding is observed between helium *vs.* sodium bonding.^{63-65,67,74,76} However, increased nitriding with a sodium bond has been reported.⁶⁶

Oxygen affects compatibility studies in two ways: (1) In out-of-pile tests, an oxide layer has been observed on the surface of UN^{66,68,71} and (U,Pu)N.^{64-66,68} Oxygen (apparently from the metal surface of the compatibility test capsule) reacted with stoichiometric UN and (U,Pu)N to form the more stable UO₂ and to increase the nitrogen potential⁶⁶ as given by Eq. 4.

Excess oxygen results in the release of nitrogen⁶⁶ as given by Eq. 5.

(2) UO₂ and U₂N₃ are completely miscible above 1200°C. A homogeneous oxygen content of 2 to 3 wt % stabilizes U₂N₃^{68,77} (and decreases the nitrogen activity). Decreased nitriding of the cladding occurs when UO₂ is present in the U₂N₃ phase.⁶⁶

CONCLUSIONS

Problems of chemical compatibility between cladding alloys and advanced FBR fuels may not be as intense as the corrosion encountered with mixed oxide fuel. The principal problem encountered in the limited number of studies carried out on advanced fuels is the change in the mechanical properties due to carburization or nitriding of the cladding. Compatibility testing needs to be extended to cladding alloys other than type 316 stainless steel and NIMONIC PE-16. In carbide fuel pins, the depth of carburization increases with sodium (*vs.* gas) bonding. The depth of carburization also increases with increasing sesquicarbide (M₂C₃) content of the fuel.

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