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# FY–18 FCRD Milestone M3NT-18OR020206061
# Steam Oxidation, Burst and Critical Heat Flux Testing of FeCrAl Cladding in the SATS




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## Introduction

Since August 2011, Oak Ridge National Laboratory (ORNL) has been evaluating the steam oxidation resistance of a wide range of materials to identify a potential accident tolerant fuel (ATF) cladding [1-14]. The primary goal of an ATF cladding is to enhance safety margins in light water reactors (LWR) during beyond design basis accident scenarios (such as the nuclear accident at Fukushima Daiichi in March 2011) without requiring changes to the reactor design or fuel enrichment, or significantly affecting reactor economics during normal operation. The research has taken advantage of the Severe Accident Test Station (SATS) [5,15] for high temperature steam oxidation testing to 1700°C and the standard integral loss of coolant accident (LOCA) experiment [9]. An ATF cladding would significantly reduce the rate of heat and hydrogen generation in the core during a coolant-limited severe accident [16-18]. In the search for materials with steam oxidation rates that are 100× slower than current Zr-based alloys at ≥1200°C, FeCrAl alloys quickly became the focus of research at ORNL including alloy development [8], physical properties [9,10,12-14,19-23] and integral data [24-27] to populate models [28-32].

While the high temperature oxidation behavior of FeCrAl alloys has been studied for almost 60 years [33,34], it was typically studied isothermally to measure reaction rates and activation energies [7] and it was not studied in steam at such high temperatures above current advanced steam concepts of 760°C [35]. More recent experiments have focused on "ramp" testing of low-Cr Fe-(10-13wt.%)Cr-(5-6%)Al compositions [6,12] and have examined the effect of varying the ramp rate or other variations in the heating schedule [25-27]. Experiments are now focusing on the new commercial C26M FeCrAl tubing that was inserted into Plant Hatch in February 2018. This report summarizes recent work (1) to compare the steam oxidation behavior of C26M to prior FeCrAl tube heats in ramp testing, (2) to study additional loss of coolant accident (LOCA) burst testing results and (3) to report efforts to establish a new experiment to evaluate critical heat flux behavior.

## Steam oxidation behavior

Based on several years of studying the high temperature steam oxidation behavior of FeCrAl compositions attractive to the ATF application, a general understanding has been developed [7,8,10,12,14]. Recent work has focused on evaluations of tube specimens and a special holder has been used to expose the tube outer diameter to steam and catch debris should breakaway oxidation occur, Figure 1. Table 1 shows the chemistry of several FeCrAl tube compositions that

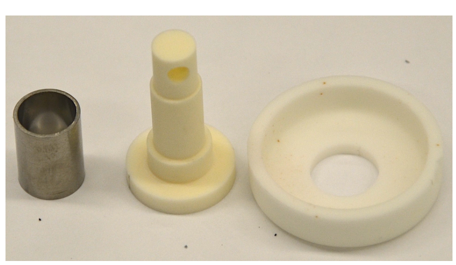

**Figure 1. Typical 9.5mm OD tube specimen, holder and catch pan for ramp testing [12].**

**Table 1. Chemical composition in weight % of relevant Fe-based alloys measured using inductively coupled plasma and combustion analyses.**

| Alloy | Fe | Cr | Al | Mo | Si | Y | Other |
|---|---|---|---|---|---|---|---|
| **B136Y** | 80.8 | 13.0 | 6.2 | <0.01 | <0.01 | 0.03 | 0.01 C, 0.001 S, 0.001 O |
| **C26M (1+2)** | bal. | 12.2 | 6.1 | 2.04 | 0.20 | 0.04 | <0.01 C,<0.005 S |
| **C26M (3rd)** | bal. | 11.9 | 6.2 | 1.98 | 0.20 | 0.03 | <0.01 C,<0.005 S |
| **APMT** | 69.2 | 21.1 | 5.0 | 2.8 | 0.59 | 0.25 | 0.2Hf,0.1Zr,<0.001 S |

were evaluated. Several different ramp schedules have been used for steam oxidation testing to match various accident or experimental test scenarios. All of the scenarios include heating to 600°C where the steam is introduced. One of the scenarios included heating to 1200°C at 20°C/min followed by a 50 min hold. The next ramp was at 11.1°C/min and Figure 2 shows the mass gain after heating to various temperatures. The results for B136Y and APMT were reported previously [12]. Both B136Y and C26M tube specimens could be heated to 1400°C followed by cooling to room temperature and show a small mass gain indicating a protective alumina scale was maintained under these conditions. Likewise, small mass gains were measured for both alloys after ramping to 1450°C. A C26M tube continued to show a small mass gain after ramping to 1475°C (Figure 2c) and an APMT tube specimen could be ramped to 1500°C and maintain an alumina scale [12]. In contrast, the B136Y tube specimens ramped to 1475°C and 1500°C showed high mass gains (i.e. breakaway oxidation) indicated that a protective alumina scale could not be formed at these temperatures. Instead, rapidly growing Fe-rich oxides completely consumed the tube specimens, Figure 2b. Higher temperature testing of C26M specimens is in progress. The current hypothesis is that reducing the Cr content from 21% to 12-13% (Table 1) reduces the maximum temperature where a protective alumina scale can form on FeCrAl in steam, particularly when the ramp rate is >2°C/min [25]. However, the protective behavior of C26M at 1475°C suggests that temperature debit is minimal for the new commercial tubing.

Several additional comparisons have been conducted between B136Y and C26M tube specimens with the mass changes summarized in Figure 3 for different peak temperatures and ramp rates. In

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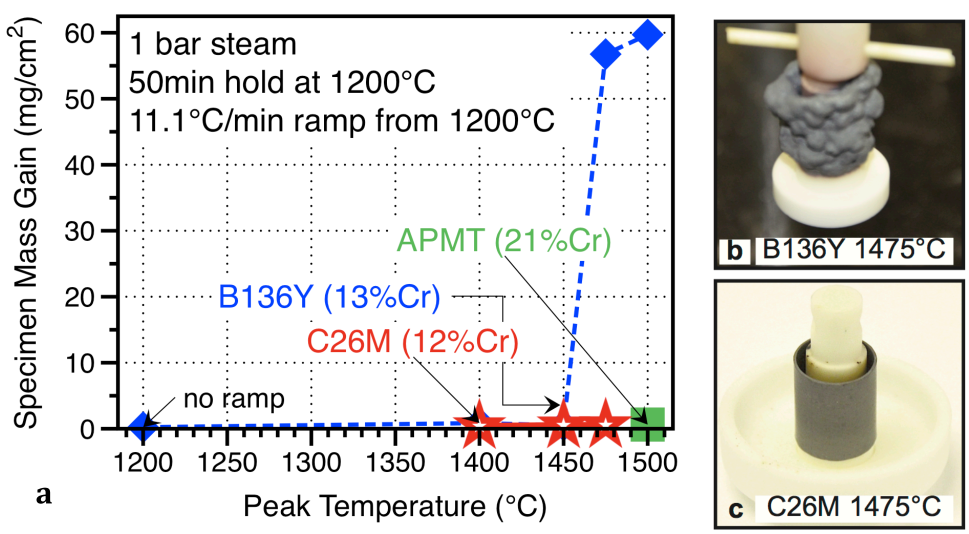


**Figure 2. (a) Specimen mass gain after holding for 50min at 1200°C followed by ramping to temperatures above 1200°C at 11.1°C/min, (b,c) images of 1475°C specimens**

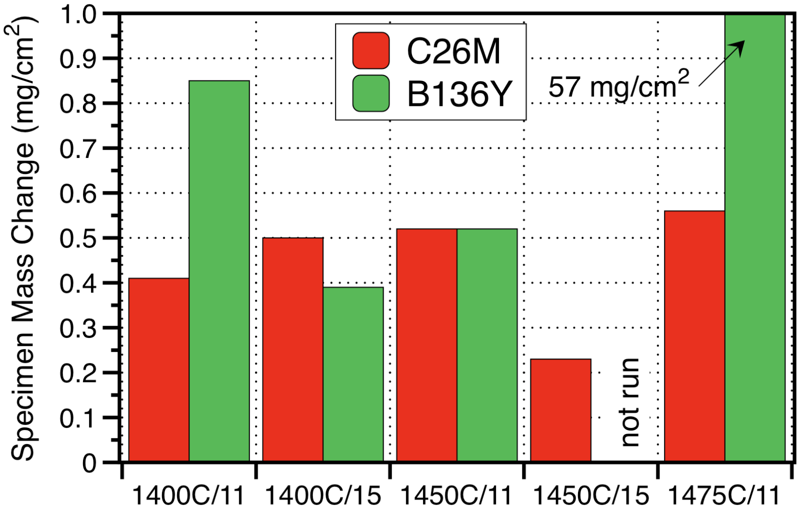


**Figure 3. Specimen mass gain for B136Y and C26M tube specimens after various exposures to 1400° and 1450°C with ramp rates of 11.1°C/min and 15°C/min.**

general, the results suggest that these two FeCrAl compositions behave similarly under these conditions with the small mass gains indicating the formation of a protective alumina scale. As noted in Figure 2, the biggest difference was for the ramp to 1475°C where the B136Y specimen could not maintain a protective alumina scale and the mass gain increased by 100 times. Characterization of the C26M reaction products is in progress.

**LOCA burst testing of commercial C26M FeCrAl tubing**

Previously, only limited burst testing was performed on FeCrAl tubing because of the limited amount of available tubing [9]. Now that more commercial tubing is available, additional burst testing has been conducted. The results for the first two batches of C26M material were reported previously [36]. The LOCA burst tests were conducted in the SATS on 30 cm long tube specimens that were filled with $ZrO_2$ pellets and internally pressurized to a fixed value followed by heating in steam to 1200°C at 5°C/s. The pressure was monitored in the tube during the experiment to determine the burst temperature. During cooling, water was injected into the system at 600°C. Figure 4 summarizes the prior results and the new results for the third batch of C26M tubing. The results are compared to results from the literature for Zr-based cladding and type 304 stainless steel [9,37,38]. As the temperature increased in the experiment and the material strength was reduced, Zr-based alloys typically ballooned (i.e. expanded due to creep) and eventually burst. Previous characterization showed that the stronger FeCrAl alloys tended to just burst without significant deformation. The expectation was that the C26M tubing would outperform the "B" series tubing (e.g. designated B135Y (13Cr-5Al) and B154Y (15Cr-4Al) in Figure 4 [9]) because of the higher tensile properties attributed to the Mo and Si additions [8], Table 1. As reported previously [36], the first batch of C26M specimens did not show improved burst temperatures compared to the 1$^{st}$ generation FeCrAl results and this was consistent with the low tensile properties reported for this batch of material, which were attributed to processing issues [39]. In the 2$^{nd}$ batch of C26M tubing, the processing issues were resolved, and the burst temperatures were consistently higher than the 1$^{st}$ generation "B" alloy results.

For the third batch of C26M tubing, several experiments were conducted at higher stress levels to examine the burst size. A few lower stress tests showed lower burst temperatures than expected.

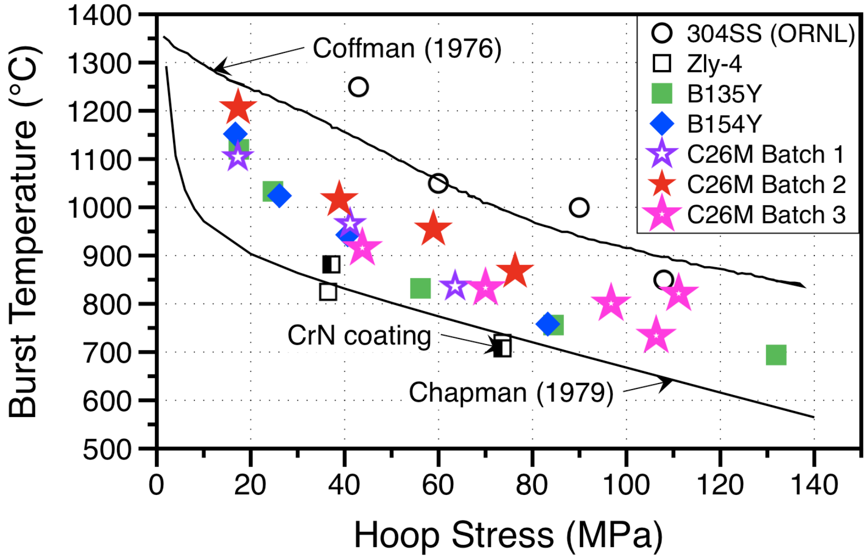


**Figure 4. Burst temperature as a function of engineering hoop stress for various cladding materials examined in the SATS (data points [9]) alongside empirical correlations (lines) from the literature for Zr-alloys [37] and 304SS [38]**

One source of error for this experiment was the measurement of the tube wall thickness, which was initially performed using calipers. To obtain more accurate values, several of the tubes from each batch will be sectioned metallographically to measure the average wall thickness and recalculate the stress values.

Figure 5 shows images of the burst tubes from four different stress levels shown in Figure 4. It is clear that above 97 MPa, the burst size increased dramatically. At 111 MPa, one of the $ZrO_2$ pellets that are surrogates for the fuel pellets was ejected. For the "B" material, a large burst occurred at

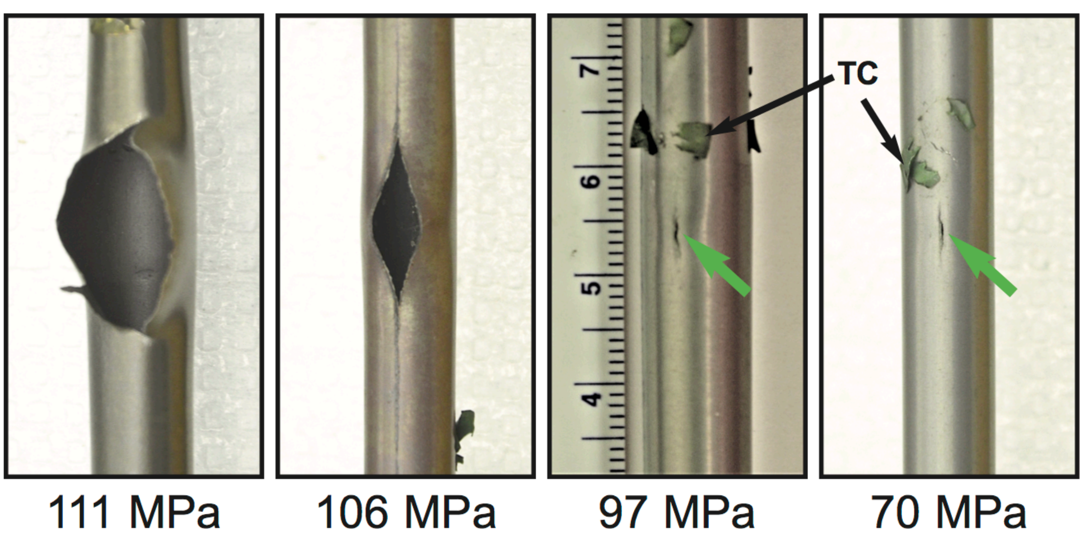


**Figure 5. Images of C26M burst at four different stress values. The small openings observed at lower stress values are marked with arrows. The locations where thermocouples (TC) were attached to the tubes is apparent.**

only 85 MPa [9] so the C26M material is showing an increase in both the burst temperature and the stress where a large burst occurs. The previous testing for C26M was all at lower stress levels and only small openings were observed [36] similar to the bursts marked by arrows in Figure 5. Additional testing is in progress to further study the burst behavior of C26M in the 90-130 MPa stress range and obtain statistical information on the burst behavior.

**Development of a critical heat flux experiment**

An additional task this year was to develop a new type of experiment to study the relative performance of C26M under critical heat flux (CHF) conditions [40-42]. Potential experiments were suggested with different peak temperatures and internal pressure on the tube specimen:

Case 1: ramp from 300 to 400°C, back to 300°C; repeat for 20-50 cycles; internal pressure 5 MPa.
Case 2: ramp from 300 to 650°C, back to 300°C; repeat for 20-50 cycles; internal pressure 5 MPa.
Case 3: ramp from 300 to 650°C, back to 300°C; repeat for 20-50 cycles; internal pressure 7 MPa.
Case 4: ramp from 300 to 800°C, back to 300°C; repeat for 20-50 cycles; internal pressure 5 MPa.

The SATS software and hardware were modified to conduct these experiments. The initial tests were based on Case 2 and 12 cycles were conducted to demonstrate the capability, Figure 6. The cooling time needs to be increased in order to allow cooling back to 300°C between each cycle but otherwise it appears to be possible to conduct these various experiments (Cases 1-4) in the SATS for 20-50 cycles.

The only Zr-alloy material currently available to compare to C26M was one Zircaloy-4 (Zly-4) tube specimen. Thus, additional testing awaits obtaining Zr-alloy tubes for comparison to C26M. Some of the results from the first set of experiments is shown in Figures 7 and 8. As expected, no change in the tube diameter could be measured for the C26M tube after 12 cycles to 650°C, however, an increase in the tube diameter could be measured for the Zly-4 tube consistent with the center of the specimen being the hottest location. Likewise, Figure 8 shows cross-sections of the outer diameter of each tube after this trial CHF simulation exposure. No oxide can be seen at this

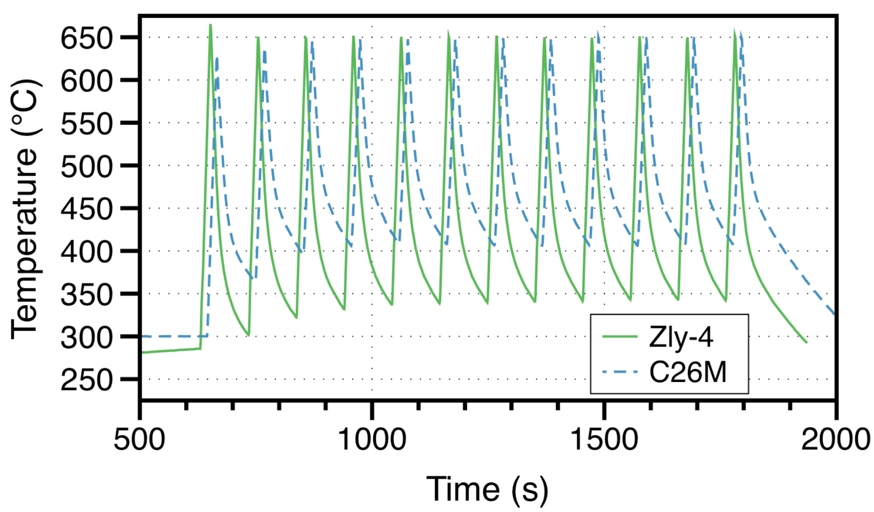


**Figure 6. Temperature as a function of exposure time for the first two CHF simulation experiments with a target peak temperature of 650°C, a 5°C/s heating rate and an internal pressure of 5MPa.**

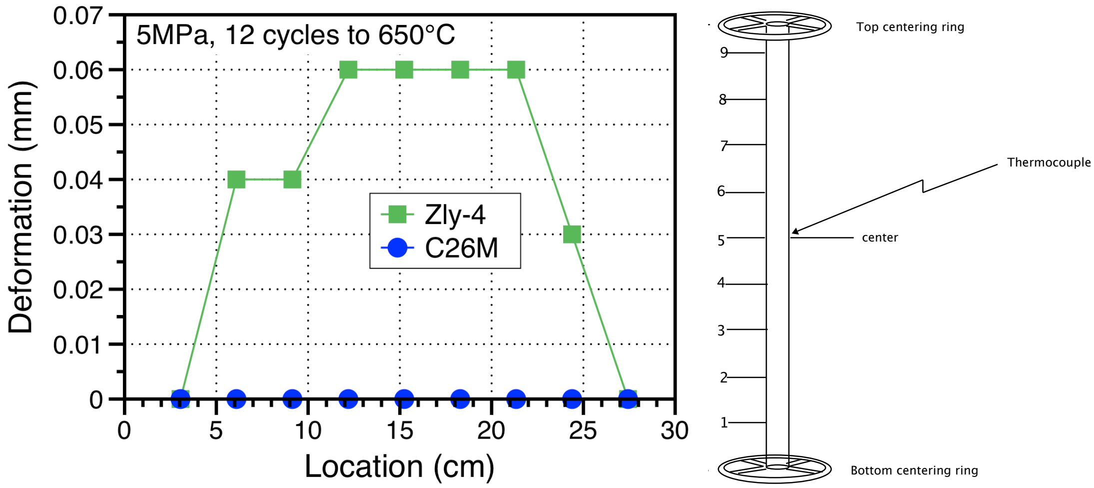


**Figure 7. Increase in tube diameter measured as a function of location on the tubes exposed under simulated CHF conditions.**

magnification on C26M, but an oxide layer can be seen on the Zly-4 tube. After the exposure, mechanical testing also can be conducted to determine the degradation in mechanical properties after these CHF simulations. Trial measurements on these two tubes are in progress.

**Summary**

This effort was focused on generating more high temperature data in the ORNL Severe Accident Test Station (SATS) on the new C26M (Fe-12Cr-6Al) commercial tubing that was inserted into Plant Hatch in February 2018. The high temperature steam oxidation testing of tube specimens has been evaluated in ramp testing at 1400°-1475°C. The behavior was very similar to the first generation B136Y composition (Fe-13Cr-6Al) except that a protective scale was formed on C26M after ramping to 1475°C. Additional LOCA burst testing has been conducted on the latest batch of C26M tubing. These initial results found that the burst size increased above 97 MPa hoop stress, which was a significantly higher stress than the weaker 1$^{st}$ generation results. Finally, based on industry feedback, a new type of test has been proposed to be relevant to the critical heat flux (CHF) conditions. Software and hardware modifications were made to enable this cyclic type experiment in the SATS. Two trial experiments were conducted on C26M and a Zr alloy (Zly-4) tube to compare their response under these conditions. The weaker Zly-4 tube deformed more and

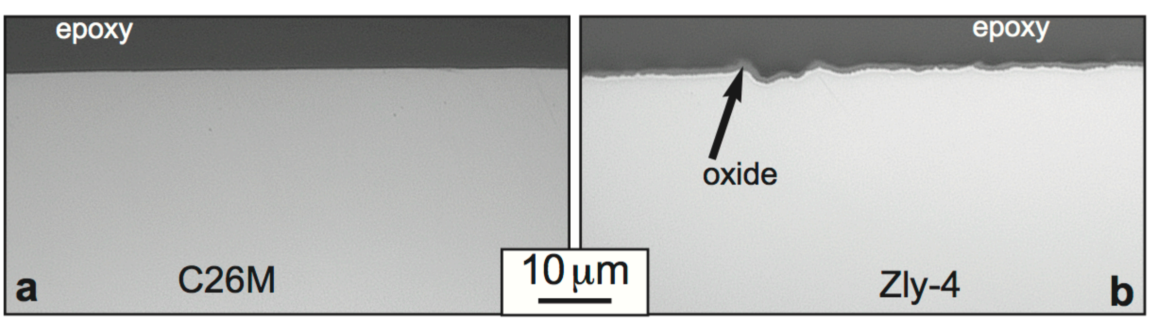


**Figure 8. Light microscopy of polished cross-sections of the outer diameter of (a) C26M and (b) Zly-4 tubes exposed under simulated CHF conditions. An oxide layer could be observed in (b).**

oxidized more than C26M under the same conditions of cycling 12 times to 650°C with an internal pressure of 5 MPa. Additional Zr alloy tubes are needed for CHF testing to continue. In addition, the current cycling parameters including peak temperature, heating rates and pressure, may be modified based on additional community feedback. Also, more oxidation and burst testing is in progress including additional post-test characterization.

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