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Post-irradiation Examination Status for the BSU-269 Project

August 2022

Fabiola Cappia, William A. Hanson, and Collin J. Knight



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Idaho National Laboratory Characterization and Advanced PIE Division Idaho Falls, Idaho 83415

http://www.inl.gov

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ACRONYMS

- ATR Advanced Test Reactor
- CCI Fuel-cladding chemical interaction
- DOE U.S. Department of Energy
- EDS energy dispersive X-ray spectroscopy
- LWR Light Water Reactor
- SEM scanning electron microscopy

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1. INTRODUCTION

High-density uranium inter-metallic uranium silicide (U₃Si₂) was initially considered as a potential advanced nuclear fuel for light water reactor (LWR) systems due to several advantages it demonstrated in thermophysical properties, chemical, and irradiation stability [1]. In addition to understanding the basic thermophysical and mechanical properties of U₃Si₂ under LWR irradiation conditions, it is important to assess its performance with advanced claddings such as FeCrAl, SiC, or conventional Zr-based claddings. Several aspects of the fuel cycle and fuel performance are being evaluated for U₃Si₂ coupled with different types of claddings [2-4]. To thoroughly characterize the U₃Si₂-cladding system, their chemical compatibilities and the potential for fuel-cladding chemical interaction (FCCI) must be addressed. Interdiffusion studies have been performed between U₃Si₂/Zry-4 and U₃Si₂/FeCrAl using diffusion couples. For the U₃Si₂/Zry-4 system, ZrSi₂ was the primary interdiffusion product observed at 800°C in addition to other secondary phases from the Zr alloving elements such as Fe and Cr. Low-melting point U_6 Fe was also observed at 1000°C [5]. For the U_3 Si₂/FeCrAl system, at temperatures above 500°C, chemical diffusion Fe-U and Fe-U-Si was observed, increasing with rising temperatures [6]. Although information from these non-irradiated tests is a fundamental first assessment, follow-up with focused irradiation tests is necessary. Irradiation can significantly alter the structure and evolution of the FCCI layers compared to the non-irradiated conditions.

This series of irradiation tests was specifically designed to focus on the assessment of FCCI between U_3Si_2 and FeCrAl, SiC, and Zry-4 claddings. The first capsule was discharged from the Advanced Test Reactor (ATR) and received at the hotcells in October 2021. The present report summarizes the status of post-irradiation examination (PIE) performed to date.

2. MATERIALS AND EXPERIMENTAL TECHNIQUES

The experiment was composed of a capsule containing a rodlet. The rodlet contained five diffusion-couple-style specimens. Each specimen consisted of a U_3Si_2 pellet, two discs of candidate-cladding material and two gadolinium foils all enclosed in a molybdenum cup. Two of the five specimens had Zry-4 discs, two SiC discs, and one FeCrAl discs. In the case of the SiC cladding discs, a ceramic ring was used to center the SiC discs over the uranium silicide pellet.

Visual examinations were conducted through a window on the capsule and rodlet. After the visual examinations, the capsule was imaged using neutron radiography to inspect the status of the Mo cups before performing capsule disassembly. Once the rodlet was extracted from the capsule, the rodlet was cut to retrieve the Mo cups then prepared for metallography and advanced PIE.

Each cup was mounted longitudinally in a stainless-steel mount and ground down to the mid plane exposing the discs, then backpotted, and re-gound. After grinding, the samples were polished with diamond suspensions to a 1 μ m finish. Metallography was conducted in the metallography box using a Leica DMi8 microscope.

3. PIE RESULTS AND DISCUSSION

Visual examination of the capsule was performed upon receipt after unloading it from the GE-100 cask. The capsule was placed on the visual examination stand and rotated to inspect the entire circumference. Figure 1a through Figure 1c shows no visible bulging or damage to the capsule which indicates no major issues. Figure 2 shows the radiography of the capsule from which the five Mo cups can be seen. From the radiograph, the cups appear intact and likely in good condition. After the capsule was disassembled the rodlet was inspected and no damage was observed (Figure 3).

After disassembly, each H-cup was mounted and ground through mid-plane to show the U3Si2 and cladding interfaces. Low-magnification overviews of the mounts are shown in Figure 4 through Figure 6. The mounts show ethanol residuals and the U3Si2 phase shows extensive pull-out (for example, see the white arrows in Figure 4). More refined sample preparation will be performed at the Irradiated Materials and Characterization Laboratory at a later stage. This preliminary investigation was performed to provide a first insight on the extent, if any, of the FCCI in each capsule. In all mounts, already at low magnification, a gap between the fuel and the cladding discs was visible, showing that there was no permanent chemical bonding between the two materials.

Higher magnification images were acquired to provide more details of the interfaces. A representative micrograph of the U3Si2/Zry-4 interface is shown in Figure 7. The gaps extended along all the interfaces, both top and bottom, in both capsules. No FCCI above the resolution limit of the optical microscope (generally $\sim 1 \mu m$) is visible, but only scanning electron microscopy (SEM) and energy dispersive x-ray spectroscopy (EDS) will confirm this preliminary observation.

The irregular surface of the SiC discs in the two H-cups shown in Figure 5 could point to a loss of integrity of the area that might have pulled out during sample preparation. A large gap remains in most of the interfaces, but signs of potential interaction could be observed towards the edges of the SiC discs in contact with the U3Si2, as highlighted by Figure 8. It cannot be excluded that these are localized areas of oxidation of the U3Si2 fuel, since it is known that these first-generation samples contained oxygen impurities that caused grey precipitates in the U3Si2 matrix, which looked exactly like the ones observed here [1,7]. Figure 9 shows an example of potential localized interaction with the FeCrAl discs. In the U3Si2 several UO2 precipitates can also be observed making it unclear if FCCI is beginning to develop or the layer is a result of pre-irradiation oxidation. SEM and EDS will be needed to determine the composition of these layers.





(b)



Figure 1. (a)-(c) Visual examinations of capsule BWCI-C1 at different azimuthal angles.



Figure 2. Neutron radiography of the capsule.



Figure 3. (a)-(b) Visual examinations of the BWCI-R1 rodlet.



(a)



Figure 4. (a)-(b) Low-magnification overview of the two U3Si2/Zry-4 specimens.



(a)



Figure 5. (a)-(b) Low-magnification overview of the two U3Si2/SiC specimens.



Figure 6. Low-magnification overview of the U3Si2/FeCrAl specimen.



Figure 7. Representative high magnification image of the U3Si2/Zry-4 interface.



Figure 8. Examples of (a) bottom and (b) top FCCI in one of the U3Si2/SiC specimens.



Figure 9. Example of a location on the U3Si2/FeCrAl interface where potential FCCI was observed.

4. SUMMARY

The first of two capsules dedicated to the study of FCCI between U3Si2 and several types of cladding have started PIE. Non-destructive examinations have been conducted on both the capsule and the rodlet before starting preparation of the diffusion-couple style specimens for metallography. The optical microscopy analyses did not show extensive interaction at this burnup level for the U3Si2/Zry-4 specimens, at least for the resolution accessible by optical microscopy. Localized, spotty interaction might have occurred between U3Si2 and SiC as well as U3Si2 and FeCrAl, but the morphological information that can be accessed with the optical microscope are not enough to undoubtly confirm that it is FCCI and not oxidation. Further advanced microscopy will elucidate more clearly if chemical interaction has started to occur in these cups and if interdiffusion has occurred in the U3Si2/Zry-4 at a scale not accessible by the optical analyses.

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