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High-Temperature Chemical Compatibility of As-Fabricated TRIGA Fuel and Type 304 Stainless Steel Cladding

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ABSTRACT

Chemical interaction between TRIGA fuel and Type-304 stainless steel cladding at relatively high temperatures is of interest from the point of view of understanding fuel behavior during different TRIGA reactor transient scenarios. Since TRIGA fuel comes into close contact with the cladding during irradiation, there is an opportunity for interdiffusion between the U in the fuel and the Fe or Ni in the cladding (Type 304 or Incoloy 800) to form an interaction zone that contains U-(Fe,Ni) phases. Assuming Type 304 cladding and based on the equilibrium U-Fe phase diagram, a eutectic can develop at a composition between the U_6Fe and UFe_2 phases. This eutectic composition can become a liquid at around 725˚C. From the standpoint of safe operation of TRIGA fuel, it is of interest to develop better understanding of how a phase with this composition may develop in irradiated TRIGA fuel at relatively high temperatures. One technique for investigating the development of a eutectic phase at the fuel/cladding interface is to perform out-of-pile diffusion-couple experiments at relatively high temperatures. This information is most relevant for lightly irradiated fuel that just starts to touch the cladding due to fuel swelling. Similar testing using fuel irradiated to different fission densities should be tested in a similar fashion to generate data more relevant to more heavily irradiated fuel. This report describes the results for TRIGA fuel/Type-304 stainless steel diffusion couples that were annealed for one hour at 730 and 800°C. Scanning electron microscopy with energy- and wavelength-dispersive spectroscopy was employed to characterize the fuel/cladding interface for each diffusion couple to look for evidence of any chemical interaction. Overall, negligible fuel/cladding interaction was observed for each diffusion couple.

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ACRONYMS

FCCI	fuel/cladding chemical interaction
HEPA	high-efficiency particulate air
INL	Idaho National Laboratory
LEU	low enriched uranium
SEM	scanning electron microscopy
SS	stainless steel
TRIGA	Training, Research, Isotopes, General Atomics
U	uranium
Zr	zirconium
ZrH	zirconium hydride

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1. INTRODUCTION

During irradiation of a nuclear fuel element, the fuel swells and eventually contacts the cladding. For TRIGA fuel elements, this results in intimate contact between the uranium-zirconium-hydride fuel and Type-304SS or Incoloy cladding [1]. As a result, interdiffusion can occur between fuel and cladding constituents to form phases. The quality of contact at the interface and in the presence of impurities and impurity-containing layers will impact the amount of interaction at a specific fuel/cladding interface location. For various TRIGA-element transient scenarios, the temperature at this interface can be relatively high and, as a result, it is of interest to develop better understanding of the temperature at which liquid phases can develop in the interaction zone and the kinetics of this reaction. Also, because the uranium is the secondary phase in a zirconium-hydride matrix, reaction with a single uranium particle does not necessarily propagate to other particles, and this could significantly affect interaction rates.

Since diffusion-couple experiments use polished samples, and pressure is applied at the fuel/cladding interface of the samples, these tests are considered conservative relative to the situation that may develop for irradiated fuel elements. This paper describes the results for TRIGA 30/20 fuel/Type-304 stainless steel diffusion couples that were annealed for one hour at 730 and 800°C. The TRIGA 30/20 fuel is comprised of 30 wt.% U, enriched at less than 20% U-235, and is henceforth called “TRIGA fuel” for convenience. This fuel is clad in Type 304 stainless steel; hence, the choice of diffusion-couple materials. But testing like this could also be done with Incoloy 800, an alternative cladding for some TRIGA fuels. After annealing, scanning electron microscopy with energy- and wavelength-dispersive spectroscopy (SEM/EDS/WDS) was employed to characterize the fuel/cladding interface for each diffusion couple to look for evidence of any chemical interaction.

2. EXPERIMENTAL

The TRIGA fuel employed in the diffusion couples was fabricated by hydriding an alloy that was a solid solution of uranium in zirconium. The zirconium was selectively hydrided, and the uranium remained as small metallic inclusions in the zirconium hydride matrix. The composition of the Type 304SS, based on certification from the vendor, is enumerated in Table 1.

Table1. Chemical composition of Type 304SS.

Element	Wt%
C	0.021
P	0.037
Si	0.298
Ni	8.33
Cu	0.374
N	0.082
Ti	0.005
Sn	0.013
V	0.07
Nb+Ta	0.046
Mn	1.77
S	0.026
Cr	18.26
Co	0.128
Mo	0.39
Nb	0.046
Al	0.004
B	0.001
Fe	70.099

Disks were machined from the 0.635-cm-diameter Type 304SS rod, and samples were sliced from the TRIGA fuel. These samples were first mechanically ground through 1200-grit sandpaper and then polished using an oil-based solution with 3-micron diamond paste to achieve a smooth, well-polished surface.

To perform a diffusion-couple experiment, a small piece of TRIGA fuel was inserted between two Type-304SS samples and placed in a stainless steel assembly (see Figure 1). A Type-304SS screw was employed to impart a force at the two interfaces between the TRIGA fuel and Type 304SS to ensure good contact during the annealing treatment. Torque was applied using a digital torque meter. The steel assembly was inserted into a conventional stainless steel vertical-tube furnace, capped with a flange at the top. The steel vessel was connected to a vacuum pump via a high-efficiency particulate air (HEPA) filter. The environment inside the vessel was pumpled down to a vacuum (10^{-3} torr) and then heated to temperature. Two diffusion-couple experiments were performed, one at 730°C and one at 800°C. Each temperature was maintained for around one hour, and then the furnace was turned off and allowed to cool overnight. Figure 2 shows the temperature profile observed for the 730°C test, based on thermocouple measurement.

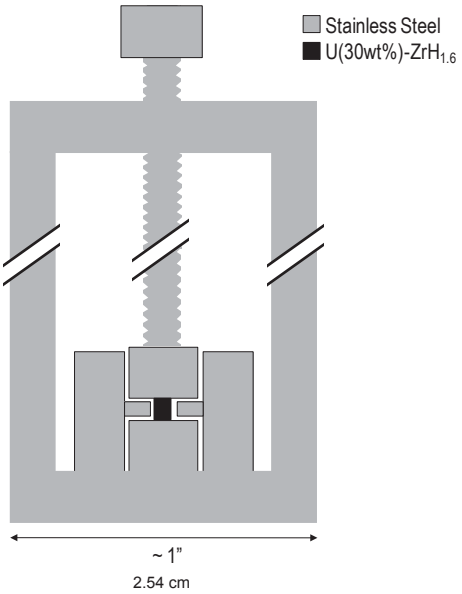


Figure 1. Schematic diagram of TRIGA fuel clampled between two Type-304SS samples inside the stainless steel assembly.

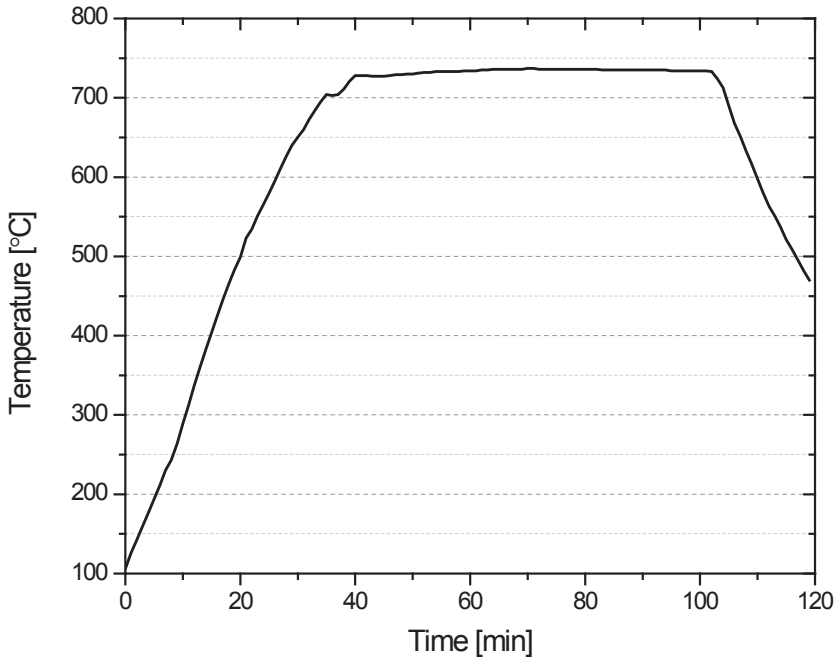


Figure 2. Temperature profile for diffusion couple experiment conducted at 730°C.

After the furnace anneal was complete, the steel assembly was removed from the furnace, and the portion of the assembly with the diffusion-couple stack was mounted in epoxy. This part of the assembly was sliced from the rest of the assembly. Each epoxy-mounted diffusion-couple stack was sliced longitudinally and then polished metallographically to a fine finish. Analysis using SEM/EDS/WDS was performed at the TRIGA fuel/Type 304SS interfaces to identify the quality of bonding, the amount of interdiffusion, and whether any evidence of melting could be observed. SEM/EDS/WDS analysis was also performed on the as-received TRIGA fuel.

3. RESULTS

3.1 As-fabricated Characterization

Figure 3 shows an SEM backscattered-electron (BSE) image of the microstructure observed for the as-received fuel, and Figure 4 presents x-ray maps that were generated for U and Zr. The uranium is contained in a fine precipitate, dispersed in a zirconium-hydride matrix. Point-to-point compositional analysis indicated that the precipitate phases also contained a few percent of Zr.

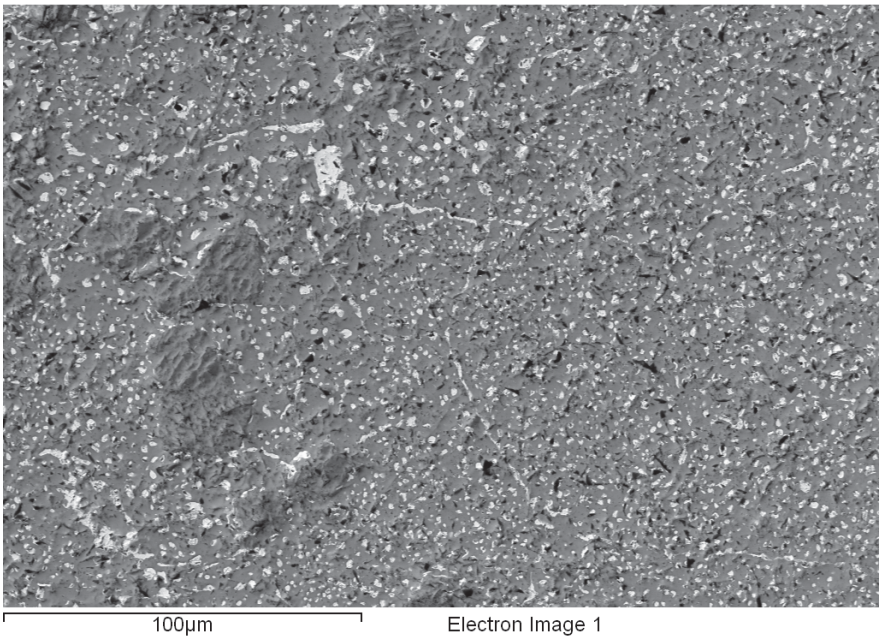


Figure 3. BSE image of the as-fabricated TRIGA fuel-element microstructure. The dark contrast phase is zirconium-hydride matrix, and the bright precipitate is the uranium-rich metallic phase. The black areas appear to be pores.

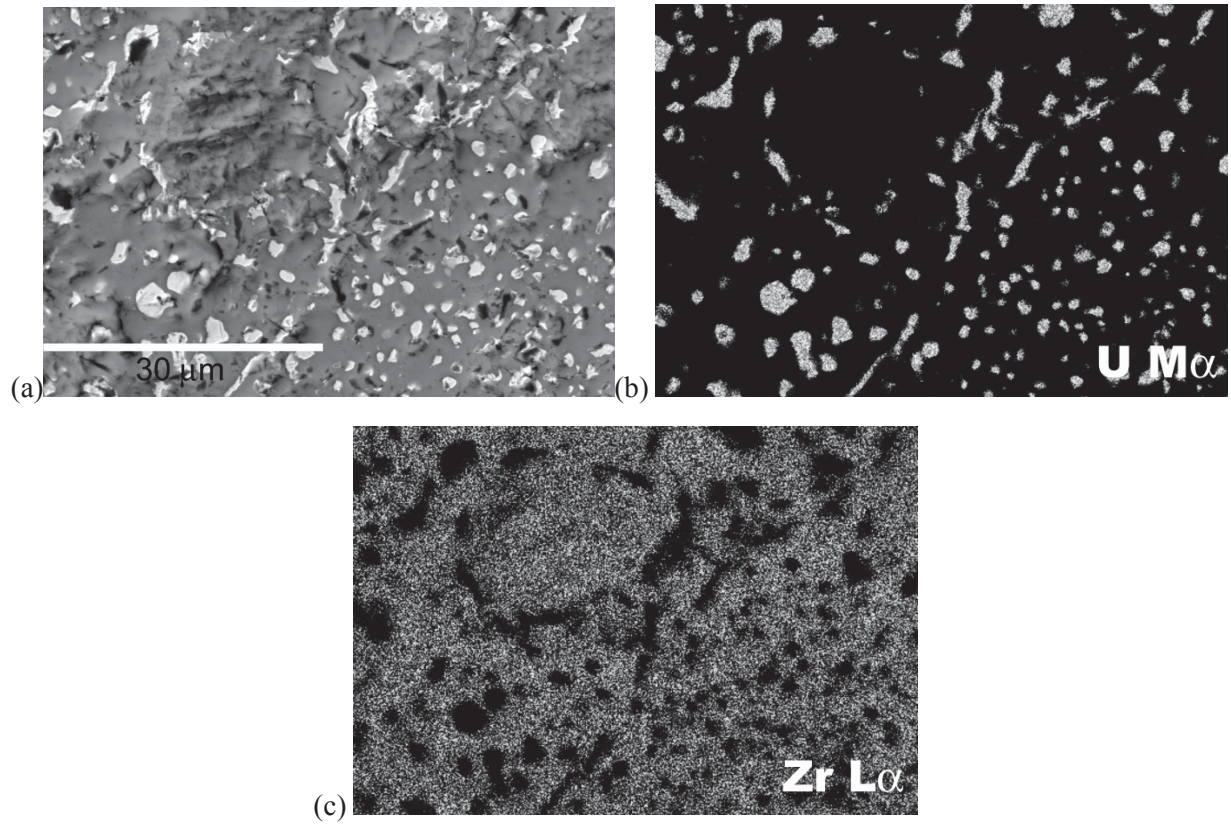


Figure 4. BSE image (a) of the as-fabricated TRIGA fuel element microstructure. WDS x-ray maps are presented in (b) for uranium and (c) for zirconium.

3.2 Diffusion Couple Annealed at 730°C for 1 Hour

Figure 5 shows low-magnification images of the microstructures observed at the TRIGA fuel/Type 304SS interfaces after a diffusion-couple assembly is heated at 730°C for 1 hour. Higher-magnification images taken at the TRIGA fuel/Type 304SS interface are presented in Figure 6. Careful analysis of the interface suggested that there was good bonding (i.e., no gaps or voids were observed). In order to look for evidence of interdiffusion of fuel and cladding components, both point-to-point and linescan analysis were employed across the fuel/cladding interface. No evidence of interdiffusion was observed.

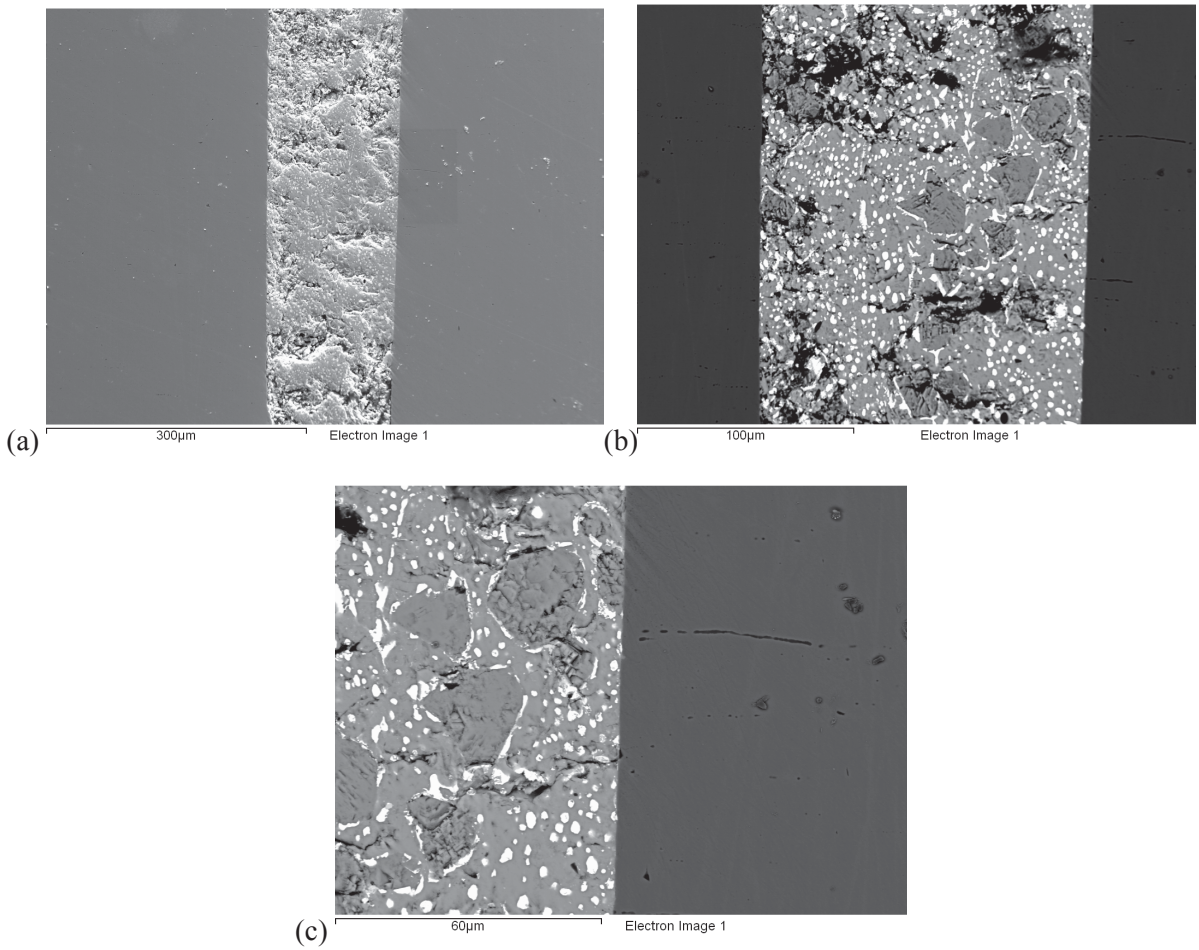


Figure 5. Low-magnification secondary electron (SE) (a) and BSE image (b) showing the microstructure of the diffusion couple comprised of TRIGA fuel in between two pieces of Type 304SS after annealing at 730°C for 1 hour. The BSE image in (c) focuses on the fuel/Type 304SS interface. Some sample pullout of the fuel is observed during polishing.

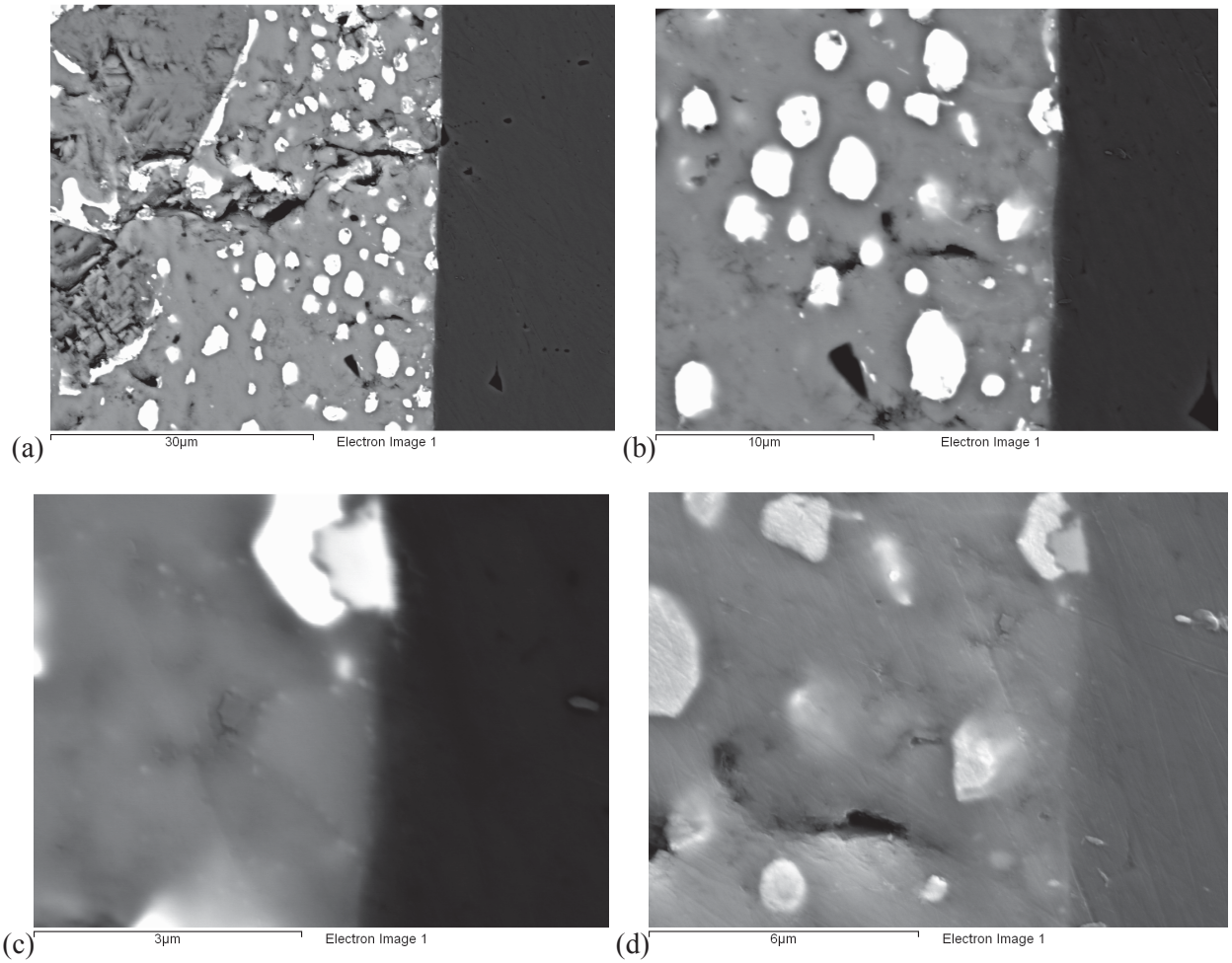


Figure 6. Higher-magnification BSE images (a-c) and a SE image (d) showing the microstructures at the TRIGA fuel/Type 304SS interfaces after an annealing treatment at 730°C for 1 hour.

3.3 Diffusion Couple Annealed at 800°C for 1 Hour

Figure 7 shows the microstructures observed at the TRIGA fuel/Type 304SS interfaces after a diffusion-couple assembly is heated at 800°C for 1 hour. Higher-magnification images taken at the TRIGA fuel/Type 304SS interface are presented in Figure 8. Good interface contact was observed all along the fuel/cladding interface, except for one location, shown in Fig. 8(b). In order to look for evidence of interdiffusion of fuel and cladding components, compositional analyses using linescans, point-to-point analysis, and x-ray mapping were employed across the fuel/cladding interface. X-ray maps for U, Zr, Fe, Ni, and Cr (see Fig. 9) and WDS linsescans for U, Zr, and Fe (see Fig. 10) show the negligible interdiffusion that was observed across the fuel/cladding interface (i.e., no cladding components were in the fuel, and no fuel components were in the cladding). A WDS linescan for oxygen was generated across the fuel/cladding interface to see whether oxide layers were present (see Fig. 10e). No apparent layer was observed.

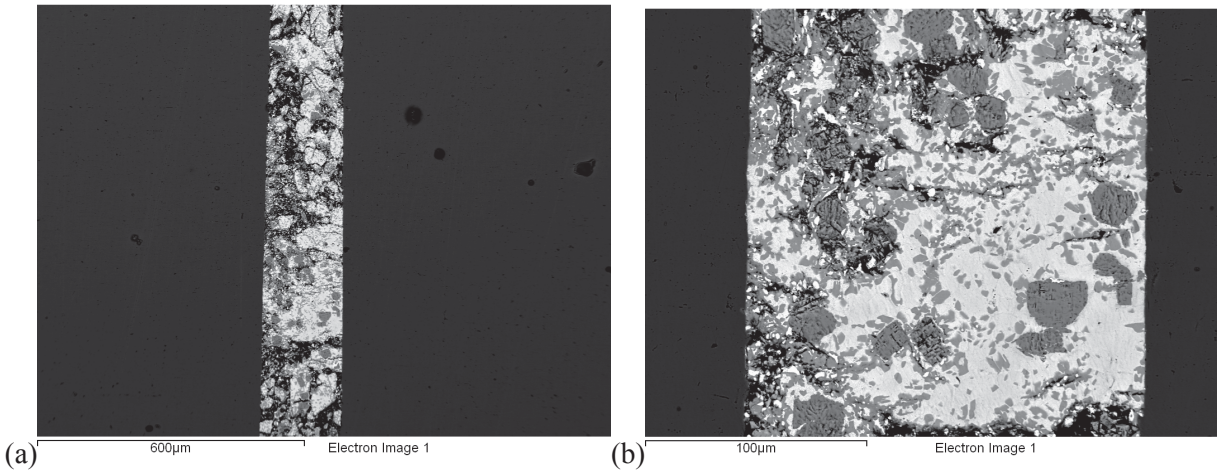


Figure 7. BSE images showing the microstructures at the two TRIGA fuel/Type 304SS interfaces after an annealing treatment at 800°C for 1 hour. Some sample pullout is of the fuel is observed during polishing.

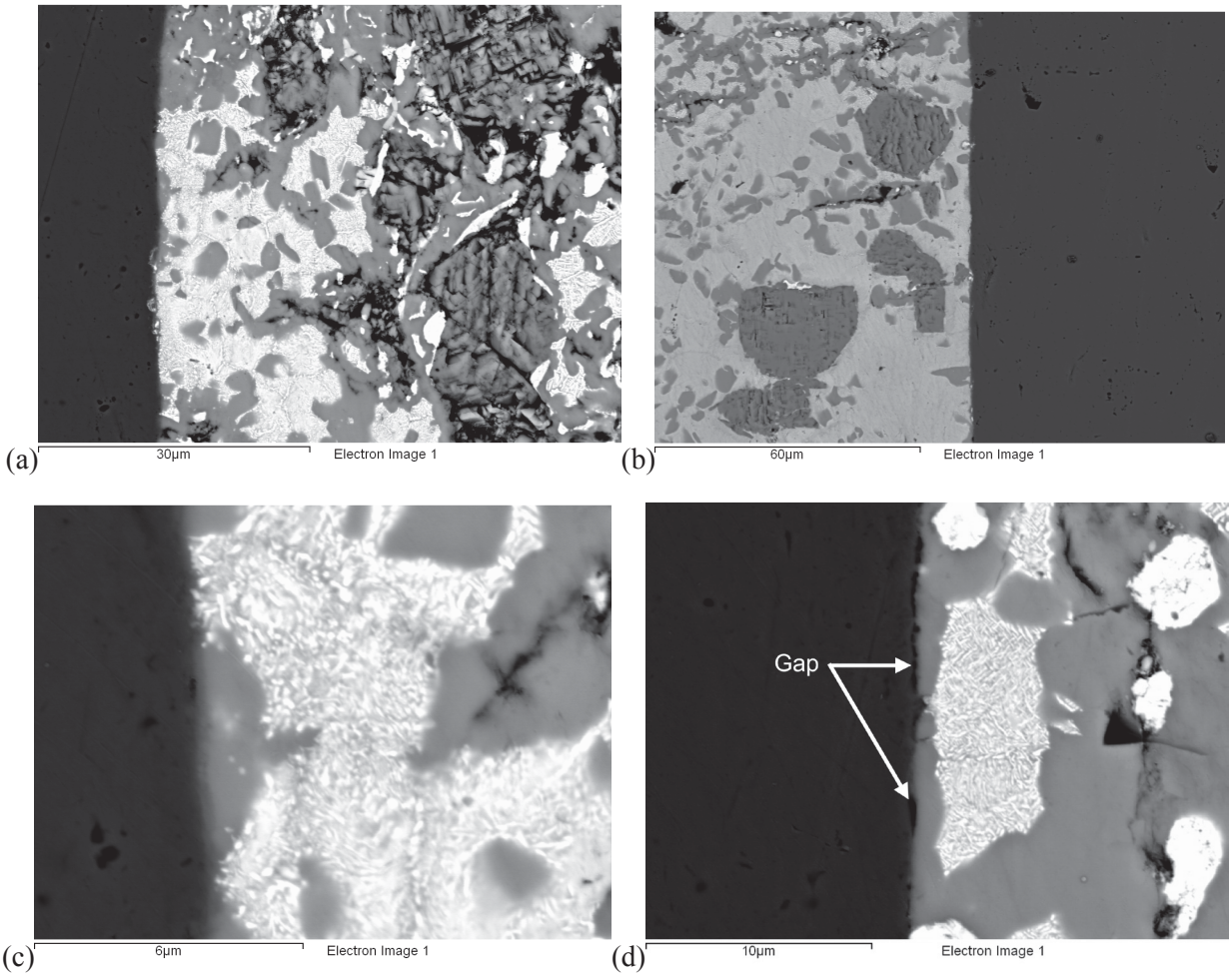


Figure 8. SE image (a) and BSE images (b-d) showing the microstructures at the TRIGA fuel/Type 304SS interfaces after an annealing treatment at 800°C for 1 hour. The only evidence of poor cladding/fuel contact was manifest as the gap shown in (d).

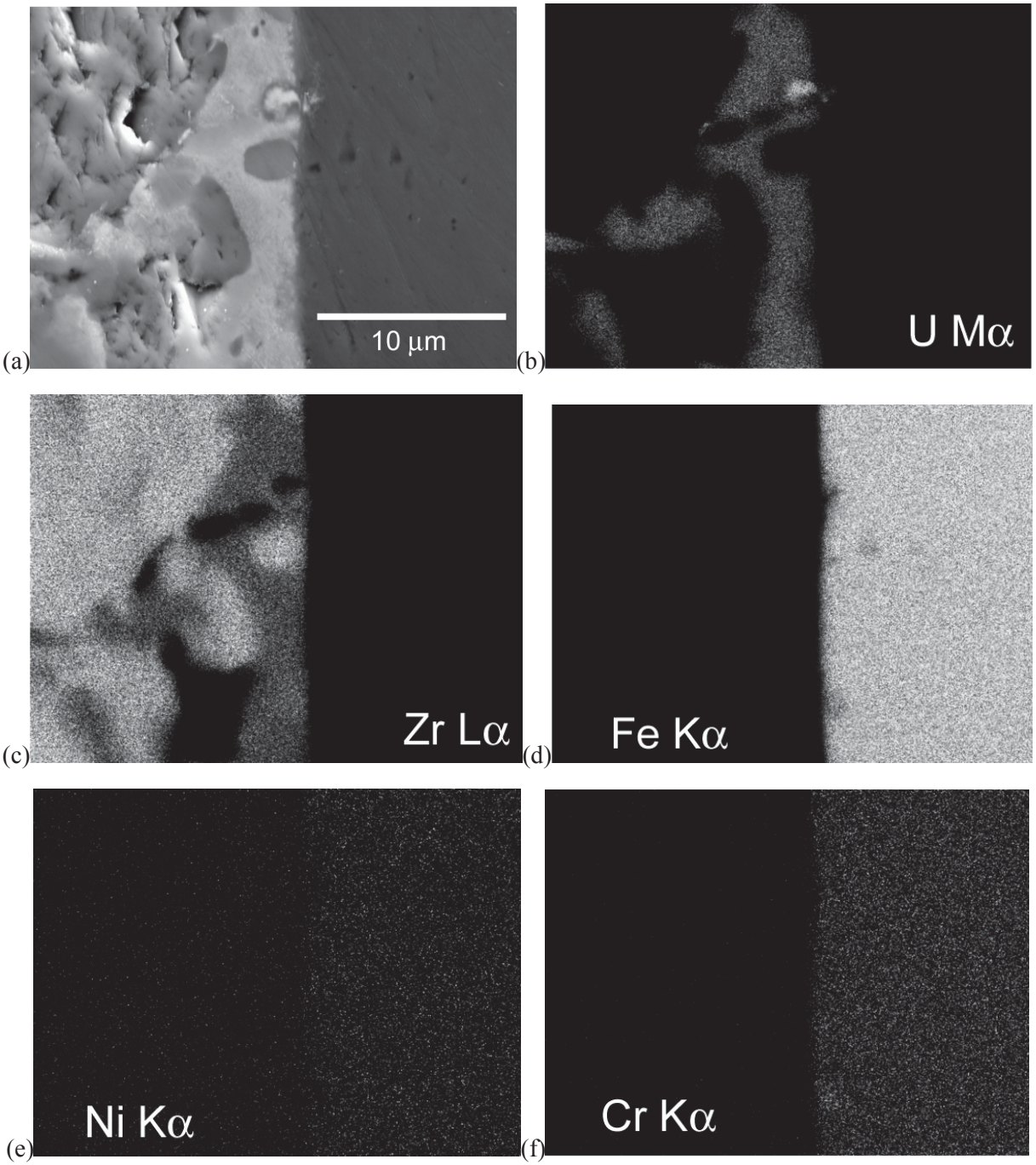


Figure 9. BSE image (a) and x-ray maps for (b) U, (c) Zr, (d) Fe, (e) Ni, and (f) Cr.

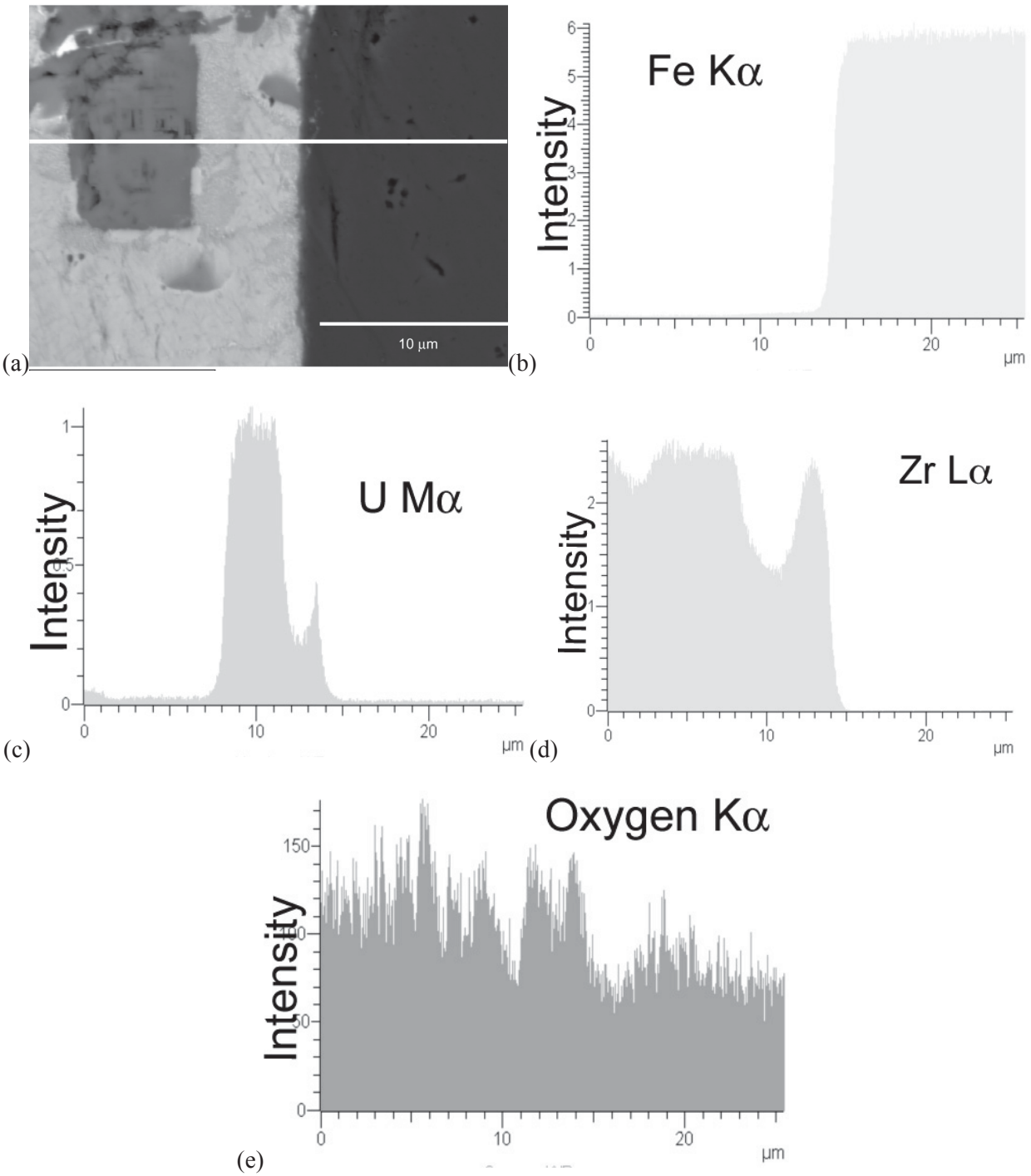


Figure 10. BSE image (a) where WDS linescans of relative intensity were taken for (b) Fe, (c) U, (d) Zr, and (e) oxygen.

4. DISCUSSION

The diffusion-couple experiments discussed in this paper can be considered as conservative tests. Typically, TRIGA fuel has a stable oxide layer on the surface [2] that could act as a diffusion barrier. However, in the case of the samples tested for this investigation, polished samples were employed, and the testing was performed in vacuum. As a result, the amount of oxygen in the system was very low and negligible oxide-layer formation was observed on the surface of the TRIGA fuel. Therefore, the chance of impurity layers being present at the fuel/cladding interface was significantly reduced. Analysis was performed for each diffusion couple at the fuel/cladding interface to look for the presence of oxide layers. None was found. Instead, well-contacted interfaces were routinely observed based on high-magnification imaging, linescan analysis, and x-ray mapping.

For the 730°C couple annealed for 1 hour, the fuel/cladding interaction of interest would only be observed in the local areas where the U-rich phase was in contact with the cladding. Since a large fraction of the TRIGA-fuel microstructure is comprised of a matrix zirconium-hydride phase, it would be expected that in only a few areas would U-rich phase be in contact with the cladding. The images in Fig. 6 show some of the local regions where U-rich precipitates are in contact with the cladding. Based on the U-Fe phase shown in Fig. 11, the annealing temperature for this diffusion couple was slightly above the 725°C temperature at which a eutectic phase could have been present at the fuel/cladding interface if there had been some fuel/cladding interaction. This would have occurred if a U-Fe composition near 10 wt.% Fe had been present in a diffusion zone. Since Zr is also present in the U-rich phase, not to mention the matrix, it would likely participate in the interdiffusion process if it were to occur. The result could be the development of a U-Zr-Fe phase. Based on the U-Zr-Fe phase diagrams available in the literature for 700 and 800°C (see Fig. 12), at certain compositions of U, Zr, and Fe, a liquid phase can develop at 800°C, but not at 700°C. Since hydrogen is a major component in the TRIGA fuel, it will participate in the interdiffusion process to some extent, depending on the temperature. The potential phases that can develop in the U-Zr-H-Fe-Ni-Cr system have not been reported. Since SEM/EDS/WDS is not an effective technique for characterizing samples with hydrogen, not much can be concluded with respect to the interdiffusion behavior of hydrogen in these diffusion couples.

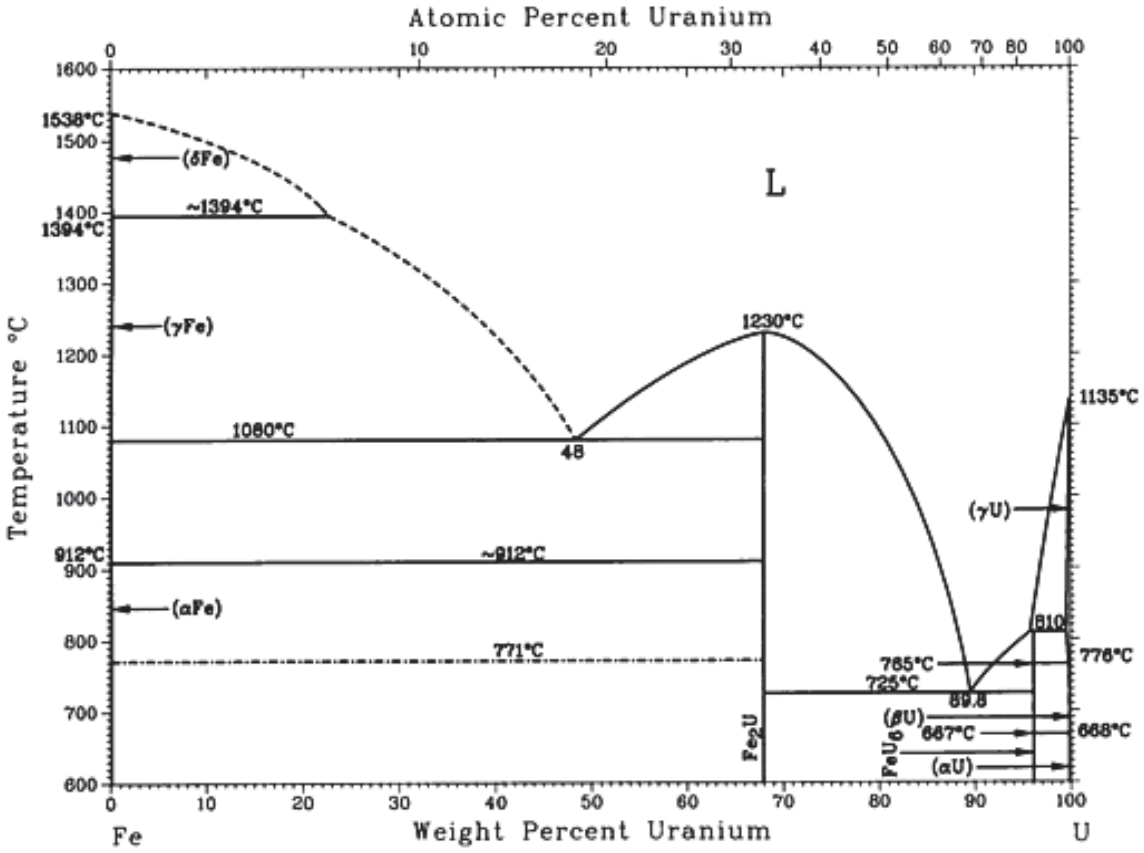
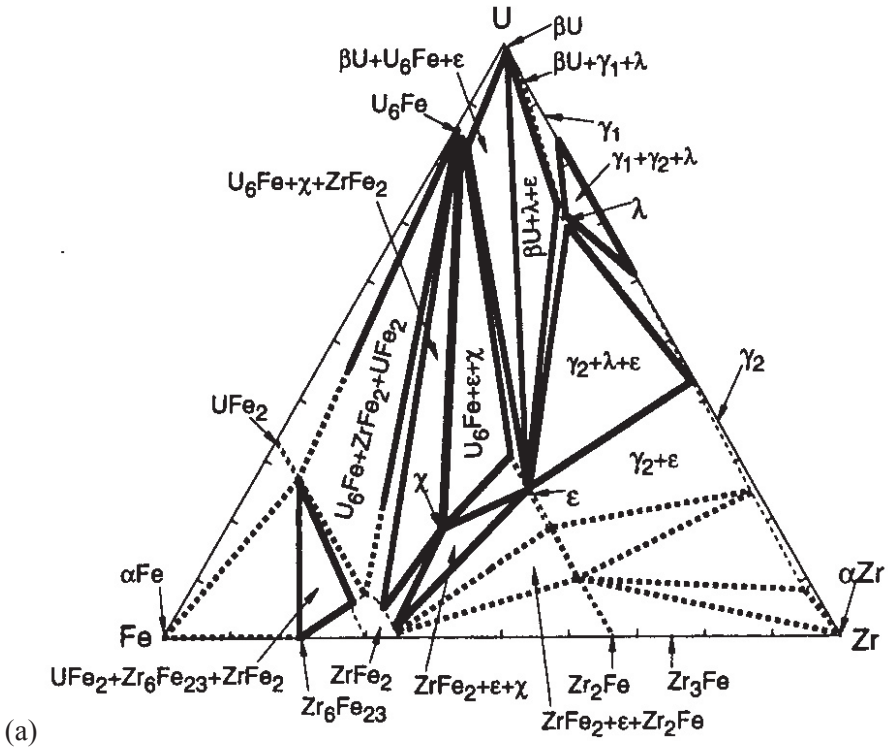
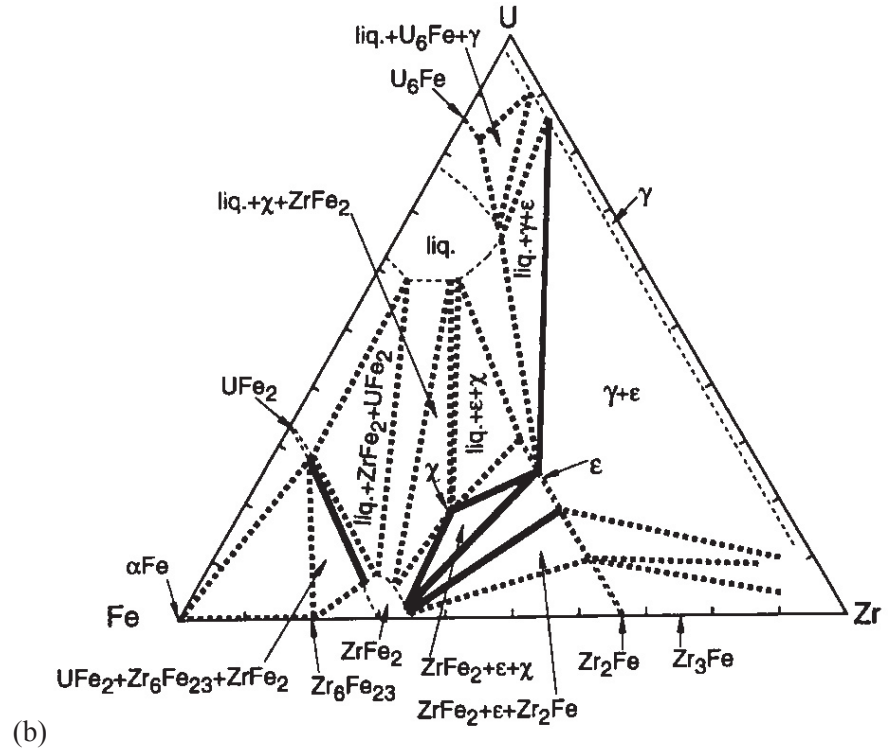


Fig. 11. U-Fe phase diagram [3].



(a)



(b)

Figure 12. Equilibrium phase diagrams at (a) 700 and (b) 800°C for the U-Zr-Fe system [4].

For the diffusion couple annealed at 800°C for 1 hour, the result was simlar to what was observed for the 730°C couple. No evidence of interaction between the TRIGA fuel and the cladding could be found. One difference observed was in the microstructure of the TRIGA fuel. Overall, more of the U-rich phase

was observed compared to what was seen in the 730°C couple (compare Figures 6 and 8). This could be due to the fact that once TRIGA fuel is exposed to temperatures above 760°C, there is a tendency for the material to experience some hydrogen dissociation [2]. This would especially be the case if the material were heated under vacuum. Also, the dehydriding is dramatically influenced by presence of surface oxide or nitride films, which are negligible for well-polished samples. The hydrogen release rate depends on the specific diffusion rate of the oxide and the internal hydrogen pressure (the equilibrium dissociation pressure), both of which increase with sample temperature as well as oxide thickness and morphology.The result of this dehydridation procsess could be an increase in the amount U-bearing phase present in the sample after some of the zirconium-hydride had dissociated during heat treatment. However, even in the case in which more U-bearing phase was in contact with the cladding, no evidence of fuel/cladding interaction could be observed.

Results for other controlled diffusion-couple experiments between as-fabricated TRIGA fuel and Type 304SS are not available in the literature for comparison. However, observations have been reported for interactions between Inconel-600 (an alloy that contains, in wt.%, 72 Ni, 14-17 Cr, and 6-10 Fe) thermocouple sheath material and TRIGA fuel [2]. It was reported that no thermocouple/fuel interaction was observed for samples heated up to 1,000°C, but at 1050°C localized melting could be observed where an Inconel-sheathed thermocouple was in direct contact with the fuel. Based on the fact that lower-temperature eutectics exist between U and major cladding constituents (U-Ni: 740°C, U-Fe: 725°C, and U-Cr: 859°C), it would be expected that melting should occur at a temperature lower than 1050°C, based on the mentioned binary phase diagrams. This suggests that for TRIGA fuel comprised of U, Zr, and H, any phases that may form due to interdiffusion with cladding constituents could be higher-melting, and there may be a significant influence of impurity-containing layers on the suface of the fuel or cladding that may impede interaction. The results of the current diffusion study can neither confirm or refute the idea that U-Zr-H-Fe-Cr-Ni phases are higher-melting since no evidence of interdiffusion was observed in the 700 and 800°C couples that resulted in the development of phases. The current study demonstrates the sluggishness of interactions when well-polished TRIGA fuel and Type 304SS cladding are in contact up to a temperature of 800°C for one hour.

The ultimate goal of performing work of the kind reported here is to shed light on when phases that form due to interdiffusion of fuel and cladding constituents that may become liquid at relatively high tempratures in irradiated fuel. If relatively high temperatures were to be reached for fresh fuel put into a reactor, then the results of the current diffusion-couple work could be applied, remembering that there is, in general, a gap between the fuel and cladding when the fuel is first fabricated. However, in order to develop understanding of how more highly irradiated fuel may perform at higher temperature, diffusion work must be performed using irradiated fuel. It has been suggested in Reference 5 that when irradiated TRIGA fuel with 45 wt.% U and Incoloy-800 cladding was heated to complete melting, there were indications (in the form of a fission-gas release) that melting at the fuel/cladding had occurred around 700°C. Further work where actual irradiated fuel is heated and examined will be required to develop better understanding of how irradiated TRIGA fuel, in the presence of fission products, interacts with irradiated Type 304SS to form phases and what the melting temperature is for these phases.

5. CONCLUSIONS

Based on the result of the SEM characterization of TRIGA fuel/Type 304 SS diffusion couples annealed for one hour at 730 and 800°C, the following conclusion can be drawn: for as-fabricated TRIGA fuel that has been polished to significantly remove any oxide layers put in intimate contact with polished Type 304SS, no diffusional interactions at the SEM scale can be observed when heated for one hour at a temperature up to 800°C. This is true even for a TRIGA fuel for which there is evidence that Zr-H dissociation may have occurred to expose more of the U-bearing phase at the TRIGA fuel/Type 304SS cladding interface.

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