Destructive Examination of a FeCrAl-UO_2 Irradiation Test

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ABSTRACT

Several destructive postirradiation examinations were performed on an irradiation specimen that coupled an early iron-chromium-aluminum (FeCrAl) candidate alloy cladding with UO_2 pellets. This irradiation test was designed to investigate the early life performance and compatibility of the FeCrAl-UO2 cladding-fuel system under prototypic light water reactor neutronic conditions. Additionally, these tests were expected to provide neutron irradiated samples for severe accident testing of this fuel system. The irradiation studied in this work was part of the ATF-1 series of drop-in style irradiations performed in the Idaho National Laboratory Advanced Test Reactor. The rodlet studied in this work is one of three similar rodlets irradiated in ATF-1 that had approximately 7.6 cm of cladding machined from a wrought FeCrAl alloy and fueled with a 6.1 cm stack of UO_2 pellets. The outer diameter of the cladding was ~0.94cm and the inner diameter was ~0.83 cm. After irradiation and non-destructive examination in Idaho this rodlet was shipped to the Oak Ridge National Laboratory hot-cells for further examination. The irradiated rodlet was sectioned into several samples for microstructural, micromechanical, and severe accident testing. During sectioning, it was noted that the fuel was not firmly bonded to the cladding and could be readily removed from small cladding slices. Microstructural characterization of fuel cross sections also revealed no significant interaction between the fuel and the cladding. Samples were also prepared for microhardness testing. To prepare for high temperature oxidation testing, the fuel was dissolved from segments of the cladding. High temperature oxidation testing of cladding segments was performed at 1200°C and 1300°C in a steam environment. Comparisons between the oxidation of this FeCrAl alloy in its neutron irradiated state, as-fabricated state, and the oxidation of Zircaloy-2 are made. The oxidation testing will be followed by ring compression testing to evaluate ductility. The microstructure of the samples after oxidation and ring compression testing will also be analyzed.

1. INTRODUCTION

Enhanced accident-tolerant fuels (ATFs) are being developed for deployment in commercial light water reactors through a collaboration of the Department of Energy Office of Nuclear Energy (DOE-NE) Advanced Fuels Campaign (AFC) and US fuel vendors. The first fueled irradiation experiments on ATF concepts, referred to as ATF-1, began at the Idaho National Laboratory (INL) Advanced Test Reactor (ATR) in 2014[1–3]. These irradiations were meant to screen many different concepts to ensure fuel–cladding compatibility and provide irradiated material for future testing. One of the ATF candidate materials for near to mid-term deployment is FeCrAl alloys for cladding[4–7]. The irradiation included three FeCrAl-UO_2 drop-in rodlets fabricated at Oak Ridge National Laboratory (ORNL) targeting burn-up values of 10 GWd/MT, 30 GWd/MT, and 50 GWd/MT. The lowest burn-up capsule (ATF-18) completed irradiation and initial post-irradiation examination (PIE) at INL [8]. The capsule was shipped to ORNL's Irradiated Fuels Examination Laboratory (IFEL) hot cell facility for additional PIE in the Severe Accident Test Station (SATS) [9,10]. This report describes destructive examinations performed on this rodlet. The microstructure of the fuel cladding interface was examined with optical microscopy. The response of the irradiated cladding to high-temperature oxidation was also tested using small ring specimens harvested from the irradiated rodlet. Pre- and post-testing weight measurements were taken to determine the weight gain. Cross-section images of the specimens were taken to measure the oxidation layer.

2. FeCrAl ATR IRRADIATION SUMMARY

ORNL has three loss-of-coolant-accident (LOCA) capsules for irradiation within the ATR: ATF-17, ATF-18 and ATF-19. The ATF-18 capsule contained an early variation of a FeCrAl alloy called C35MN, which is Fe-13Cr-5Al with minor additions of Mo and Nb [4]. The cladding was filled with UO_2 pellets and irradiated to a 10.4 GWd/mtU burnup [11]. The rodlet had approximately 7.6 cm of cladding machined from a wrought FeCrAl alloy and fueled with a 6.1 cm stack of UO_2 pellets. The outer diameter of the cladding was ~0.94cm and the inner diameter was ~0.83 cm. The tubes used in this irradiation were created by gun drilling rather than extrusion which is the typical method of fabricating cladding tubing. The gun drilling created a non-prototypic microstructure in the tube, but this difference is inconsequential in studying fuel-cladding interaction. The gun-drilled nature of the tube also raised suspicions that the tube itself might be vulnerable to brittle microcracking. It should be noted that the ATF-1 experiment was double encapsulated with a stainless steel "capsule" that surrounded the rodlet which consisted of a sealed cladding tube and fuel pellets. Before the capsule was shipped to ORNL, non-destructive PIE was performed at INL [8]. The capsule was checked for fission gas release before being disassembled, and fission gas was detected in the capsule. The fission gas release for the fuel was found to be 0.35%, which is in line with expectations from the Vitanza curve for irradiated UO_2 fuel [12]. No obvious flaws were detected in visual exams or neutron radiography, and the exact nature of the rodlet failure could not be determined. There was some measurable change in the cladding diameter. The cladding diameter measured between 9.39 and 9.4 mm which is greater than the maximum reported diameter of 9.37 mm in the as-built documentation. This indicates there may be some irradiation or thermally induced cladding strain. However, 9.39 and 9.4 mm is within the tolerance of the drawings for the tube.

3. STEAM OXIDATION OF ATF-18 SPECIMENS

After irradiation and nondestructive examination at INL, the ATF-18 rodlet was shipped to the ORNL hot cells. The irradiated rodlet was sectioned into several samples for high-temperature steam oxidation tests. During sectioning, it was noted that the fuel was not firmly bonded to the cladding and could be readily removed from small cladding slices. This was expected for a drop-in style irradiation in ATR where there is no external pressure to drive the cladding creep-down that is normally seen in light water reactors. To prepare for high-temperature oxidation testing, the fuel was dissolved from segments of the cladding. High-

temperature stem oxidation tests of the ATF-18 rodlet were performed in the SATS at the IFEL. SATS consists of two modules: one for integral testing of LOCA scenarios and the other with a high-temperature furnace for testing fuel segments [9,13]. The LOCA furnace has been successfully used in LOCA fragmentation tests on high-burnup fuels [10]. The tests of irradiated FeCrAl C35MN specimens in this report were conducted with the high-temperature furnace. Figure 1 shows the alumina specimen holder used in this work.

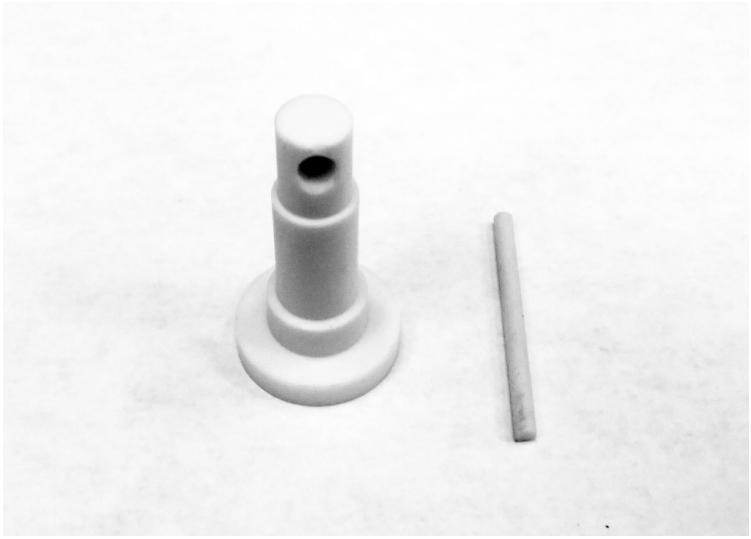


Figure 1. Alumina Specimen holder used for the SATS high-temperature steam oxidation tests.

3.1 Steam Oxidation Tests with Unirradiated ATF-18 Surrogates

Before the in-cell oxidation tests of the irradiated ATF-18 specimen, out-of-cell benchmark tests were conducted on C35MN surrogates (same alloy, heat, and fabrication procedure) to provide baseline data for the in-cell tests. These out-of-cell tests with unirradiated C35MN surrogates parallel the thermal history of the in-cell tests with irradiated AFT-18 specimens, particularly to see how the FeCrAl alloy C35MN tubing would perform at temperatures ≥1200°C. All tests were performed at atmospheric pressure with the tube in the sample holder shown in Figure 1. The surrogate specimens were approximately 9.4 mm in outer diameter (OD) and 12.5 mm long.

The specimen was first ramped from room temperature to 600°C under an argon atmosphere at a rate of 20°C/min. Following this test segment, the argon supply was shut off, steam was supplied to the test section, and the temperature was ramped to 1200°C at 7.5°C/min. The sample was held at 1200°C for one min and then slowly ramped to 1300–1400°C at 1.82°C/min to avoid temperature overshoot. Steam was supplied to the test section by injecting water into a preheat furnace at ≈0.056 g/s (200 mL/h). The tests and the temperature at which the tests were stopped are illustrated in Figure 2. Following the steam oxidation runs, the specimens were weighed.

To determine an accurate post-test specimen weight, it is important that the specimen be free of moisture. In this work, the specimens were dried in stagnant air for at least 2 hours. The drying time was verified by weight measurements: the specimen weight continued to decrease during the drying process until it reached a minimum and held at that minimum. The post-test specimen weight was measured to the nearest 0.1 mg using a calibrated balance. The weight gain was determined by subtracting the pretest weight from the post-test weight and normalizing this value to the steam-exposed surface area of the specimen. The surface area of the specimen was calculated by including the inner tube surface, the outer tube surface, and both ends.

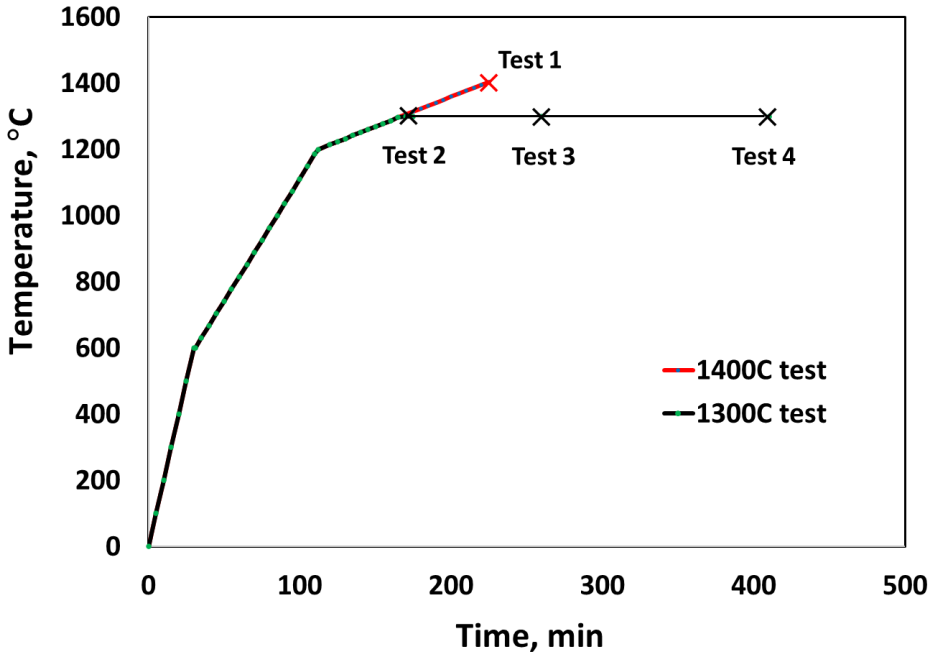


Figure 2. Heating segment illustration for out-of-cell high-temperature steam oxidation tests.

Table 1 lists the test conditions and weight gain results from the steam oxidation tests of FeCrAl alloy C35MN tubing specimens. Specimens melted as soon as the temperature reached 1400°C. However, the C35MN was very stable at 1300°C. Post-test images of these tests are provided in Figure 3. It is generally believed that the oxidation pickup of the FeCrAl alloy is characterized by a parabolic rate law. For the oxidation tests at 1300°C, the ramp-up and cooling durations were the same. Therefore, the oxidation at the hold temperature can be considered an isothermal process. and the oxidation rate can be considered a constant for the multi-oxidation tests at a target temperature.

Figure 5 shows oxidation parabolic curves of the unirradiated C35MN specimens oxidized at 1300°C. The test data points vs. the isothermal oxidation time are in good agreement with a linear fit to the experimental data, which indicates good control of isothermal oxidation temperature. For isothermal oxidation at a target temperature of 1300°C, the weight-gain coefficient can be determined to be 3.8×10^{-9} $(mg/cm)^2/s$ by the slope of the fits in Figure 4, which is about two orders of magnitude lower than the coefficient of zirconium alloys.

Table 1. Test conditions and weight gain values for unirradiated ATF-18 surrogate C35MN tubing specimen oxidized in steam.

Test ID	Max. temperature (°C)	Time at max. temperature (min)	Measured weight gain (mg/cm^2)	Measured OD oxide µm	Measured ID oxide µm	Sample surface
1	1400	1	N/A	N/A	N/A	Melted
2	1300	1	0.336	2.1 ± 0.2	2.0 ± 0.1	Lustrous black
3	1300	90	0.586	3.7 ± 0.2	3.5 ± 0.4	Lustrous black
4	1300	240	0.762	5.4 ± 0.3	5.0 ± 0.5	Lustrous black

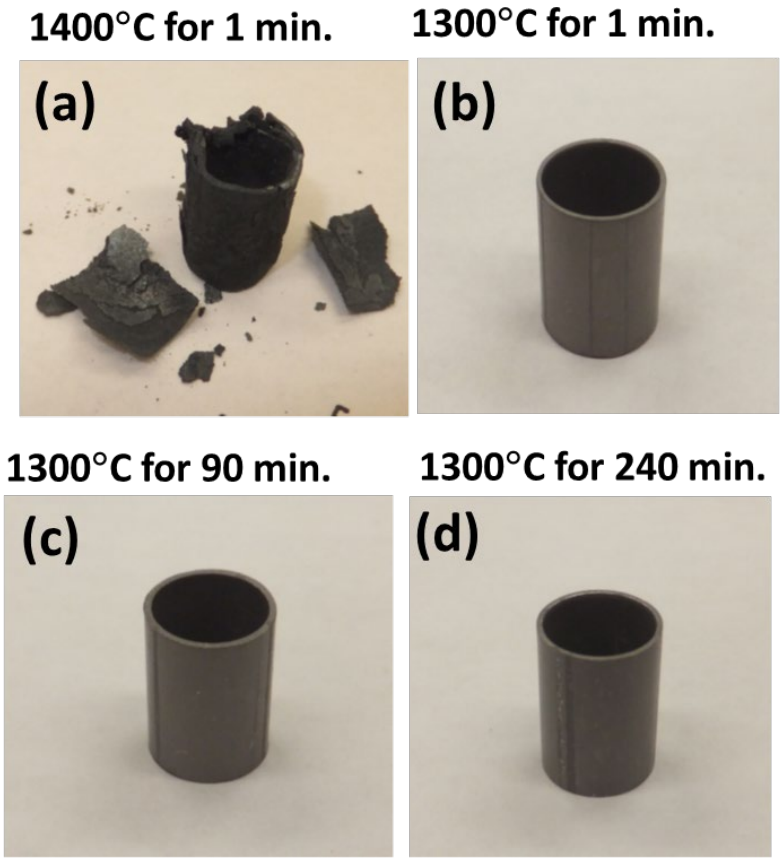


Figure 3. Post-test images of high-temperature unirradiated steam-oxidized FeCrAl alloy C35MN tubing specimens.

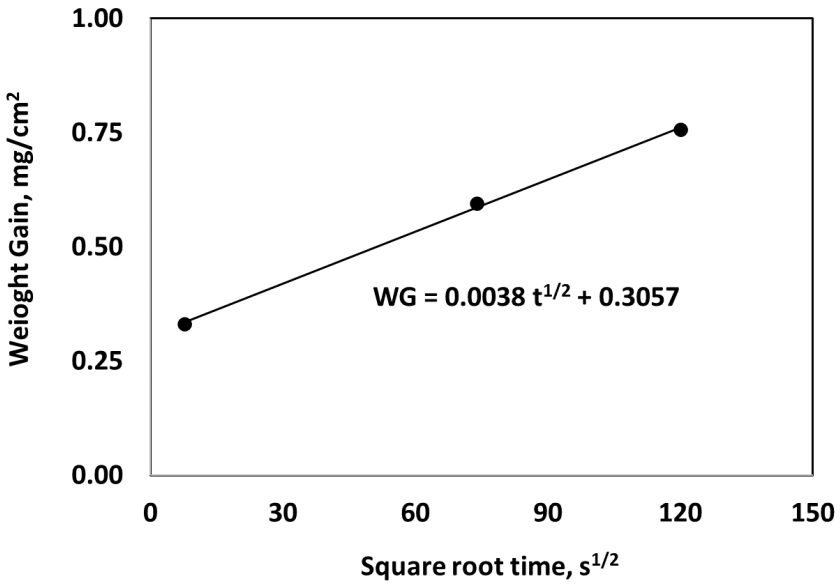


Figure 4. Steam oxidation parabolic curve of unirradiated FeCrAl C35MN at 1300°C.

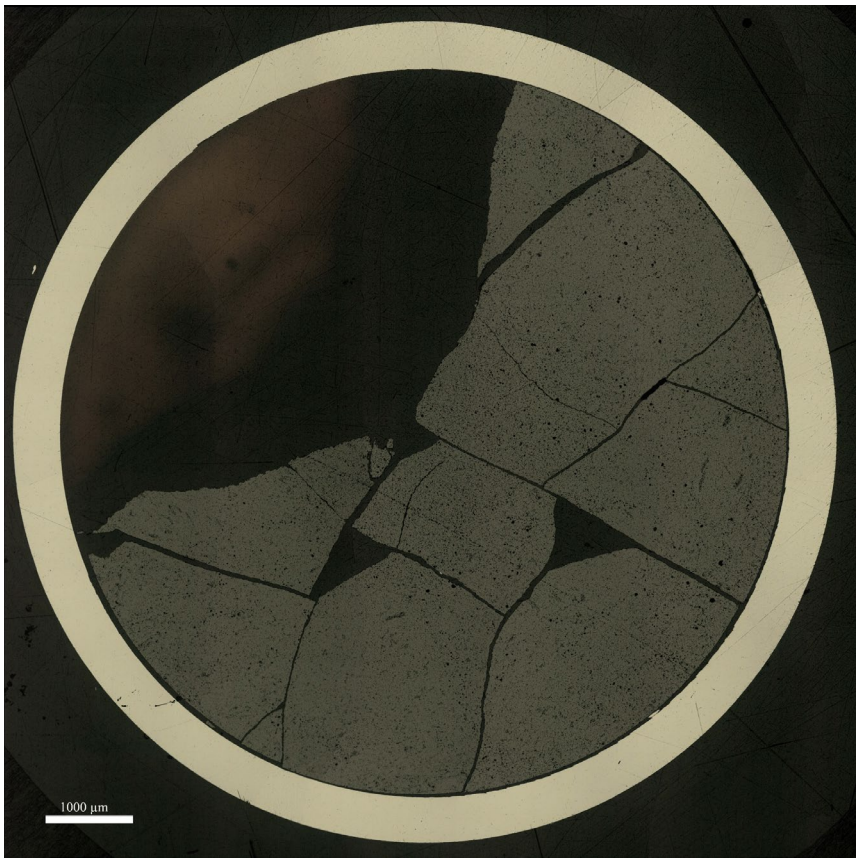
3.2 Steam Oxidation Tests with Irradiated ATF-18 Specimens

Pretest characterization was performed on the ATF-18 rodlet to determine the fuel, fuel-cladding bond and cladding behavior. Figure 5a shows a low-magnification image of the fuel morphology. The fuel partially fell out during sample preparation. A fuel-cladding bond was not found, which is expected for a low-burnup rod such as the ATF-18. Figure 5b is a higher-magnification image showing a gap between the cladding and fuel.

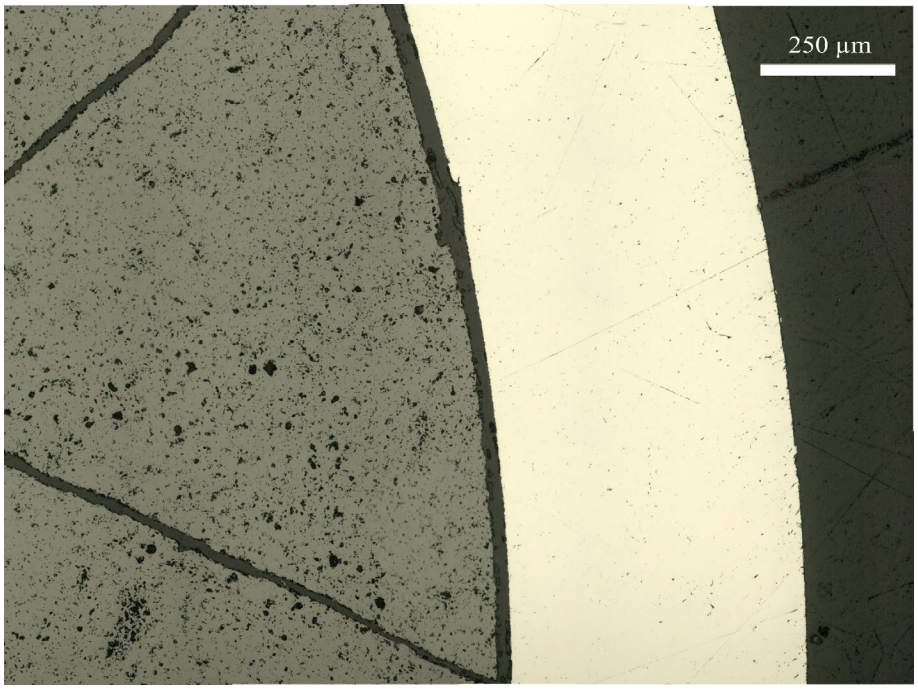
High-temperature steam oxidation tests of the irradiated ATF-18 specimens were performed with the in-cell SATS. To get a precise weight gain measurement, the uranium oxide fuel in cladding segments was dissolved in nitric acid at room temperature, then the cladding segments were rinsed in water and dried before high-temperature oxidation testing. The average OD of the ATF-18 rodlet was approximately 9.40 mm. The sample length for the high-temperature oxidation tests was between 4.57 and 6.96 mm.

All in-cell tests of irradiated ATF-18 specimens were conducted with the standard sample holder (see Figure 1) under the same test conditions as used for unirradiated out-cell surrogate specimen tests. The specimen was first ramped from room temperature to 600°C under an argon atmosphere at a rate of 20°C/min. Following this test segment, the argon supply was shut off, steam was supplied to the test section, and the temperature was ramped to 1200°C at 7.5°C/min. The sample was either held at 1200°C or slowly ramped to 1300°C at 1.82°C/min. The tests and temperatures at which the in-cell tests were stopped are illustrated in Figure 6. Figure 7 is the temperature history of the in-cell test ATF-18G2.

Although unirradiated specimens performed very well at 1300°C, the irradiated specimens started melting as soon as the temperature reached 1300°C. However, the ATF-18 specimen was very stable at 1200°C. In addition, the oxygen pickup at 1200°C was extremely low, under the limit of the scale for weight gain measurements. Therefore, no weight gain data were reported for the ATF-18 specimens steam oxidized in the in-cell SATS. Post-test images of these tests at 1200° and 1300°C are provided in Figure 8 and Figure 9. Table 2 lists the test conditions and results of the steam oxidation tests of irradiated ATF-18 tubing specimens.



(a)



(b)

Figure 5. (a) Fuel morphology for the ATF-18 rodlet, (b) high-magnification image showing a gap between the fuel- and cladding.

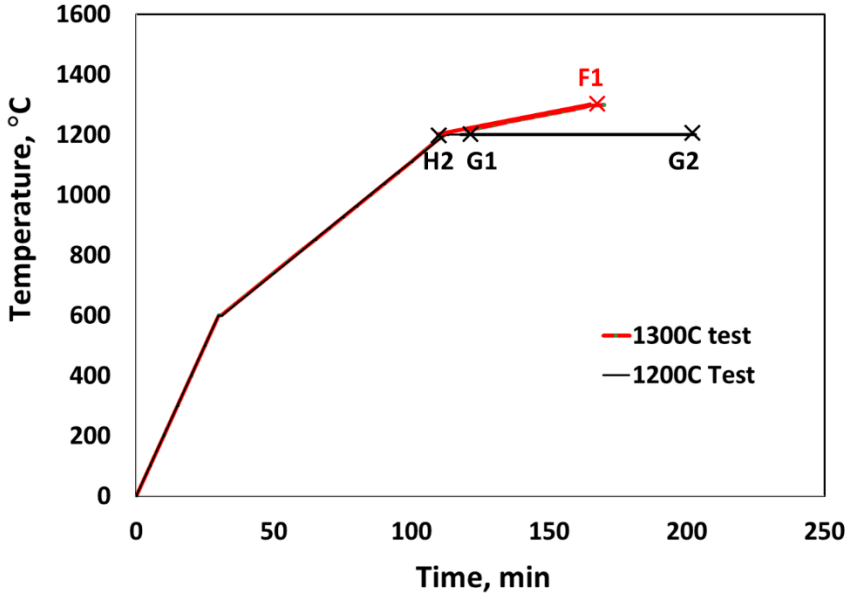


Figure 6. Heating segment illustration for in-cell high-temperature steam oxidation tests.

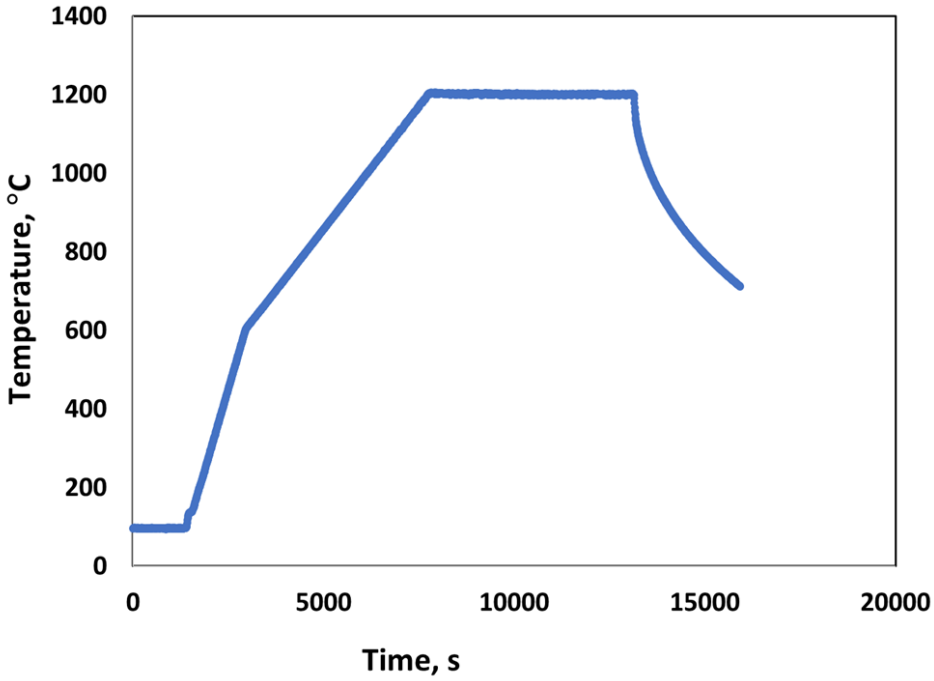


Figure 7. Temperature history of the in-cell SATS test of irradiated ATF-18G2.

Figure 8. Post-test images of irradiated ATF-18 specimens oxidized at 1200°C.

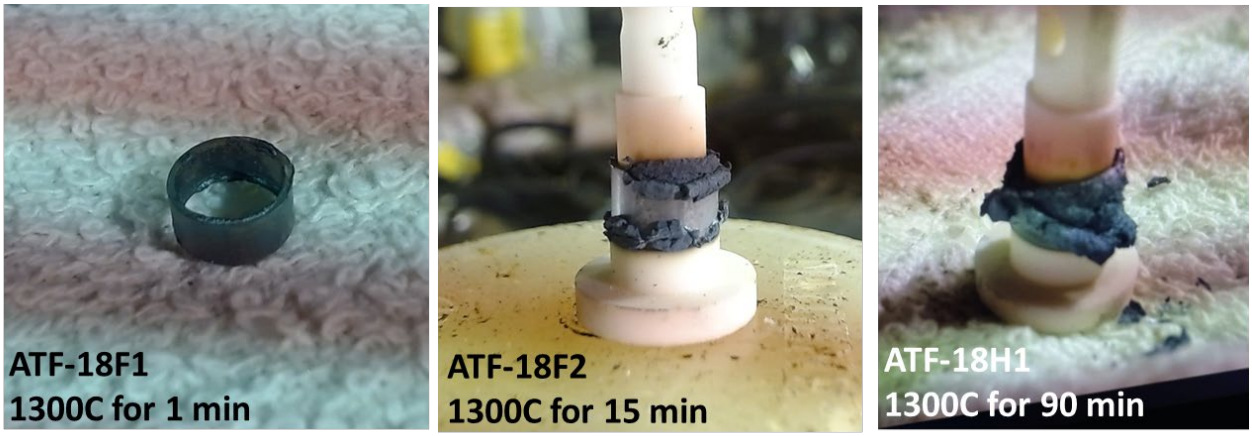


Figure 9. Post-test images of irradiated ATF-18 specimens oxidized at 1300°C.

Table 2. Test conditions and results for in-cell SATS tests of irradiated ATF-18 steam specimens.

Test ID	Max. temperature (°C)	Time at max. temperature (min)	Calculated equivalent cladding reacted for Zry-4 (%)	Sample surface
H2	1200	1	16	Lustrous black
G1	1200	15	35	Lustrous black
G2	1200	90	79	Lustrous black
F1	1300	1	–	Partially melted
F2	1300	15	–	Melted
E	1300	90	–	Melted

The microstructures of the ATF-18 oxidation specimens were examined using optical microscopy (Figure 10). The images were taken digitally using a Leica microscope. The resolution of the Leica microscope at maximum magnification was approximately 1 μm. Figure 10 shows low-magnification images for ATF-18 specimens after the high-temperature oxidation in the SATS. High magnification micrographs at eight azimuthal locations were taken every 45° of the cross-sectioned samples to observe oxidation formation. The oxide layer thickness was at the limit of resolution for the microscope for the 1200°C samples. The oxide thickness for the G2 sample was approximately 2.5 μm on the outer diameter.

Enhanced oxidation was observed on the inner surface at several areas of the ATF-18F1 (see Figure 10d), which was oxidized at 1300°C for 1 min. Similar reactions likely occurred with the F1 and F2 samples. This phenomenon was not observed for unirradiated ATF-18 surrogate specimens oxidized in steam at 1300°C up to 240 min. Unirradiated samples of C35MN were run in the in-cell SATS system after receiving a similar acid wash and rinse sample preparation treatment, and the unirradiated material matched the out-of-cell behavior reported in Section 3.1. Several independent thermocouple tests were also performed to confirm proper function of the SATS high temperature oxidation furnace. For the irradiated specimens oxidized at 1200°C, the enhanced oxidation was not found. More study of irradiated FeCrAl alloys is needed to understand the mechanism of enhanced inner diameter oxidation at 1300°C.

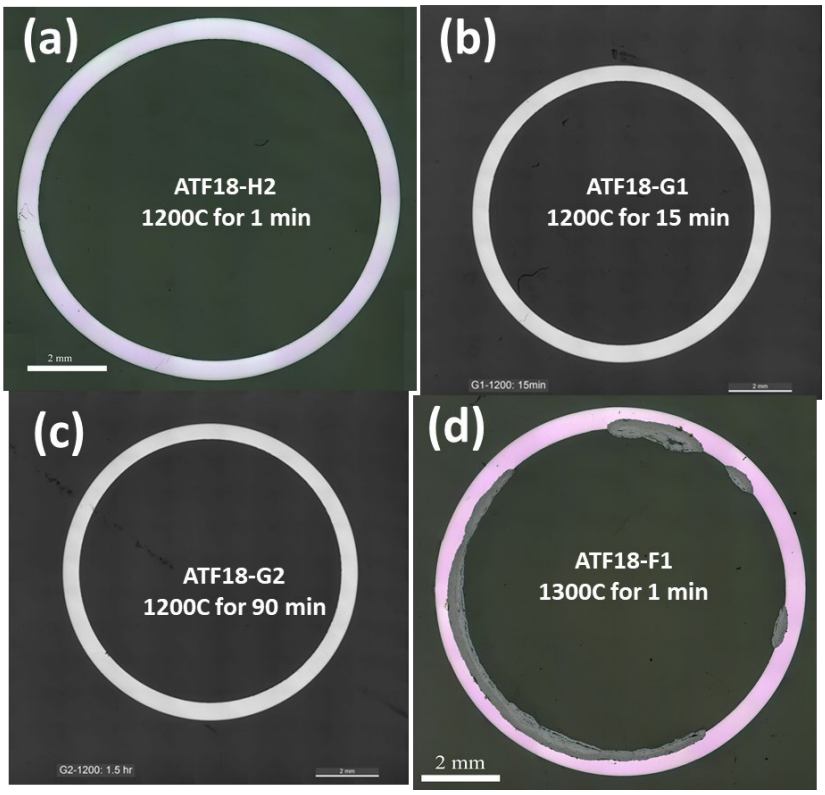


Figure 10. Low-magnification images of the irradiated ATF-18 specimens after the high-temperature oxidation in the SATS. Note the enhanced oxidation on the inside of the 1300C specimen (d). The pink color is an artifact.

4. CONCLUSIONS

FeCrAl-UO_2 test capsules were fabricated at ORNL and irradiated at the ATR. Following irradiation, samples were sectioned from the irradiated rod ATF-18 for oxidation kinetics study to assess the candidate cladding high-temperature oxidation performance following irradiation. The high-temperature oxidation tests were conducted in the ORNL SATS at 1200 and 1300°C. Weight measurements were taken before and after oxidation testing. Cross-sections of the cladding were metallographically mounted and optical microscopy was performed. Measurements of the oxidation layer before and after high-temperature testing were collected. The results indicate the irradiated FeCrAl C35MN alloy provided good thermal stability up to 1200°C. The irradiated specimens started melting as soon as the temperature reached 1300°C, although the unirradiated C35MN surrogate specimens were very stable at 1300°C for 240 min. Enhanced oxidation was observed on the inner surface of the irradiated FeCrAl C35MN specimen oxidized at 1300C for 1 min. More study of irradiated FeCrAl alloys is needed to understand the mechanism of enhanced oxidation at 1300°C.

5. REFERENCES

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