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A Review of Compatibility of IFR Fuel and Austenitic Stainless Steel

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Interdiffusion experiments have been conducted to investigate the compatibility of various austenitic stainless steels with U-Pu-Zr alloys, which are alloys to be employed as fuel for the Integral Fast Reactor being developed by Argonne National Laboratory. These tests have also studied the compatibility of austenitic stainless steels with fission products, like the minor actinides (Np and Am) and lanthanides (Ce and Nd), that are generated during the fission process in an IFR. This paper compares the results of these investigations in the context of fuel-cladding compatibility in IFR fuel elements, specifically focusing on the relative interdiffusion behavior of the components and the types of phases that develop based on binary phase diagrams. Results of interdiffusion tests are assessed in the light of observations derived from post-test examinations of actual irradiated fuel elements.

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1. Introduction

Argonne National Laboratory's Integral Fast Reactor (IFR) utilizes a U-Pu-Zr metallic fuel clad in stainless steel [1-2]. Once irradiated, this U-Pu-Zr fuel swells and contacts the cladding. As a result, fuel, cladding, and fission product components interdiffuse and form potentially low melting intermetallic phases that may affect the structural integrity of the cladding. This fuel cladding interaction (FCI) has been investigated using systematic diffusion couple techniques [3-8] that focus on interactions between cladding, both martensitic and austenitic, and U-Zr fuel, U-Pu-Zr fuel, and Ce (a major fission product). This paper will compare the results from interdiffusion experiments using the austenitic stainless steel cladding D9[1] with those from recent melting tests using U-19Pu-10 wt.% Zr fuel and 15-15 Ti stainless steel[2], an austenitic stainless steel close in composition to D9 [9]. Additionally, it will compare the results from diffusion tests and melting tests with those from recent analyses of actual irradiated IFR specimens clad with D9. In the light of available phase diagrams, this paper will discuss the particular phases that can be expected to form in an irradiated D9 clad IFR fuel element in the context of FCI, with comments on the relative diffusion behavior of the fuel, cladding, and fission product components.

2. Main Observations From Diffusion Couple Studies

Keiser and Dayananda [3-5] investigated the interdiffusion behavior of U-23Zr[3] fuel bonded with Fe, Ni, various alloys of the Fe,Ni,Cr system, and the cladding materials HT9 (a martensitic steel with a nominal composition in wt.% of 12.02Cr, 0.57Ni, 1.03Mo, 0.34V, 0.51W, 0.50Mn, 0.22Si, 0.21C, bal Fe), D9, and 316 (an austenitic steel with a nominal composition in wt.% of 16.30Cr, 10.50Ni, 2.11Mo, 1.47Mn, 0.67Si, 0.05C, bal Fe). They noted that the presence of Ni had a drastic effect on the diffusion

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[1] D9 is an austenitic stainless steel with a nominal composition in wt.% of: 13.45Cr, 15.61Ni, 1.65Mo, 0.32Ti, 0.01V, 0.01W, 2.05Mn, 0.85Si, 0.034C, 0.0051N, 0.0062O, bal Fe.

[2] 15-15 Ti has the following ranges in composition in wt.%: 0.08-0.12C, 14.5-15.5Cr, 14.5-15.5Ni, 1.0-1.4Mo, 0.3-0.55Si, 1.5-2.0Mn, 0.04-0.06B, 0.3-0.55Ti, 0.05-0.1(Co+Ta), 0.015Cu, 0.015P, 0.015S, 0.05V, 0.01Ca, 0.01N, bal. Fe.

[3] All compositions will be in atomic percent unless otherwise noted.

structure size and morphology and the relative interdiffusion behavior of the components in diffusion couples annealed at 700°C. The U-23Zr versus D9 couple had a relatively large diffusion structure with many phases. The U-23Zr versus D9 couple had the following intermetallic phases based on point-by-point analysis utilizing a scanning electron microscope and energy-dispersive spectroscopy (SEM-EDS): σ (an approximate composition of 50Fe-50Cr), U(Fe,Ni,Cr)$_2$, U$_6$(Fe,Ni,Cr), high-Zr precipitates, (Zr,U)(Fe,Ni,Cr)$_2$, (Zr,U)$_2$(Fe,Ni,Cr), and a (U,Zr) matrix with up to 6 at.% Fe. The intermetallic designations of the phases are based on binary phase diagrams [10]. The designation of a phase corresponds to the ratio of the sum of the cladding elements (Fe+Ni+Cr) to the sum of the fuel elements (U+Zr). The fuel and cladding elements are listed in parentheses in decreasing order of concentration. All phases will be designated in this fashion throughout this paper. Table 1 enumerates the typical composition ranges for the various phases observed in a U-23Zr versus D9 couple.

Interdiffusion experiments were also conducted at 650°C using U-22Pu-23Zr fuel and D9 cladding [6]. The U-22Pu-23Zr versus D9 couple exhibited the development of 5 distinct phases. These phases included: (U,Pu)(Fe,Ni,Cr)$_2$, (Zr,U)(Fe,Cr,Ni)$_2$, (Zr,U)(Fe,Cr,Ni), (U,Pu,Zr) matrix, and (Zr,U,Pu)$_2$(Fe,Ni). The typical compositions for these phases are presented in Table 2. One common observation noted for both the U-23Zr versus D9 and U-22Pu-23Zr versus D9 couple is that, of the fuel elements, U forms intermetallics with Fe on the cladding side of the diffusion structure and Zr forms intermetallics with Fe on the fuel side of the diffusion structure. This reflects the fact that U diffuses faster towards the cladding than the other fuel components, and Fe diffuses faster towards the fuel than the other cladding components.

To investigate lanthanide fission product interactions with cladding materials, Tortorici and Dayananda performed isothermal furnace anneals using Ce (a major fission product in IFR fuels) and Fe, Ni, selected alloys in the Fe-Ni-Cr system [7], and selected cladding materials [8]. The Ce versus D9 couple developed the three distinct phases Ce(Fe,Ni,Cr)$_3$, Ce(Fe,Ni)$_2$, and Ce$_7$(Ni,Fe)$_3$. The compositions of these phases are presented in Table 3. The Ce$_7$(Ni,Fe)$_3$ was consistently found in all of the Ni-containing couples on the Ce side of the diffusion structure. In Ce versus austenitic steel couples, Ni diffuses faster than Fe.

3. Melting Tests

Recently, Sari et al. [9] reported results from isothermal melting tests. These tests investigated the melting temperatures that would be observed if various alloys containing

U, Pu, Zr, minor actinides, or rare earths were sandwiched between two 15-15 Ti pins, placed in a dilatometer, heated to temperatures at which large dimensional changes occurred, and then cooled to room temperature. Supposedly, the dilatometric analysis enabled one to detect minor phase transformations and to determine the onset of melting at the interface of an alloy-steel diffusion couple. The heat treatment was repeated several times for each sample to check the accuracy of the test as a function of the heating rate and of the state of the surfaces in contact. The specific alloys that were sandwiched between the 15-15 Ti steel pins included (in wt.%, see Table 4 for at.%): U-19Pu-10Zr, U-19Pu-10Zr-2Am-3Np-3.6Nd-1.2Ce-0.2Y, Nd-16Ce-4Y-8Pu-20Am, and Nd-10Ce-10Gd-10Y. All alloys were cast by arc melting. The U-Pu-Zr alloys containing minor actinides and rare earths dissolved Am and Np but not the rare earth components. Rare earth alloys were able to dissolve Am and Pu. Those alloys that consisted of U, Pu, Zr, minor actinides and rare earths were two-phase with a U-Pu-Zr-Np matrix and Am-Pu-Rare Earth (RE) precipitates. These investigators reported seeing noticeable interaction between the various alloys and the 15-15 Ti steel using electron probe microanalysis and metallographic examination, and visually they observed collapse of the samples due to the formation of low melting reaction products.

The intimacy of contact of the samples, the sample cleanliness, and the conditions of the sample surfaces are all unknown for these tests, but interdiffusion did occur and phases did form. These tests were conducted to promote diffusion and to determine when particular phases would melt, not to determine the diffusion kinetics of the process. So, in this respect these tests are noteworthy. Table 4 shows the experimental conditions for the tests, the atomic percent composition of the alloys, and the temperature at which the specimens lost their integrity and collapsed due to the formation of low melting reaction products. The melting temperatures are reported to be accurate to within ±10 degrees. Four different types of couples were run for this investigation, as shown in Table 4, namely the CR13-Steel, CR12-Steel, REMAAL-Steel, and REAL-Steel couples.

The following will describe the types of phases that developed for each of these couples.

3.1   CR13-Steel Couple

The CR13-Steel couple reportedly developed 3 multi-phase regions. The compositions of the various phases are listed in Table 5. By converting the wt.% compositions reported by Sari et al. for the various phases to ones in at.%, it is observed: region 1 contained the three phases $(U,Pu,Zr)(Fe,Cr,Ni)_2$, $(Zr,U,Pu)(Fe,Cr,Ni)_2$, and $(U,Pu,Zr)_6(Fe,Cr,Ni)$; region 2 contained the four phases $(U,Pu,Zr)(Fe,Cr,Ni)_2$, $(U,Pu,Zr)_6Fe$,

$(Zr,U,Pu)(Fe,Cr,Ni)_2$, and $(Zr,U,Pu)_2(Fe,Ni)$; and region 3 consisted of the 3 phases $(U,Pu,Zr)_6Fe$, $(Zr,U,Pu)(Fe,Cr,Ni)_2$, and $(Zr,U,Pu)_2(Fe,Ni)$. Hence, this couple developed 4 distinct types of phases viz. $(U,Pu,Zr)(Fe,Cr,Ni)_2$, $(U,Pu,Zr)_6Fe$, $(Zr,U,Pu)(Fe,Cr,Ni)_2$, and $(Zr,U,Pu)_2(Fe,Ni)$.

3.2 CR12-Steel Couple

The CR12-Steel couple developed phases similar in concentration to those observed for the CR13-Steel couple. Six distinct phases were detected in this couple, and they included: $(U,Pu,Zr)(Fe,Cr,Ni)_2$, $(U,Pu)_6Fe$-with 2 at.% Np, $(Zr,U)(Fe,Cr,Ni)_2$, $(Zr,U)(Fe,Cr)$, $(Zr,U,Pu)_2(Fe,Ni)$, and $(Ce,Nd,Am,Pu,U)(Ni,Fe,Cr)_3$. The phase compositions are listed in Table 6.

3.3 REMAAL-Steel Couple

The REMAAL (Nd-Ce-Am-Y-Pu alloy) versus steel couple formed four phases. The phase compositions are listed in Table 7. Heading from the cladding towards the fuel, a $(Nd,Ce,Pu,Am,Y)Fe_2$ phase developed followed by a $(Nd,Ce,Am,Y)_3Ni$ phase, which contained 42 at.% Nd and 18 at.% Ce, and two phases that were composed of Ce, Nd, Am, and Y. Pu was only detected in the $(Nd,Ce,Pu,Am,Y)Fe_2$ phase, with a concentration of 16 at.%. This amount is higher than the 5 at.% Pu in the original REMAAL alloy.

3.4 REAL-Steel Couple

The REAL (Nd-Ce-Gd-Y alloy) versus steel couple had a diffusion structure very similar to the one described for the REMAAL-Steel couple, except that no $(Nd,Ce,Pu,Am,Y)Fe_2$ phase developed. The reported phases for this couple include: $(Nd,Ce,Y,Gd)_3(Ni,Fe)$-consisting of 50 at.% Nd, two lanthanide-rich (Nd,Ce,Y,Gd) phases, $(Nd,Ce,Y,Gd)(Ni,Fe)_5$, $(Nd,Ce,Y,Gd)(Fe,Ni,Mn)$, and $(Nd,Ce,Y,Gd)Fe_2$.

In terms of the melting results where heating rates were varied along with the number of annealing cycles, Sari et al. report little difference in melting temperatures for the CR13-Steel or CR12-Steel couples (see Table 4). The lowest melting temperature observed for either couple was near $720°C$. On the other hand, the REMAAL-Steel and REAL-Steel couples, which contained no U or Zr, melted at temperatures nearer $625°C$.

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4. As-Irradiated Results

A U-16Pu-23Zr fuel element with D9 cladding irradiated to 11.3 at.% burnup, where burnup is the amount of heavy metals fissioned, has been analyzed using an electron microprobe [11]. Particular attention was given to the development of phase layers at the fuel-cladding interface. The major observations from this study were: (1) Pu is found in high concentrations in a phase layer closest to the unreacted cladding; (2) Fe and Ni penetrate into the fuel to a maximum depth of 175 mm; (3) Ni-Zr phases are observed in the diffusion structure; (4) of the lanthanide-series fission products, Ce and Nd are found in the highest amounts in all regions of the diffusion structure; (5) a 40 µm thick phase layer develops along the fuel-cladding interface, and it contains Ce, Nd, La, and some Cr; (6) lanthanides are not found in high concentrations in the phase nearest the cladding; (7) Fe seems to form phases with U and Pu towards the fuel and primarily with Pu nearest the unreacted cladding; and (8) negligible Ni is observed in the phase nearest the unreacted cladding.

Another electron microprobe analysis of an irradiated U-16Pu-23Zr fuel element with D9 cladding has revealed the development of two different lanthanide-containing phase near the fuel-cladding interface [12]. These phases contain the elements U, Pu, Zr, Nd, Ce, La, Pr, and Pd. The compositions for "Phase 1" and "Phase 2" in atomic percent are 1.15U-1.91Pu-0.3Zr-20.22Pd-23.67La-17.6Ce-3.79Pr-31.35Nd and 3.81Pu-28.94La-29.21Ce-3.91Pr-34.12Nd, respectively. This demonstrates that lanthanide-rich phases will develop near the cladding in irradiated fuel elements.

Related to melting temperatures in irradiated fuel elements, the first signs of liquefaction will depend on many factors, e.g. burnup, linear power, etc, and one of the largest contributing factors is the interdiffusion of lanthanides and cladding constituents to form low melting phases. For an irradiated fuel element with D9 cladding, the lowest observed melting temperature was around 675°C [13]. In this fuel element, which was irradiated to a high burn-up resulting in the generation of a large amount of fission products, the first signs of melting were observed along the fuel-cladding interface where lanthanide-cladding constituent phases developed.

5. Discussion

5.1 Phase Development

If one compares the various types of phases that develop in these reported investigations, it becomes clear that the relative diffusion behavior in the various D9 couples is similar. Firstly, by comparing the U-23Zr versus D9 couple with the U-22Pu-23Zr versus D9 couple, as presented in Tables 1 and 2, one can conclude that even with the presence of Pu no drastic changes occur in the types of phases that will form. With the presence of Pu, the major phases that will develop in the diffusion structure are $(U,Pu)(Fe,Ni,Cr)_2$, $(Zr,U)(Fe,Cr,Ni)_2$, $(Zr,U)(Fe,Cr,Ni)$, (U,Pu,Zr) matrix, and $(Zr,U,Pu)_2(Fe,Ni)$. This result has been corroborated by Sari et al. who report the same types of phases for the CR13-Steel couple, as shown in Table 5. In Sari et al.'s CR12-Steel couple (see Table 6) where not only fuel constituents are present but also lanthanides and minor actinides, the same types of phase develop as discussed previously. A $(Pu,Nd,Ce,Am)(Ni,Fe)$ phase is the only phase that seems to develop due to the presence of lanthanides and minor actinides. This phase could possibly be predicted to develop based on the Pu-Ni, Ce-Ni, and Nd-Ni binary phase diagrams, where the only common intermetallic with Ni on the Pu, Ce, and Nd side of the phase diagram is one with a 50-50 ratio. Furthermore, the development of this Pu-lanthanide-Ni intermetallic in the CR12-Steel couple demonstrates that Ni plays an increased role in determining what phases will form in this couple compared to the U-23Zr versus D9 or U-16Pu-23Zr versus D9 couples where Fe played the most important role.

When the elements U and Zr are not present in diffusion couples with Pu, lanthanides, minor actinides, and D9, completely new types of phases develop based on the REMAAL-Steel and REAL-Steel couples reported by Sari et al, as presented in Tables 7 and 8. When Pu is present in a couple, it is only observed in notable concentrations in the $(Pu,Am,Ce,Nd)(Fe,Ni,Mn,Cr)_2$ phase closest to the unreacted cladding. Based on the Pu-Fe and Am-Fe[4] phase diagrams, the 2 major elements in this phase viz. Pu and Am will form $PuFe_2$ and $AmFe_2$, respectively, when combined with Fe. Nd, which exists in larger concentrations than Ce, and Ni dictate the next phase to develop towards the lanthanide-minor actinide alloy. According to the Nd-Ni and Ce-Ni phase diagrams a Ce_7Ni_3 and Nd_7Ni_3 phase are possible intermetallics for these elements with Ni, and only Nd will form the Nd_3Ni type phase. Yet, the Nd_3Ni type phase is the one observed to form in the REMAAL-Steel couple, probably due to the fact that Nd exists in higher

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[4] The Am-Fe phase diagram has not been calculated but some thermodynamic information is available for this system, and an $AmFe_2$ intermetallic will reportedly form [10].

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concentrations in the original alloy. In the Ce versus D9 couple reported by Tortorici [8] (see Table 3) a Ce$_7$Ni$_3$ type of phase does in fact form. So it is clear that without Nd present it is likely for this intermetallic to form. Comparing the relative diffusion behavior of the Ni and Fe, it seems that even though D9 contains less Ni than Fe, the Ni is diffusing faster than Fe because Ni intermetallics are found closer to the REMAAL alloy side of the diffusion zone. This is consistent with Tortorici and Dayananda who report faster diffusion for Ni and the resulting formation of the Ce$_7$(Ni,Fe)$_3$ intermetallic on the Ce side of the diffusion structure. Therefore, Ni dictates the types of intermetallics that will form on the lanthanide alloy side of a lanthanide alloy versus D9 cladding diffusion couple.

In reference to the effect of Pu concentrations on intermetallic formation, the REAL-Steel couple, which does not contain Pu, does develop a (Nd,Ce,Y,Gd)Fe$_2$ phase that was also observed in the REMAAL-Steel couple. Yet, overall, more types of phases develop in the non-Pu-containing REAL-Steel couple than the REMAAL-Steel couple. This suggests that the presence of Pu will reduce the number of observed phases in a diffusion couple that contains lanthanides and minor actinides. In both couples, lanthanide-rich phases containing Nd, Ce, and Y have been observed, thereby shedding light on what lanthanide-rich phases are likely to form when Nd and Ce are present.

Based on electron microprobe analysis results, irradiated IFR fuel elements with D9 cladding exhibit similar interdiffusion behavior and phase development as compared to the diffusion couple experiments and melting tests discussed in this paper. A Pu-rich phase, probably (Pu,Nd,Cr)Fe$_2$, seems to form nearest the unreacted cladding in the irradiated fuel element, and this phase has negligible Ni. Then, moving towards the fuel, the presence of the fission products Nd and Ce have been identified along with the cladding element Ni, thereby suggesting the possible formation of a (Pu,Nd,Ce)(Ni,Fe) phase. Continuing to move towards the fuel various intermetallics between fuel components and cladding components may form. These can include all or some of the following intermetallics: (U,Pu,Zr)(Fe,Cr,Ni)$_2$, (U,Pu)$_6$Fe, (Zr,U)(Fe,Cr,Ni)$_2$, (Zr,U)(Fe,Cr), and (Zr,U)$_2$(Fe,Ni). In addition, phases consisting almost entirely of lanthanides have been identified to exist by Cohen and Sanecki. The lanthanide-rich phases observed in Sari et al.'s REMAAL-Steel and REAL-Steel couples had similar ratios of Ce and Nd compared to those observed in an irradiated fuel element. This suggests that since lanthanides have mutual solubility and little solubility in the (U, Pu, Zr) matrix they will form lanthanide-rich phases in a fuel element. Also, in cases where lanthanides migrate through the fuel and form lanthanide-rich phases near the cladding, intermetallics may develop due to interdiffusion of lanthanides and cladding constituents.

In this type of circumstance, when U and Zr are not present, a NdNi$_3$ type of intermetallic may form based on the REMAAL-Steel couple.

5.2 Phase Liquefaction

The fuel components U, Pu, and Zr dictate what phases will form at the fuel/cladding interface, even with the presence of minor amounts of actinides and lanthanides, based on the CR12-Steel couple. In this couple, the same phases develop as for the CR13-Steel couple and the U-22Pu-23 Zr versus D9 couple annealed at Argonne (compare Tables 2, 5, and 6), except for the (Pu,Nd,Ce,Am)(Ni,Fe) phase. Even with formation of this phase, the CR12-Steel couple melted at temperatures comparable to what is observed for couples between U-Pu-Zr fuel and austenitic cladding, namely 720-730°C. Based on binary phase diagrams, the CeNi, PuNi, and NdNi intermetallics will melt at 680°C, 800°C and 780°C, respectively. The concentration of Ce is only 5.2 at.% in the (Pu,Nd,Ce)(Ni,Fe) phase while the concentrations of Nd and Pu are 12.9 and 18.4 at.%, respectively. Therefore, based on the results of the CR12-Steel couple, this phase may have melted near 730°C along with the eutectic combination of (U,Pu,Zr)(Fe,Cr,Ni)$_2$ and (U,Pu,Np)$_6$Fe which can melt at 725°C according to the U-Fe phase diagram.

The REMAAL-Steel and REAL-Steel couples display lower melting temperatures than the CR12-Steel and CR13-Steel couples, as shown in Table 4. Melting was observed in these couples at around 620-630°C. The presence of Pu dictated what phase would form nearest the unreacted cladding, viz. (Pu,Am,Ce,Nd)Fe$_2$. The binary PuFe$_2$ intermetallic melts at around 1240°C. Whereas the presence of Nd caused the (Nd,Ce,Am)$_3$Ni phase to form. The binary Nd$_3$Ni intermetallic melts at around 600°C, and a eutectic develops between Nd and Nd$_3$Ni that exhibits melting at around 570°C. Therefore, it is possible that the appearance of an Nd$_3$Ni type of phase contributed to the overall liquefaction of the diffusion structures in these couples.

For actual IFR fuel elements with D9 cladding irradiated to high burn-ups, melting has been observed near 675°C, and based on the diffusion couple results it may be due to eutectic melting that develops between lanthanide-rich phases and the Nd$_3$Ni type of multicomponent phase. In the cases where only small amounts of minor actinides and lanthanides are available in the fuel, the melting behavior of the irradiated fuel will be close to that of the CR12-Steel couple where melting was seen near 725°C. On the other hand, when large lanthanide-rich phases develop adjacent to the cladding, which can be the case for fuel elements irradiated to high burn-ups, a eutectic can develop with the

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Nd3Ni type of multicomponent phase. This eutectic may be where melting first occurs locally at temperatures near 675°C.

6.   Conclusions

The following are the main conclusions of this paper on compatibility of fuel and cladding in IFR fuels with austenitic stainless steel cladding:

1.   U-Pu-Zr fuel alloys coupled with an austenitic stainless steel cladding develop five major phases, namely (U,Pu,Zr)(Fe,Cr,Ni)$_2$, (Zr,U)(Fe,Cr,Ni)$_2$, (U,Pu,Zr) matrix, (Zr,U)(Fe,Cr), and (Zr,U)$_2$(Fe,Ni), and the presence of lanthanides and minor actinides adds a new phase, (Pu,Nd,Ce,Am)(Ni,Fe).

2.   Lanthanide-actinide alloys without U and Zr coupled with an austenitic stainless steel exhibit a (lanthanide)Fe$_2$ type of phase, a Nd3Ni type of phase, and lanthanide-rich phases. When Pu is not present in the couple, additional types of phases such as NdNi and Nd5Ni develop.

3.   The presence of Ni, not Fe, in lanthanide-actinide alloy/austenitic stainless steel cladding couples dictates the types of phases that form on the lanthanide-actinide alloy side of the diffusion structure.

4.   A (Pu,Nd,Ce)Ni type of phase forms in couples between austenitic stainless steel cladding and alloys with U, Pu, Zr, and small amounts of lanthanides and minor actinides. The development of this phase does not lower the expected 725°C melting temperature based on the U-Fe phase diagram.

5.   When lanthanide-actinide alloys without U and Zr are bonded to an austenitic stainless steel cladding, a (Pu,Nd,Ce)Fe$_2$ and Nd3Ni type of phase develops, along with lanthanide-rich phases. Based on the binary Nd-Ni phase diagram, the appearance of the Nd3Ni type of phase adjacent to a lanthanide-rich phase can initiate eutectic melting at temperatures near 600°C.

6.   Irradiated IFR fuels with D9 cladding that do not develop large lanthanide-rich phases at the fuel-cladding interface will exhibit evidence of melting at around 725°C. Whereas, in cases where lanthanide-rich phases do develop near the cladding, a eutectic can develop with the Nd3Ni type of multicomponent phase that will initiate melting at lower temperatures. Those fuel element locations where this eutectic develops may be

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the locations where liquid phases first develop in fuel elements that show signs of liquefaction at temperatures near 675°C.

7.   Future Work

Further investigation of FCI is required for IFR fuel clad with martensitic stainless steels that do not contain Ni, since these types of steels are being considered for use in IFR fuel elements. The presence of Ni has been found to drastically affect the types of intermetallics observed at the fuel-cladding interface, and it is imperative to better understand phase formation when Ni is not present in the cladding. This work should involve experiments like those conducted by Sari et al. using U-Pu-Zr alloys with lanthanides and minor actinides. Furthermore, these alloys should deploy other fission product elements, like Pd, that may play a role in determining the intermetallic phases that form at the fuel-cladding interface.

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Table 1. Composition Ranges for the Phases Observed in the U-23Zr versus D9 Couple in at.% [5].

Phase	Fe	Ni	Cr	U	Zr
σ	56	6	35	2	0
U(Fe,Ni,Cr)$_2$	33-42	10-17	12-22	28-34	0
U$_6$(Fe,Ni)	14	4	1	80	1
Zr precipitates	2	2	1	14	80
two (Zr,U)(Fe,Ni,Cr)$_2$	27	4	35	5	27
phases	8	42	6	4	36
two (Zr,U)$_2$(Fe,Ni,Cr)	13	22	1	2	59
phases	13-31	4-24	0	23-25	38-39
(U,Zr) matrix	6	2	1	72	17

Table 2. Typical Compositions of the Phases Detected in the U-22Pu-23Zr versus D9 Couple in at.% [6].

Phase	Fe	Ni	Cr	U	Pu	Zr
(U,Pu,Zr)(Fe,Cr,Ni)$_2$	49	7	13	24	3	4
(U,Pu,Zr) Matrix	5	1	0	70	18	7
(Zr,U)(Fe,Cr,Ni)$_2$	37	3	25	2	0	32
(Zr,U)(Fe,Cr,Ni)	36	3	15	14	0.6	32
(Zr,U,Pu)$_2$(Fe,Ni)	21	18	0	18	3	40

Table 3. Typical Compositions of the Phases Detected in the Ce versus D9 Couple in at.% [8].

Phase	Fe	Ni	Cr	Ce
Ce(Fe,Ni,Cr)$_3$	50	4	18	28
Ce(Fe,Ni)$_2$	63	3	0	36
Ce$_7$(Ni,Fe)$_3$	1	29	0	70

Table 4. The Compositions of the Alloys Coupled with 15/15 Ti, the Experimental Conditions and the Results for Sari et al. Melting Tests [9].

Couple	Alloy Compositions (at.%)	Cycle	Heating Rate (K/min.)	Temperature (C) Melting
CR13/3 Steel	$U_{61}Pu_{16}Zr_{23}$ 15/15 Ti	1 1a 2	2 2 7	820 740 747
CR13/3H2 Steel	$U_{61}Pu_{16}Zr_{23}$ 15/15Ti	1 2 3	2 7 2	827 727 722
CR12/3 Steel	$U_{51}Pu_{15.6}Zr_{22}Am_{1.6}Np_{2.5}Nd_{5}Ce_{1.7}Y_{0.5}$ 15/15Ti	1 2 3	2 2 7	792 727 737
CR12/3H2 Steel	$U_{51}Pu_{15.6}Zr_{22}Am_{1.6}Np_{2.5}Nd_{5}Ce_{1.7}Y_{0.5}$ 15/15Ti	1 1a 2 3 4 5 6	2 2 2 5 7 7 7	747 727 697 712 717 727 712
REMAAL Steel	$Nd_{55.7}Ce_{17.6}Y_{9}Pu_{5.1}Am_{12.7}$ 15/15Ti	1 2	7 2	697 625
REAL Steel	$Nd_{66.2}Ce_{9.7}Gd_{8.7}Y_{15.3}$ 15/15Ti	1 2 3 4 5	7 1 2 2 2	732 625 630 630 617

Table 5. Composition of the Types of Phases Detected in the CR13-Steel Couple in at.% [9].

Constituent	Region 1			Region 2				Region 3		
	UFe_2	Unknown	$ZrFe_2$	UFe_2	U_6Fe	$ZrFe_2$	$ZrFe$	U_6Fe	$ZrFe_2$	Zr_2Fe
Cr	11.48	3.99	16.14	8.69	0	19.63	0	0	19.61	0.21
Fe	47.87	28.18	44.62	50.78	14.73	42.62	31.28	14.68	42.15	23.79
Ni	5.55	2.06	4.71	6.76	0.36	4.71	12.89	0	2.54	14.44
Zr	7.62	2.65	17.79	4.84	0.23	19.73	28.45	1.15	31.18	38.78
U	22.76	43.42	15.38	26.4	75.01	12.28	23.52	75.47	4.22	20.39
Pu	4.72	19.69	1.36	2.53	9.67	1.03	3.86	8.7	0.31	2.38

Table 6. Composition of the Phases Detected in the CR12-Steel Couple in at.% [9].

Constituent	$(U,Pu,Zr)(Fe,Cr,Ni)_2$	$(U,Pu,Np)_6Fe$	$(Pu,Nd,Ce,Am)(Ni,Fe)$	$(Zr,U)(Fe,Cr,Ni)_2$	$(Zr,U)(Fe,Cr)$	$(Zr,U)_2(Fe,Ni)$
Cr	12.32	0.81	1.0	11.94	7.08	0.46
Fe	51.86	13.54	4.2	49.81	48.89	25.15
Ni	4.61	0.72	45.64	2.14	0.96	11.15
Zr	7.8	0.92	0.71	32.18	27.01	35.74
Ce	0.71	0	5.2	0.05	0	0.08
Nd	0.21	0	12.9	0	0	0
U	19.31	68.96	1.53	3.32	15.43	23.94
Np	0.42	2.92	0.22	0.16	0.12	0.65
Pu	2.63	12.13	18.39	0.39	0.5	2.68
Am	0.12	0	10.22	0	0	0.15

Table 7. Composition of the Phases Detected in the REMAAL-Steel Couple in at.% [9].

Constituent	$(Pu,Am,Ce,Nd)(Fe,Ni,Mn,Cr)_2$	$(Nd,Ce,Am)_3(Ni,Fe)$	(Nd,Ce,Am,Y)	(Nd,Ce,Am,Y,Ni)
Cr	1.94	0.49	0.29	0.56
Mn	1.43	0.47	0.55	0
Fe	59.97	2.29	0.27	0.26
Ni	0.57	22.46	0.51	3.5
Y	3.66	5.18	5.07	1.49
Ce	4.25	18.09	23.05	35.72
Nd	3.04	41.37	58.96	49.77
Pu	16.25	0.47	1.11	0.36
Am	9.89	9.17	10.2	8.33

Table 8. Composition of the Phases Detected in the REAL-Steel Couple in at.% [9].

Constituent	$(Nd,Ce,Y,Gd)Fe_2$	$(Nd,Ce,Y,Gd)(Fe,Mn,Ni)$	$(Nd,Ce,Y,Gd)_5(Ni,Fe)$	(Nd,Ce,Y,Gd)	(Nd,Ce,Y,Gd)	$(Nd,Ce,Y,Gd)_3(Ni,Fe)$
Cr	0.9	0.18	0	0	0	0
Mn	0.99	7.8	0.23	0.25	0.26	0
Fe	63	38.52	5.54	0.25	0.25	3.18
Ni	0.93	3.73	10.33	0	0	20.93
Y	16.1	8.13	12.8	9.69	4.75	10.63
Ce	2.66	14.12	9.18	12.3	21.49	8.85
Nd	9.39	23.41	55.16	69.29	69.58	49.65
Gd	6.03	4.11	6.76	8.22	3.67	6.76