Characterization of phases formed between U-Pu-Mo fuels and Fe-12Cr cladding

Assel Aitkaliyeva, James W. Madden, Brandon D. Miller, Cynthia A. Papesch, James I. Cole

Idaho National Laboratory, Idaho Falls, ID, 83415

Keywords

Metallic fuel, fuel-cladding chemical interaction (FCCI)

Abstract

Exposure to high temperatures and irradiation can lead to interaction between fuel and cladding constituents, inter-diffusion, and formation of brittle or low-melting phases. Therefore, understanding of fuel-cladding interaction (FCCI) is critical for evaluation of fuel performance in a reactor environment. In this contribution, phases formed between U-22Pu-4Mo and U-25Pu-15Mo (in wt%) fuel alloys and Fe-12Cr cladding were characterized using scanning and transmission electron microscopy (SEM/TEM) techniques. Phases formed within FCCI layers in both alloys were identified by implementing selective area diffraction pattern analysis as $\text{Cr}_{0.3}\text{Mo}_{0.7}$ (Im-3m), Fe_2U (Fd-3m), UCrFe (Fd-3m), and Fe_2Pu (Fd-3m). Phases formed at the end of the FCCI layer in the U-22Pu-4Mo alloy included UCrFe (Fd-3m), Fe_2U (Fd-3m), and Cr_2FeO_4 (Fd-3m) while in the U-25Pu-15Mo alloy the phases were consistent with $\text{Cr}_{0.49}\text{Fe}_{0.51}$ ($\text{P4}_2/\text{mnm}$), $\text{Cr}_{0.8}\text{Fe}_{0.2}$ (Im-3m), and UCrFe (Fd-3m).

1. Introduction

Uranium-plutonium-molybdenum (U-Pu-Mo) and uranium-plutonium-zirconium (U-Pu-Zr) fuels, known for their high burn-up and good thermal response, have been considered as candidate fuels for advanced fast reactors [1]. Upon exposure to radiation in nuclear reactor environment, nuclear fuel can swell and transport fission products to the cladding. The contact between fuel matrix fission products and cladding promotes chemical and mechanical interactions, which can compromise integrity of the fuel and cladding. Since chemical compatibility between fuel and cladding is crucial for safe operation of the reactor, it is imperative to understand and mitigate interdiffusion between fuel and cladding. Extensive studies have been carried out on phase relations in binary, ternary, and quaternary U-Pu systems [2-5]. Phases formed in quaternary Fe-U-Pu-Zr systems have been investigated previously but no information on complex Fe-Cr-U-Pu-Mo systems has been found [4-5]. Higher conductivity, lower thermal expansion, and higher melting points of U-Pu-Mo alloys offer several advantages over U-Pu-Zr fuels [6-7]. For instance, Mo is a stronger γ -stabilizer than Zr and therefore should provide stable swelling behavior for U-Pu-Mo alloys [6-7]. In addition, Mo has a lower reaction potential with the cladding, such as Fe, as compared to Zr [6-7]. Lower constituent

redistribution in U-TRU-Mo fuel alloys, which includes migration of minor actinides and lanthanides towards the cladding, offers another advantage of replacing Zr with Mo in U-TRU-Zr systems. This can be explained by the existence of a single γ -phase with body-centered-cubic structure over the fuel operating temperatures (550-950°C) in U-Pu-Mo alloys as compared to three phase fields for U-Pu-Zr alloys over the same temperature region [8-9].

Phases formed in Pu-based systems have traditionally been investigated using thermal analysis and wavelength and energy dispersive spectroscopy (WDS/EDS) in scanning electron microscopes (SEM) [4-5]. The issues associated with characterization of Pu-phases using EDS include unavailability of U and Pu standards for appropriate EDS calibration, spatial resolution, magnification-dependent sensitivity of the technique, and overlaps of U and Pu EDS peaks, all of which restrict accurate quantitative chemical analysis of Pu-based phases in SEM. To avoid such uncertainties, detailed structural and chemical composition analysis of formed complex phases needs to be conducted in transmission electron microscopes (TEM). However, preparation of cross-sectional transmission electron microscopy specimens from radioactive diffusion couples is challenging, if not impossible, by conventional sample preparation means. Conventional sample preparation techniques, such as electro-polishing and broad-beam ion milling, do not permit localized sampling at the spatial resolution of few microns and require close-in hands-on manipulation of the sample for extended periods of time in a glovebox and radiological hood environment. This issue can be prevailed by implementing focused ion beam technology in preparation of site-specific lift-outs from the formed brittle fuel-cladding interaction layers with minimal hands-on sample processing. This allows a thorough cross-sectional examination of all phases formed during annealing. In this contribution, phases formed between U-Pu-Mo fuel alloys and Fe-12Cr cladding after heat treatment at 700°C were investigated using a variety of microscopy tools.

2. Materials and methods

The fuel alloys with nominal compositions of U-21.7Pu-4.3Mo and U-24.8Pu-14.6Mo (in wt%) were arc cast in an inert atmosphere glovebox using electric discharge plasma arc melter, homogenized, and then sectioned into several pieces. The produced fuel **slugs** were sliced and used to assemble diffusion couples. Two diffusion couples were assembled from U-21.7Pu-4.3Mo and U-24.8Pu-14.6Mo (in wt%) fuel ingots and Fe-12Cr cladding. **To simplify, U-21.7Pu-4.3Mo will be referred to as U-22Pu-4Mo and U-24.8Pu-14.6Mo as U-25Pu-15Mo in the text.** Prior to assembling, surfaces of cladding and fuel slugs were polished through a 3 μm diamond solution to a mirror polish. Each fuel slug was placed in contact with cladding material and compressed in a Kovar steel device, which was lined with tantalum foil on the interior for prevention of an undesirable reaction between diffusion couples and the jig. After assembling, diffusion couples were placed in a furnace and annealed at 700°C for 75 hours under continuous

argon flow. Immediately after annealing, the whole assembly was quenched in water to preserve the phases formed upon heat treatment. Diffusion couples were then removed from the compression device, embedded in epoxy, sectioned longitudinally to expose fuel-clad interfaces, and polished through a 1 μm diamond solution to expose fresh surface material. Since ultra-high purity argon used for annealing can contain oxygen and other reacting gases, formation of oxides at the initial interface cannot be overruled. It is possible that some of the formed phases have been stabilized by impurities.

Cross-sectional specimens for transmission electron microscopy (TEM) were prepared in a FEI QUANTA 3D field emission gun (FEG) dual beam focused ion beam/scanning electron microscope (FIB/SEM) using a lift-out approach. Prior to TEM specimen preparation, surface of diffusion couples was examined using energy and wavelength dispersive spectroscopy (EDS/WDS) techniques in JEOL JSM 7000F field emission SEM operated at accelerating voltage of 20 kV. SEM was used to determine the exact location of the fuel-cladding interaction zones and to examine the formed microstructure. To reduce charging in the SEM, a layer of Pd coating was applied to the sample surface prior to the insertion into the instruments. Specimens for transmission electron microscopy were characterized in a JEOL JEM 2010 TEM operated at 200 kV and equipped with Bruker Quantax 200 Esprit 1.9 software and a Bruker 133 silicon drift detector.

3. Results and discussion

3.1 U-22Pu-4Mo alloy

Annealing at 700°C for 75 hours resulted in formation of a substantial fuel-cladding chemical interaction (FCCI) layers in the diffusion couple constructed from U-22Pu-4Mo fuel alloy and Fe-12Cr cladding. Backscattered electron micrographs (BSE) in Figure 1 provide a) low magnification overview of the fuel-clad interface and b) higher magnification view of phases formed within the FCCI layer. The acquired micrographs and X-ray maps revealed substantial diffusion of the fuel into the cladding that resulted in pronounced phase formation. The width of FCCI layer was non-uniform with average value at about 570 ± 30 μm . Detailed examination of the fuel-clad interface revealed formation of several phases within the bulk of FCCI region, identified with white arrows in Fig. 1b. BSE micrograph (Fig. 1b) shows a light-contrast matrix surrounding darker irregular shaped polygons, dark-contrast spherical features containing light-contrast central zones, and irregularly shaped intermediate contrast features. Based on the variations in the widths and sharpness of the edges of different features, one can assume that the features are planar and have different orientations relative to the specimen surface.

Figure 1. a) Low magnification backscattered electron (BSE) micrograph of the FCCI layer and b) higher magnification BSE micrograph of phases formed between U-22Pu-4Mo fuel and Fe-12Cr cladding. Different phases formed within FCCI layer are identified with white arrows in b). Scale bars denote 100 μm in a) and 10 μm in b).

EDX point scans and WDX maps were collected from each identified phase. However, unavailability of standards for transuranic elements in the EDX/WDX software prevented obtaining meaningful quantitative or semi-quantitative compositional data and thus the results are represented in the form of qualitative X-ray map data. X-ray maps depicting spatial distributions of various elements were collected by individually mapping characteristic X-rays from each element of interest with the WDX spectrometer. Figure 2 shows representative X-ray maps collected across the formed interaction zone and corresponding secondary electron micrograph. Each map was adjusted for contrast and brightness to emphasize spatial variations in intensity. Brighter areas in each map represent higher concentrations of the element of interest than less bright areas in the same map. Note that equally bright areas in different maps do not represent comparable concentrations. Elemental X-ray maps shown in Fig. 2 revealed segregation of Mo with Cr and Fe with U and Cr. In addition segregation of Fe with U and Pu with U has been noted. This suggests formation of four distinctive phases within the formed FCCI layer.

Figure 2. Scanning electron micrograph and corresponding X-ray maps of the phases formed between U-22Pu-4Mo fuel and Fe-12Cr cladding upon annealing at 700°C. Scale bars denote 10 μm .

X-ray maps shown in Fig. 2 indicate that the dark-contrast spherical phase is enriched with Cr and Mo, the dark elongated phase is Fe and U rich, the lighter elongated phase features segregation of Fe, U, and Cr, and the lightest phase/matrix has high concentrations of Pu and U. Based on segregation of the elements in X-ray maps, initial phase identification was conducted by consulting alloy phase diagram database [10]. To our knowledge, there are no published phase diagrams that include U, Pu, Mo, Fe, and Cr. Therefore, existing binary and ternary phase diagrams have been consulted during this stage of phase identification. According to the published Cr-Mo binary phase diagram, Cr-Mo enriched phase formed at 700°C in the diffusion couple should correspond to the bcc (Cr, Mo) phase ($\text{Cr}_{0.3}\text{Mo}_{0.7}$) [11-12]. The Fe-U binary phase diagrams indicate formation of following phases at 700°C: Fe_2U , FeU_6 , and orthorhombic U [13-14], which suggests that Fe-U enriched phase could correspond to any of these phases. Assessment of Pu-U binary phase diagrams showed formation of the following phases: βU , (ϵPu , γU), ($\epsilon + L$), and a liquid phase [15-16]. Precise identification of the phases could not be carried out from qualitative X-ray maps shown in Fig. 2.

Instead, identification of the formed phases was conducted in TEM by comparing experimental selective area diffraction (SAD) patterns to simulated patterns from published crystal structures produced using JEMS software [17]. In addition, d spacing of each diffraction pattern was measured and compared to the published crystallographic data. To ensure a thorough characterization and identification of all phases, seven cross-sectional TEM specimens (each approximately $15\ \mu\text{m} \times 10\ \mu\text{m} \times 0.1\ \mu\text{m}$ in size) were lifted-out from different locations within the formed interaction layer. One of the cross-sectional TEM specimens was prepared from the area shown in Fig. 2. Selective area

diffraction (SAD) patterns of phases shown in Fig. 2 were collected, analyzed, and provided in Fig. 3. Phase 1, enriched with Cr and Mo, was identified as $\text{Cr}_{0.3}\text{Mo}_{0.7}$; phase 2, enriched with Fe and U, was identified as Fe_2U ; and phase 3, enriched with Fe, U, and Cr was identified as UCrFe . Due to the varying FIB milling rate and subsequent specimen thickness variation, the Pu-U enriched matrix was not sufficiently thin for SAD analysis and therefore identification of the Pu-U enriched phase has not been conducted.

Figure 3. Scanning electron (SE) micrograph of phases formed after annealing of U-22Pu-4Mo fuel with Fe-12Cr cladding and selective area diffraction (SAD) patterns of phases identified in TEM (highlighted with white arrows). Top row of SAD patterns corresponds to experimental and bottom row to simulated electron diffraction patterns. Scale bar in SE micrograph denotes $10\ \mu\text{m}$.

Figure 4a) shows a low magnification bright-field (BF) transmission electron micrograph overview of the TEM specimen lifted-out from the end of the diffusion zone. Numbers provided in Fig. 4a) indicate location of different phases, which were identified as $\text{Cr}_{0.03}\text{Fe}_{0.97}$, UCrFe , Fe_2U , and Cr_2FeO_4 using SAD analysis (Fig. 4). Experimental and simulated electron diffraction patterns are provided at the bottom of Fig. 4 for reference.

In addition, a higher magnification TEM micrograph of the phases formed at the end of the diffusion zone is provided in Fig. 4b). As it can be seen, the Fe_2U phase is encapsulated by Cr_2FeO_4 phase.

Figure 4. a) Low magnification and b) higher magnification bright field (BF) transmission electron micrographs of the phases formed within fuel-cladding interaction layer, and SAD patterns of phases labeled 1-4. Top row of SAD patterns corresponds to experimental and bottom row to simulated electron diffraction patterns. Scale bars denote 2 μm in a) and 0.2 μm in b).

3.2. *U-25Pu-15Mo* alloy

As in case of the *U-22Pu-4Mo* alloy, the *U-25Pu-15Mo* alloy forms a notable FCCI layer upon annealing at 700°C for 75 hours in contact with Fe-12Cr cladding. After

exposure to high temperatures, significant diffusion of the fuel into the cladding was observed and a BSE micrograph of the formed FCCI layer is provided in Fig. 5a). The width of the FCCI layer was non-uniform with an average value of about $580 \pm 40 \mu\text{m}$. BSE micrographs (e.g. Fig. 5a and b) show a light-contrast matrix surrounding darker irregular shaped polygons and irregular intermediate contrast features, and light-contrast central zones bracketed by dark-contrast spherical features. Contrast variations within the FCCI layer, observed in BSE micrographs, suggest that the sample contains more than one phase. Based on the observed contrast variations, the presence of at least four phases has been identified (denoted with white arrows in Fig. 5b). Similar to the previously discussed diffusion couple, dark-contrast spherical features were abundantly present in this diffusion couple. However, density of observed phases formed in [U-25Pu-15Mo](#) alloy in contact with Fe-12Cr cladding exceeds that of the [U-22Pu-4Mo](#) alloy.

Figure 5. a) Low magnification BSE micrograph of the FCCI layer and b) higher magnification BSE micrograph of phases formed between [U-25Pu-15Mo](#) fuel and Fe-12Cr cladding. Different phases formed within FCCI layer are identified with white arrows in b). Scale bars denote 100 μm in a) and 10 μm in b).

Phase constituents developed in the alloy during annealing of the diffusion couple were analyzed using X-ray maps collected with WDX spectrometer. To determine spatial distribution of elements, WDX maps and SEM micrographs were correlated and are provided in Fig. 6. The elemental constituents can be classified into several groups in which they exhibit similar behavior and segregate into common phases. Qualitative examination of SEM micrographs and X-ray maps shown in Fig. 6 suggests segregation of Mo with Cr and Fe with U. In addition, segregation of Fe with U and Cr in one phase and Pu with U into another phase has been observed. This is in agreement with the results obtained for [U-22Pu-4Mo](#) alloy, which indicates that despite the differences in density, similar phases form in both diffusion couples. Based on existing binary phase diagrams, the features observed in Fig. 6 could correspond to one of the following phases: i) Cr-Mo enriched region to bcc (Cr, Mo) phase ($\text{Cr}_{0.3}\text{Mo}_{0.7}$) [11-12]; ii) Fe-U enriched phase to

either Fe_2U , FeU_6 , or orthorhombic U [13-14]; and iii) Pu-U enriched features to βU , (εPu , γU), ($\varepsilon + \text{L}$), or a liquid phase [15-16].

Figure 6. Scanning electron micrograph and corresponding X-ray maps of the phases formed between U-25Pu-15Mo fuel and Fe-12Cr cladding. Scale bars denote 10 μm .

For consistency, the same number of lift-outs was prepared for TEM characterization from the U-25Pu-15Mo diffusion couple as it has been done in the case of the U-22Pu-4Mo diffusion couple. Similarly, phase identification was conducted in TEM by comparing experimental selective area diffraction patterns to simulated patterns. TEM examination of cross-sectional specimen prepared from the area shown in Fig. 6, confirms that the dark-contrast spherical feature corresponds to the $\text{Cr}_{0.3}\text{Mo}_{0.7}$ phase, light-contrast feature identified with white arrow is Fe_2U , and the intermediate-contrast features are UCrFe . Indexed experimental SAD patterns of identified phases and simulated electron diffraction patterns are provided in Fig. 7. In addition to these three phases, examination of cross-sectional lamella revealed formation of Fe_2Pu phase beneath sample surface in the same region. Even though Fe_2Pu phase has not been observed in U-22Pu-4Mo diffusion couple, its existence cannot be overruled because of rapid oxidation of Pu-containing phases upon exposure to air. It is possible that Fe_2Pu phase was present in U-22Pu-4Mo diffusion couple but was one of the most oxidized phases that could not be characterized using SAD analysis. Further work needs to be conducted to identify all Pu-enriched phases in both diffusion couples.

Figure 7. Scanning electron (SE) micrograph of phases formed after annealing of **U-25Pu-15Mo** fuel with Fe-12Cr cladding and selective area diffraction (SAD) patterns of phases identified in TEM (highlighted with white arrows). Top row of SAD patterns corresponds to experimental and bottom row to simulated electron diffraction patterns. Scale bar in SE micrograph denotes 10 μm .

Low magnification overview of the TEM lamella prepared from the end of the diffusion zone is provided in Fig. 8a. Experimental electron diffraction patterns were collected to identify the phases formed towards the end of the interaction zone (Fig. 8). Similarly to the **U-22Pu-4Mo** alloy (shown in Fig. 4), in this diffusion couple light-contrast cladding is consistent with a $\text{Cr}_{0.03}\text{Fe}_{0.97}$ phase and dark-contrast interaction product between cladding and fuel is consistent with **UCrFe** phase. However, in this alloy Fe_2U and surrounding Cr_2FeO_4 phases have not been observed. Instead, formation of two additional Fe-Cr enriched phases has been found: $\text{Cr}_{0.49}\text{Fe}_{0.51}$ and $\text{Cr}_{0.8}\text{Fe}_{0.2}$. **It could be in part related to oxidation of U-22Pu-4Mo alloy during transfer between instruments.**

Despite the efforts to promptly transfer samples to TEM upon unloading from the FIB, samples were exposed to air for ~5 minutes in order to take radiological smears from the equipment and follow safe radiological practices. Higher magnification bright field TEM micrograph of these phases, formed at the end of the diffusion zone, and their corresponding SAD patterns are shown in Fig. 8b. The summary of all observed phases in both alloys is provided in Table 1.

Figure 8. a) Low magnification and b) higher magnification BF TEM micrographs of phases formed between U-25Pu-15Mo fuel and Fe-12Cr cladding, and SAD patterns of phases labeled 1-4. Top row of SAD patterns corresponds to experimental and bottom row to simulated electron diffraction patterns. Scale bars denote 2 μm in a) and 0.2 μm in b).

Diffusion couple	Phase	Space group	Unit cell parameters, nm
U-22Pu-4Mo vs. Fe-12Cr	Cr _{0.3} Mo _{0.7}	Im-3m (229)	a=0.297
	Fe ₂ U	Fd-3m (227)	a=0.7067
	UCrFe	Fd-3m (227)	a=0.7144
	Cr ₂ FeO ₄	Fd-3m (227)	a=0.8376
	Cr _{0.03} Fe _{0.97}	Im-3m (229)	a=0.2869
U-25Pu-15Mo vs. Fe-12Cr	Cr _{0.3} Mo _{0.7}	Im-3m (229)	a=0.297
	Fe ₂ U	Fd-3m (227)	a=0.7067
	UCrFe	Fd-3m (227)	a=0.7144
	Fe ₂ Pu	Fd-3m (227)	a=0.7183
	Cr _{0.49} Fe _{0.51}	P4 ₂ /mmn (136)	a=b=0.8796; c=0.4558
	Cr _{0.8} Fe _{0.2}	Im-3m (229)	a=0.2872
	Cr _{0.03} Fe _{0.97}	Im-3m (229)	a=0.2869

Table 1. Summary of phases observed in both alloys.

As it has been mentioned, SEM/WDX analysis showed that the FCCI matrix in both diffusion couples is enriched with U and Pu, which according to existing phase diagrams could correspond to βU , (ϵPu , γU), or ($\epsilon + L$) phases. Multiple cross-sectional TEM specimens containing U-Pu enriched matrix have been prepared in the FIB tool but due to differential thinning of diverse phase constituents in these multi-component diffusion couples, U-Pu enriched phases were not sufficiently thin to acquire SAD patterns. Since both U and Pu have larger atomic masses as compared to other elements in these samples, U-Pu enriched phases thin at a much slower rate. In addition, Pu-enriched phases oxidized more rapidly during specimen transfer between instruments than other phases within prepared lamella. In some cases, when U-Pu matrix was thinned more compared to the other phases within same lamella, rapid oxidation of the matrix lead to its transformation into a brittle and highly unstable phase upon exposure to air for short periods of time (30 min or less). Because of these issues, identification of U-Pu enriched phases in TEM has not been conducted. Further work will be conducted to overcome these issues.

Recent paper published by Kim *et. al.* suggested advantages of using U-TRU-Mo fuels over U-TRU-Zr fuel alloys [8]. Fuel constituent migration at the normal fuel operating temperatures should be more pronounced in U-Pu-Zr fuel than U-Pu-Mo alloy due to the differences in thermochemical properties between adjoining phases in the fuel. The redistribution kinetics in U-Pu-Zr fuel has been shown to increase with Pu composition [8]. Since U-Pu-Mo alloy exists in a single γ phase field in the temperature range between 550-950°C, absence of large chemical potential differences that drives constituent migration was assumed to prevent migration of constituents. The results shown in this paper suggest fuel constituent redistribution in U-Pu-Mo fuels to the extent comparable, if not exceeding that of U-Pu-Zr fuels. It is known that Pu provides catalytic

effect and promotes constituent migration by interdiffusion. Since the reported experiments have been conducted in isothermal conditions, migration and reaction between fuel alloy constituents and cladding constituents in these diffusion couples are due to chemical affinities between the elements. There is no microstructural evidence that different concentrations of Pu in fuel ingots influences phases formed in these diffusion couples. However, high concentrations of Pu (>20wt%) in the alloys enhanced constituent migration and extended the length of FCCI layer. For comparison, FCCI layer thickness reported for U-12Pu-9Mo fuel against 304 SS cladding at 650°C was 20 μm [18]. Both discussed diffusion couples formed similar phases upon annealing, with minor differences at the end of the FCCI layer. This article described simpler phases (containing 2-3 elements) formed in these alloys. Identification of more complex Pu-enriched phases will be conducted in the future. Phase identification has been affected by the unavailability of phase diagrams that include more than 3-4 elements, particularly U, Pu, Mo, Fe, and Cr.

4. Summary

This paper identifies and characterizes phases formed between alloys with nominal compositions of U-22Pu-4Mo and U-25Pu-15Mo and Fe-12Cr cladding. Scanning electron and transmission electron microscopy were employed in characterization of formed phases. SEM/WDX examination of both diffusion couples indicates substantial fuel-cladding interaction at 700°C, with thicknesses of FCCI layers in the range of $570 \pm 30 \mu\text{m}$. The results show substantial migration of constituents in U-Pu-Mo fuels. In addition, high concentrations of Pu (>20wt%) enhanced constituent migration and resulted in formation of an extensive FCCI layer. Implementation of FIB instrument allowed preparation of cross-sectional TEM specimens from these transuranic samples for the first time. This allowed determination of compositions and structures of individual phases using the TEM. Phases formed within the fuel-cladding interaction layer in both diffusion couples included $\text{Cr}_{0.3}\text{Mo}_{0.7}$, Fe_2U , UCrFe , and Fe_2Pu . At the end of the diffusion zone in the U-22Pu-4Mo alloy, UCrFe , Fe_2U , and Cr_2FeO_4 phases formed. The $\text{Cr}_{0.49}\text{Fe}_{0.51}$, $\text{Cr}_{0.8}\text{Fe}_{0.2}$ (Im-3m), and UCrFe phases formed in U-25Pu-15Mo alloy at the end of its diffusion zone. The Pu-U enriched matrix phase has not been identified using SAD analysis but based on existing phase diagrams, it could correspond to either βU , (εPu , γU), or ($\varepsilon + L$) phases. Despite the advantages of U-Pu-Mo fuels over U-Pu-Zr alloys, which include higher conductivity, lower thermal expansion, and high melting point, more work is needed to determine suitability of this fuel for fast reactor applications.

Acknowledgements

This work is supported by the U.S. Department of Energy, under DOE Idaho Operations Office Contract DE-AC07-05ID14517, as part of Fuel Cycle Research and Development (FCRD) program of US Department of Energy.

U.S. Department of Energy Disclaimer

The U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U.S. Government purposes. This information was prepared as an account of work sponsored by an agency of the U.S. Government. Neither the U.S. Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. References herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the U.S. Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the U.S. Government or any agency thereof.

References:

1. D. E. Burkes, J. R. Kennedy, T. Hartmann, C. A. Papesch, D. D. Keiser Jr., *J. Nucl. Mater.* **396**, 49 (2010).
2. M. Kurata, T. Ogata, K. Nakamura, T. Ogawa, *J. Alloys Compounds* **271**, 636 (1998).
3. K. Nakamura, M. Kurata, T. Ogata, A. Itoh, M. Akabori, *J. Nucl. Mater.* **275**, 151 (1999).
4. M. Kurata, K. Nakamura, T. Ogata, *J. Nucl. Mater.* **294**, 123 (2001).
5. K. Nakamura, T. Ogata, M. Kurata, T. Yokoo, M. A. Mignanelli, *J. Nucl. Mater.* **304**, 63 (2002).
6. Y.-S. Kim, G. L. Hofman, A. M. Yacout, T.-K. Kim, in: T. Okazaki, J. Bouchard, T. Takeda, Y. Oka (Eds.), *Proc. Internat. Confr. On Fast Reactors and Related Fuel Cycles (FR09), Challenges and Opportunities*, IAEA-CN-176, Kyoto, Japan, 2009, pp. 1-9.
7. A. Landa, P. Söderlind, P. E. A. Turchi, *J. Nucl. Mater.* **414**, 132 (2011).
8. Y.-S. Kim, G. L. Hofman, A. M. Yacout, *J. Nucl. Mater.* **392**, 164 (2009).
9. Y.-S. Kim, S. L. Hayes, G. L. Hofman, A. M. Yacout, *J. Nucl. Mater.* **359**, 17 (2006).
10. ASM Alloy Phase Diagrams Database, P. Villars, editor-in-chief; H. Okamoto and K. Cenzual, section editors; <http://www1.asminternational.org/AsmEnterprise/APD>, ASM International, Materials Park, OH, 2006.
11. P. E. A. Turchi, L. Kaufman, Z. L. Liu, *CALPHAS: Comput. Coupling Phase Diagrams Thermochem.* **30**, 70 (2006).
12. W. Trzebiatowski, H. Ploszek, J. Lobzowski, *Anal. Chem.* **19**, 93 (1947).
13. M. Kurata, T. Ogata, K. Nakamura, T. Ogawa, *J. Alloys Compd.* **271**, 636 (1998).
14. H. Okamoto, in: M. E. Kassner, D. E. Peterson (Eds.), *Phase Diagrams of Binary Actinide Alloys*, The Materials Information Society, Materials Park, OH, 1995.
15. H. Okamoto, *J. Phase Equilib.* **17**, 372b (1996).
16. F. H. Ellinger, R. O. Elliott, E. M. Cramer, *J. Nucl. Mater.* **3**, 233 (1959).
17. P. Stadelmann, JEMS (EMS Java version), in: CIME-EPFL, Lausanne, Switzerland, 2004.
18. D. M. Poole, J. K. Critcheley, J. A. C. Davidson, P. M. French, E. N. Hodkin, M. J. F. Notley, in: E. Grison, W. B. Lord, R. D. Fowler (Eds.), *Plutonium 1960*, Clever-Hume Press, London, 1961, p.627.

Caption

Figure 1. a) Low magnification backscattered electron (BSE) micrograph of the FCCI layer and b) higher magnification BSE micrograph of phases formed between U-22Pu-4Mo fuel and Fe-12Cr cladding. Different phases formed within FCCI layer are identified with white arrows in b). Scale bars denote 100 μm in a) and 10 μm in b).

Figure 2. Scanning electron micrograph and corresponding X-ray maps of the phases formed between U-22Pu-4Mo fuel and Fe-12Cr cladding upon annealing at 700C. Scale bars denote 10 μm .

Figure 3. Scanning electron (SE) micrograph of phases formed after annealing of U-22Pu-4Mo fuel with Fe-12Cr cladding and selective area diffraction (SAD) patterns of phases identified in TEM (highlighted with white arrows). Top row of SAD patterns corresponds to experimental and bottom row to simulated electron diffraction patterns. Scale bar in SE micrograph denotes 10 μm .

Figure 4. a) Low magnification and b) higher magnification bright field (BF) transmission electron micrographs of the phases formed within fuel-cladding interaction layer, and SAD patterns of phases labeled 1-4. Top row of SAD patterns corresponds to experimental and bottom row to simulated electron diffraction patterns. Scale bars denote 2 μm in a) and 0.2 μm in b).

Figure 5. a) Low magnification BSE micrograph of the FCCI layer and b) higher magnification BSE micrograph of phases formed between U-25Pu-15Mo fuel and Fe-12Cr cladding. Different phases formed within FCCI layer are identified with white arrows in b). Scale bars denote 100 μm in a) and 10 μm in b).

Figure 6. Scanning electron micrograph and corresponding X-ray maps of the phases formed between U-25Pu-15Mo fuel and Fe-12Cr cladding. Scale bars denote 10 μm .

Figure 7. Scanning electron micrograph of phases formed after annealing of U-25Pu-15Mo fuel with Fe-12Cr cladding and selective area diffraction (SAD) patterns of phases identified in TEM (highlighted with white arrows). Top row of SAD patterns corresponds to experimental and bottom row to simulated electron diffraction patterns. Scale bar in SE micrograph denotes 10 μm .

Figure 8. a) Low magnification and b) higher magnification BF TEM micrographs of phases formed between U-25Pu-15Mo fuel and Fe-12Cr cladding, and SAD patterns of phases labeled 1-4. Top row of SAD patterns corresponds to experimental and bottom

row to simulated electron diffraction patterns. Scale bars denote $2\ \mu m$ in a) and $0.2\ \mu m$ in b).

Table 1. Summary of phases observed in both alloys.

Figures

Figure 1.

Figure 2.

Figure 3.

Figure 4.

Figure 5.

Figure 6.

Figure 7.

Figure 8.

Diffusion couple	Phase	Space group	Unit cell parameters, nm
U-22Pu-4Mo vs. Fe-12Cr	$\text{Cr}_{0.3}\text{Mo}_{0.7}$	Im-3m (229)	a=0.297
	Fe_2U	Fd-3m (227)	a=0.7067
	UCrFe	Fd-3m (227)	a=0.7144
	Cr_2FeO_4	Fd-3m (227)	a=0.8376
	$\text{Cr}_{0.03}\text{Fe}_{0.97}$	Im-3m (229)	a=0.2869
U-25Pu-15Mo vs. Fe-12Cr	$\text{Cr}_{0.3}\text{Mo}_{0.7}$	Im-3m (229)	a=0.297
	Fe_2U	Fd-3m (227)	a=0.7067
	UCrFe	Fd-3m (227)	a=0.7144
	Fe_2Pu	Fd-3m (227)	a=0.7183
	$\text{Cr}_{0.49}\text{Fe}_{0.51}$	P4 ₂ /mmn (136)	a=b=0.8796; c=0.4558
	$\text{Cr}_{0.8}\text{Fe}_{0.2}$	Im-3m (229)	a=0.2872
	$\text{Cr}_{0.03}\text{Fe}_{0.97}$	Im-3m (229)	a=0.2869

Table 1.