

IRRADIATION TESTING OF ACCIDENT TOLERANT FUEL FeCrAl CLADDING

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Abstract:

The research, development, and deployment (RD&D) of accident tolerant fuel (ATF) FeCrAl cladding for light water reactors is a large international multi-program—multi-laboratory effort. This RD&D effort for ATF FeCrAl cladding is being driven by the expected gains in safety margins based on ATF FeCrAl claddings having excellent performance in simulated (both computationally and experimentally) design basis and beyond design basis scenarios. To date efforts have been exhaustive and include production of valuable data on alloy development, environmental testing (water-side corrosion and steam oxidation), mechanical performance, thin-walled tube production, and radiation tolerance to name a select few. Central to this extensive database is irradiation testing and evaluation. Irradiation tests are typically accelerated within simplified environments to reduce the time and cost of the testing while still allowing for a systematic evaluation of dominant variables contributing to any possible degradation of ATF FeCrAl cladding when in service. These tests range from materials screening tests using simple drop-in capsules containing tensile specimens irradiated in the High Flux Isotope Reactor to more complex, instrumented flow loop tests of shortened UO₂-containing FeCrAl fuel pins irradiated in the Halden reactor. These tests are combined with a wide range of post-irradiation examination (PIE) techniques and analyses. PIE typically includes both non-destructive and destructive evaluations to assess FeCrAl performance for a range of degradation issues such as radiation-embrittlement, water-side corrosion, cladding creep, and/or pellet-cladding interaction. This work provides a summary of these wide-ranging irradiation and PIE efforts.

Keywords: *FeCrAl, Accident Tolerant Fuel (ATF), Cladding, Radiation*

I. Introduction

Iron-chromium-aluminum (FeCrAl) alloys are currently being evaluated for commercial light water reactor (LWR) use as an accident tolerant fuel (ATF) cladding. FeCrAl alloys are an attractive alloy class for ATF cladding applications due to enhanced oxidation resistance in elevated temperature steam when compared to Zr-based alloys^{1–4}. Increased oxidation resistance is expected to provide gains in safety margins for both design basis and beyond design basis scenarios. The possible increase in safety margins has led to an extensive research, development, and deployment (RD&D) effort for nearly a half-a-decade. The primary goal of this RD&D effort is to deploy select alloy(s) as part of a Lead Test Rod (LTR) or a Lead Test Assembly (LTA) within a commercial LWR.

The deployment of a LTR/LTA requires an extensive database of material properties and fabrication scale-up demonstrations. Hence, the RD&D program's primary focus is on accident scenario testing, safety/economic analysis, fabrication studies (including commercial interfaces with vendors), unirradiated materials testing and evaluation, and accelerated irradiation testing. Data generated from these activities has resulted in compilation of a robust and vast material database. In particular, accelerated irradiation testing has allowed for a systematic evaluation of the dominant variables contributing to any possible degradation of ATF FeCrAl cladding when in service within a LWR. Irradiation testing has included a wide range of tests such as simple drop-in capsules containing tensile specimens to more complex, instrumented flow loop testing of a FeCrAl-clad, UO₂-fuel concept. Irradiation tests are complemented with extensive non-destructive and destructive post-irradiation examination (PIE) techniques to evaluate the performance of selected FeCrAl cladding concepts. This work provides a brief overview of FeCrAl irradiation campaigns that have been completed or are on-going and, when applicable, select results generated from PIE.

II. Overview of Irradiation Programs

An overview of the irradiation campaigns to evaluate the FeCrAl alloy concepts for ATF cladding applications is summarized schematically in Fig. 1. In short, four research reactors are being used including the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory, the Halden Research Reactor, the Advanced Test Reactor (ATR) at Idaho National Laboratory, and the Massachusetts Institute of Technology Reactor (MITR). Several ion beam facilities are also being used including the Intermediate Voltage Electron Microscope (IVEM)-Tandem in-situ irradiation facility at the Argonne National Laboratory, the ion beam facility at Los Alamos National Laboratory and University of Michigan Ion Beam Laboratory (MIBL). These irradiation campaigns were designed to evaluate specific concerns regarding FeCrAl. The facility in which each of the campaigns was conducted was selected based on the facility capabilities, availability, and suitability for evaluating the technical issue of interest. Details on specific campaigns will be discussed in later subsections. Ion beam testing has been focused on fundamental

scientific understanding of irradiation effects on FeCrAl⁵. Although this understanding is important, it has little impact on deployment of a LTR or LTA and hence this work is omitted from further discussion.

Fig. 1 also shows a range of different alloys used within the various irradiation campaigns. The wide range of alloys is the result of a tiered approach in the multi-phase development program for FeCrAl alloys. In this approach, lower complexity examinations such as mechanical properties evaluations are completed on simple FeCrAl alloys. The results of these lower complexity examinations are used to inform more complex and integral tests. The simpler FeCrAl alloys, designated with a “B” in their first-place alpha-numeric designation (and FC15AY in Fig. 1) are deemed “Generation I” FeCrAl alloys. These alloys typically contain only Fe, Cr, Al, and Y, and are not considered optimized compositions or microstructures. Generation I alloys are primarily used to evaluate trends due to composition changes in Cr and/or Al. Significant findings on these alloys are provided in multiple references^{6,7}.

The more complex campaigns such as creep studies (FCREEP, Halden Creep) and flowing loop tests (ATF-2, IFA-796) use the more complex FeCrAl alloys. These alloys are deemed “Generation II” alloys and have designations where the firstplace alpha-numeric designation has a “C”. The second-place holder designates the Cr content in wt.% + 10% and the thirdplace holder designates the Al content in wt.%. The remaining alpha-numeric designations are related to minor alloying elements. For example, C35M refers to a Generation II alloy with 13 wt.% Cr, 5 wt.% Al, 2 wt.% Mo, Y additions, and a Fe balance. Generation II alloys have optimized microstructures and fall within the “lean-alloy” chemistry class (<15 wt.% Cr) of the FeCrAl family. All Generation II FeCrAl alloys have been produced using traditional wrought alloy fabrication techniques. Other alloys in Fig. 1 include an experimental oxide dispersion strengthened (ODS) variant, 125YF, and commercial alloys such as Alkrothal 720 (K720), Kanthal APMT, and Kanthal AF.

II.A. HFIR irradiations

All HFIR irradiations utilize simple drop-in capsules that are irradiated in the central flux trap region (the region with the highest fast neutron flux) of the core. These drop-in capsules have no active temperature monitoring. Instead, irradiation temperatures are verified using passive SiC thermometry. Post-irradiation dilatometric measurements of the SiC thermometry are made during isochronal annealing. The irradiation temperature can be determined with reasonable accuracy by comparing the instantaneous coefficient of thermal expansion during heating vs. that during cooling of the passive thermometry⁸. Irradiation design temperatures are achieved by varying the capsule fill gas and/or the size of insulating gas gaps. The size of the insulating gas gaps depend on the amount of heat generated by neutron and gamma heating of the specimens and the supporting capsule components. These simple drop-in capsules provide fast and cost effective materials screening tests including post-irradiation flat-sheet tensile properties (FCAY and FCAT campaigns), embrittlement and fracture properties (FCAB campaign), cladding tube mechanical properties (FTUBE), and irradiation creep properties (FCREEP). Images of the FCAT and FCAB unassembled irradiation capsules are shown in Fig. 2 and Fig. 3, respectively.

The FCAY and FCAT campaigns are complementary programs. The FCAY campaign represents the first irradiation testing on modern-era FeCrAl alloys. This campaign was designed specifically to determine irradiation hardening and embrittlement in FeCrAl alloys and to develop structure-property relationships to provide guidance on alloy development. The FCAY program used capsules containing 36 SS-J type (5 mm x 1.2 mm x 0.5 mm gauge) flat-sheet dog bone specimens⁹ per capsule, with six specimens of six different alloys in each capsule. Irradiation target temperatures were set to 320°C with irradiation doses of 0.3 dpa, 0.5 dpa, 1.8 dpa, 7.0 dpa, and 13.8 dpa. Design values for dose and temperature were selected to determine hardening at prototypical pressurized water reactor (PWR) temperatures up to end-of-life dose levels. Postirradiation evaluations have included determination of mechanical properties^{9,10}, irradiation-enhanced precipitation^{9,11,12}, irradiation-induced dislocation loop formation^{9,13,14}, and irradiation-induced swelling^{9,13} as a function of composition and irradiation dose. PIE techniques have

included tensile testing, fractography, transmission and/or scanning transmission electron microscopy (STEM), atom probe tomography (APT), and small angle neutron scattering (SANS). All irradiations within FCAY and the subsequent PIE have been completed. In summary, FCAY irradiations demonstrated within Generation I FeCrAl alloys that irradiation-hardening and embrittlement can be attributed to both dislocation loop formation (of both $\frac{a}{2}\langle 111 \rangle$ and $a\langle 100 \rangle$ Burgers vector) and precipitation of the Cr-rich α' phase and the size and number density of both of these microstructural features is dependent on the microstructure, irradiation dose, and the Cr content of the FeCrAl alloys. Findings from the FCAY irradiations were the primary driving factor for lean-composition FeCrAl alloys used in Generation II FeCrAl alloy development.

The FCAT campaign includes irradiations of Generation II FeCrAl alloys as follow-on work to the completed irradiation and PIE of Generation I alloys in the FCAY campaign. The FCAT campaign uses nearly identical irradiation capsules including 24 SS-J type tensile specimens as well as novel SS-2E type miniature tensile specimens¹⁵. Both bulk specimens and welded FeCrAl specimens were contained within each irradiation capsule. To complement the FCAY campaign, irradiations were targeted to 330°C at 1.8 dpa, 7.0 dpa, and 13.8 dpa. Additionally, irradiations at temperatures of 200°C and 550°C were also included with the same target dose. These conditions were included in the irradiation test matrix to determine the critical role of temperature on both dislocation loop formation and precipitation of the Cr-rich α' phase. To date, 1.8-1.9 dpa irradiations have been completed for all three irradiation temperatures. Preliminary PIE including tensile testing, STEM, APT, and SANS have been completed. Results from the FCAT campaign indicate similar findings to the FCAY campaign but also has highlight the importance of irradiation temperature on the radiation tolerance of Generation II FeCrAl alloys.

The FCAB irradiation campaign serves to determine the role of Cr content on the precipitation of the Cr rich α' phase and the resulting changes, if any, on the fracture properties of FeCrAl alloys. The FCAB irradiation capsules are designed to fit four sub-sized pre-cracked Charpy V-notch (M4-PCCVN) specimens and four SiC thermometry samples. Each M4-PCCVN specimen contains four notches. Each capsule

contains two specimens of two different alloys. The RD&D program is interested in developing a relation between embrittlement (shift in the T_0 temperature) and the change in the yield stress of a given FeCrAl alloy. Hence, the FCAB program mimics the irradiation temperatures and doses of the FCAT campaign except that the lowest dose condition (1.8 dpa) is omitted, as it is expected that saturation in the change in yield strength would not be achieved at doses below 2 dpa¹⁰. The lowest dose (7.0 dpa) capsules have completed irradiation and are currently awaiting PIE to meet the goals of the irradiation campaign. The

FTUBE and FCREEP irradiation campaigns are the least mature HFIR irradiation campaigns. The FTUBE program serves to extend the work of the FCAY and FCAT irradiation programs from sheet-type feedstock to prototypical commercially-manufactured FeCrAl cladding tubes. Each irradiation capsule will hold up to two shortened segments of FeCrAl cladding and be irradiated at either prototypical PWR or boiling water reactor (BWR) irradiation temperatures and doses. The target cladding geometry for the campaign is a 17x17 array PWR cladding geometry. Design analyses and capsule assembly for the FTUBE capsules have been completed and irradiations are on-going. The FCREEP capsules are leveraging a design concept developed outside the RD&D program. The pressurized creep tubes used within the capsule will utilize an embossed foil to provide a constant heat transfer path from the creep tube to the surrounding capsule components throughout the irradiation. The embossed foil allows for a constant creep tube temperature despite significant irradiation creep, which would otherwise result in reduction of an insulating gas gap and a subsequent reduction in the temperature of the creep tube¹⁶. Conceptual designs for the FCREEP campaign irradiation capsules have been completed and irradiations are expected to begin in September 2017.

II.B. Halden irradiations

The Halden irradiations are designed to provide critical data regarding irradiation creep (Halden creep) and performance under PWR-type flowing loop conditions (IFA-796). Halden was selected for both of these irradiation campaigns due to the sophisticated and reliable instrumentation that can be incorporated into Halden experiments. For example, the Halden creep campaign uses flowing gas in the annular regions

of the irradiation capsules and feedback from multiple thermocouples to enable precise control of the irradiation temperature. The target irradiation temperature was set at 350°C. Additionally, the applied stress is controlled by varying the internal gas pressure of a sealed bellows. Cladding strain is determined using a calibrated linear variable differential transformer (LVDT). Such sophisticated instrumentation enables the production of the reliable data that is needed within the RD&D program, especially since only a limited number of alloys and irradiation conditions could be investigated.

The Halden creep experiment uses dog-bone tensile specimens to evaluate uniaxial creep. Specimens were tested in the unstressed and stressed conditions to determine both the swelling-induced dimensional changes and the strains resulting from both swelling and creep. For the stressed specimens, the stress level is varied as a function of time (or irradiation dose) to determine the creep rate as a function of applied stress. The initial stress was set at 325 MPa, which is representative of the expected stress that thin-walled FeCrAl cladding could experience in typical LWR geometries upon fuel pellet-cladding contact. Due to the in-situ nature of the experiment, the Halden creep experiment has been continuously generating data since its insertion. Preliminary analysis of the creep data indicated that the in-pile irradiation creep and out-of-pile thermal creep strain rates (completed separately from the in-pile tests) are similar, if not identical. The similarity in creep rates could be attributed to the low irradiation dose rate, which could result in a relatively large thermal creep component within the in-pile examinations. To confirm this, the aforementioned FCREEP experiment was launched within the HFIR to better differentiate between thermal and irradiation creep in candidate FeCrAl cladding.

The IFA-796 irradiation campaign is performed in a PWR test loop with a six-fuel rod configuration in an annular assembly. Five of the six positions in IFA-796 house four segmented, drop-in fuel rods while the sixth position contains a fully instrumented fuel rod. Candidate FeCrAl claddings will be included in both segmented rod positions and the fuel-length rod positions. The irradiation will be performed under prototypic PWR conditions with inlet and outlet coolant temperatures of 295°C and 320°C, respectively, and a pressure of 155 bar. The water chemistry consists of 2-3 ppm Li with B addition aimed at maintaining

a pH of ~ 7.2 . Fuel pellet enrichment in each rod will be tailored for a beginning-of life (BOL) linear heat rate (LHR) of ~ 25 kW/m and a fast neutron flux on the order of $\sim 2 \times 10^{13}$ n/cm²/s. Segmented rods will be investigated using non-destructive techniques such as visual inspection, Eddy current, and profilometry during reactor outages. PIE will also be performed at the end of the irradiation campaign and will include destructive evaluations such as optical metallography. The IFA-796 irradiation campaign is expected to begin in the Summer of 2017.

II.C. ART irradiations

ATR irradiation tests have focused on pellet-clad interactions and hence contain both prototypical UO₂ fuel and FeCrAl cladding within both irradiation campaigns. The first irradiation campaign, ATF-1, included simple drop-in capsule experiments. These experiments were developed to demonstrate irradiation capabilities and determine the hermeticity of the cladding and the welds, fuel-clad performance, and structural stability of the fuel rodlets under prototypical PWR irradiation conditions. ATF-1 used a double encapsulated irradiation design, which eliminated clad-coolant interactions and cladding creep due to the external coolant pressure. Two different irradiation geometries were used for FeCrAl cladding within ATF-1: 1.) shortened fuel rodlets containing standard PWR geometry UO₂ (shown in Fig.4 through Fig.6) and 2.) fuel-clad diffusion couple assemblies (shown in Fig.7 through Fig.9). The diffusion couple assemblies were developed, assembled, and irradiated to determine the effect of composition, grain size, and surface oxidation of the cladding on pellet-cladding chemical interaction. These experiments put the cladding and fuel in constant contact during irradiation. The shortened rodlets were designed to simulate standard fuel assembly geometry. For both assemblies, the targets were irradiated to burnup levels of 10, 30, and 50 GWd/Mt. The first rodlet irradiated to 10 GWd/Mt has completed irradiation and is currently being prepared for PIE.

A follow-on study to the ATF-1 irradiation campaign has been conceptualized and is currently in development. This program, named ATF-2, abandons the double encapsulated design in favor of a test loop where novel clad and fuel-clad designs can be tested in a neutron environment with simultaneous exposure

to prototypical PWR coolant. The primary goal of ATF-2 is to demonstrate the viability of a FeCrAl cladding concept under the most representative conditions, thus bringing FeCrAl cladding technologies further into commercial maturity. Additionally, the ATF-2 irradiation campaign serves as a direct complement to the Halden IFA-796 irradiation campaign. The design consists of a 2x3 arrangement of candidate fuel pins stacked in the axial direction with lengths of four or twelve inches. The top fuel pins in each stack can be instrumented. Fabrication of ATF-2 is currently on-going with a planned insertion in late calendar year 2017.

II.D. MIT irradiations

The MIT irradiation campaign is to test coolant-clad interactions under typical PWR water chemistries. Three modules are currently being irradiated: one in core, one in the core periphery, and one outside of the core. From this approach, the effects of neutron irradiation, gamma/radiolysis effects, and water chemistry effects on coolant-clad interactions can be determined independently. Irradiation capsules contain a mixture of sheet-type and clad tube coupons in an annular array with inlet and outlet holes on the top and bottom of the specimen holder assembly. The MITR is a lowpower reactor and hence the specimens will not be tested to end-of-life dose conditions for a typical commercial power reactor. Irradiation has begun and the irradiation was extended for two cycles of the MITR. PIE is expected to begin within calendar year 2017 and will include visual inspection, sample massing, and possible S/TEM investigations of the corrosion products.

III. Conclusions

A multi-year, multi-program effort focused on the RD&D of ATF FeCrAl claddings for commercial LWRs is ongoing. Success within this program could result in significant increases in the safe operation of nuclear power plants within the United States and abroad. To achieve this goal, a primary focus of the RD&D program is evaluating the performance of FeCrAl claddings in both normal and off-normal conditions. For this, extensive irradiation testing is required. These tests range from simple drop-in capsules to complex instrumented loop testing. Completed irradiation campaigns have had significant impact on the

RD&D program including the promotion of the lean-Cr composition FeCrAl alloys for ATF cladding use. Ultimately, the irradiation programs have enabled the foundation to be established for a LTR/LTA within a commercially operating LWR in calendar year 2018.

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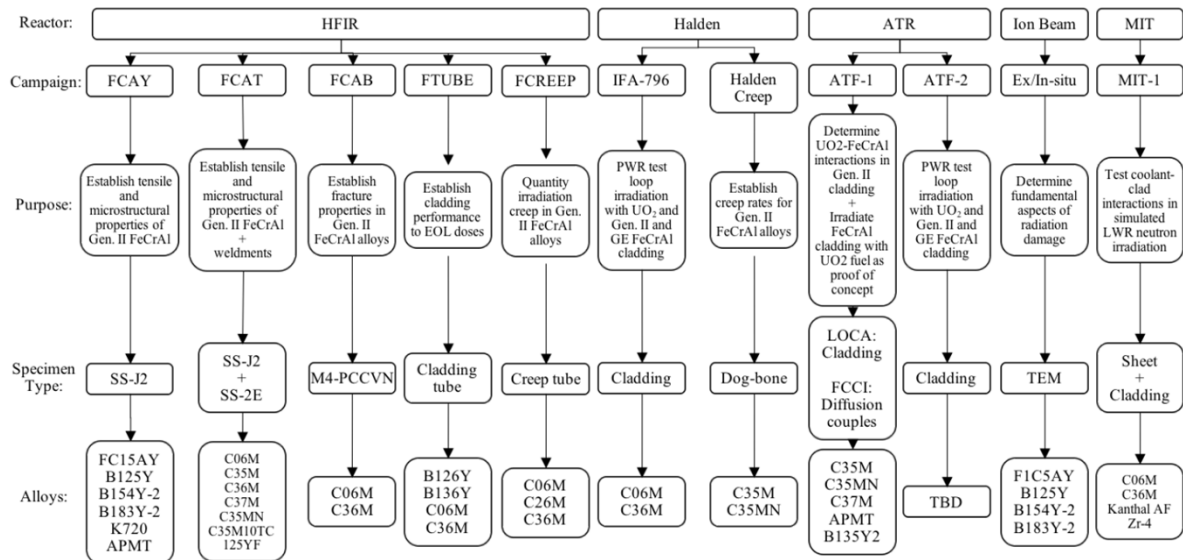


Fig. 1. Summary of irradiation campaigns for evaluating the ATF FeCrAl fuel cladding concept(s).



Fig. 2. Representative FCAT Capsule

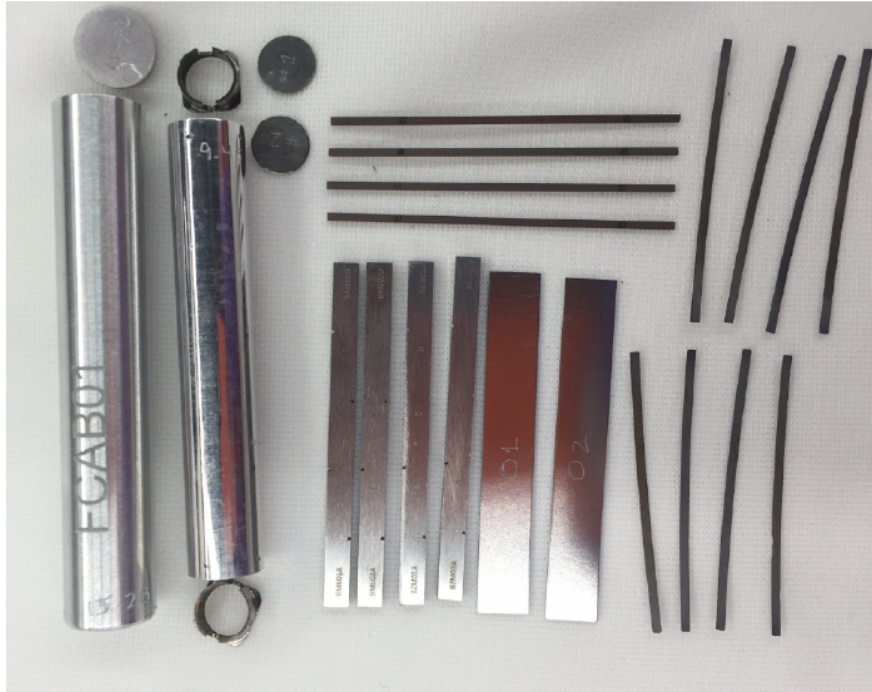


Fig. 3. Representative FCAB Capsule

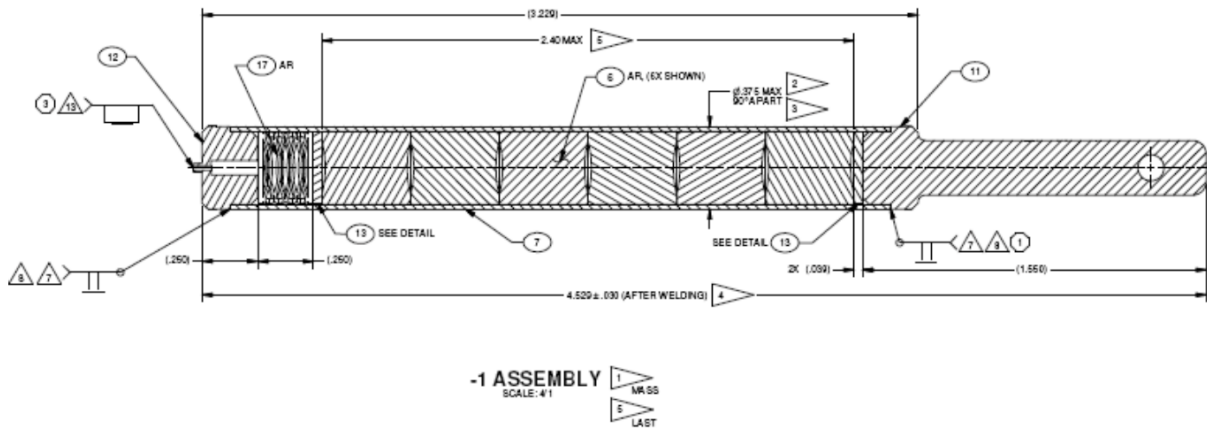


Fig. 4. Schematic of ORNL shortened FeCrAlATF rodlet.

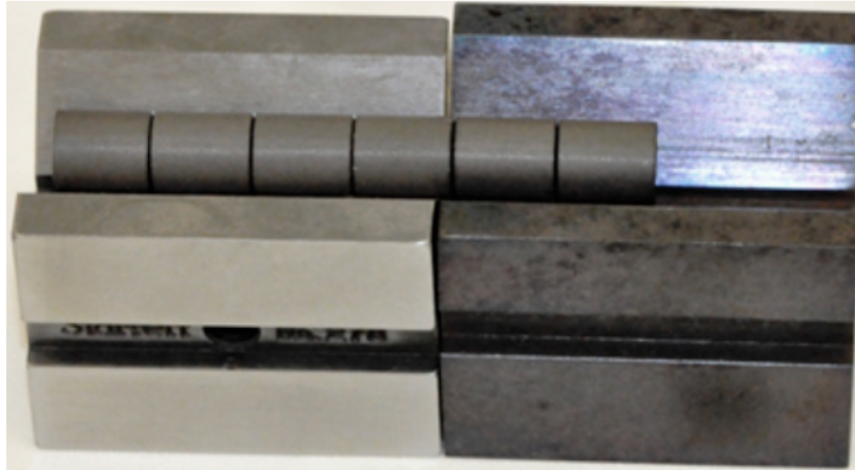


Fig. 5. Prototypical PWR fuel



Fig. 6. FeCrAl steel capsule containment

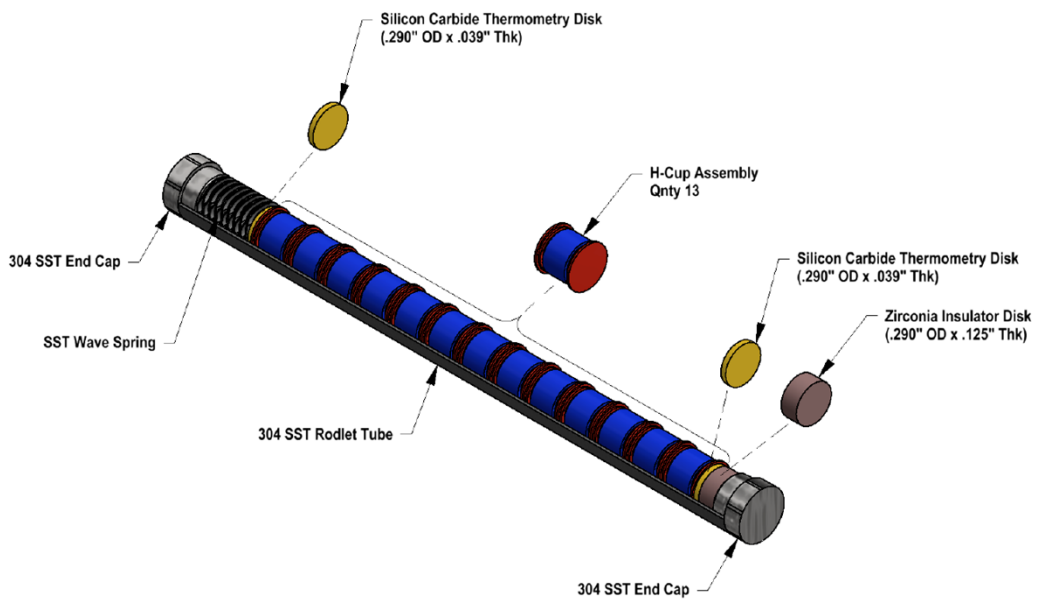


Fig. 7. Schematic of ORNL FCCI-ATF rodlet.



Fig. 8. "H-cup" piece parts and engraved sample coins.

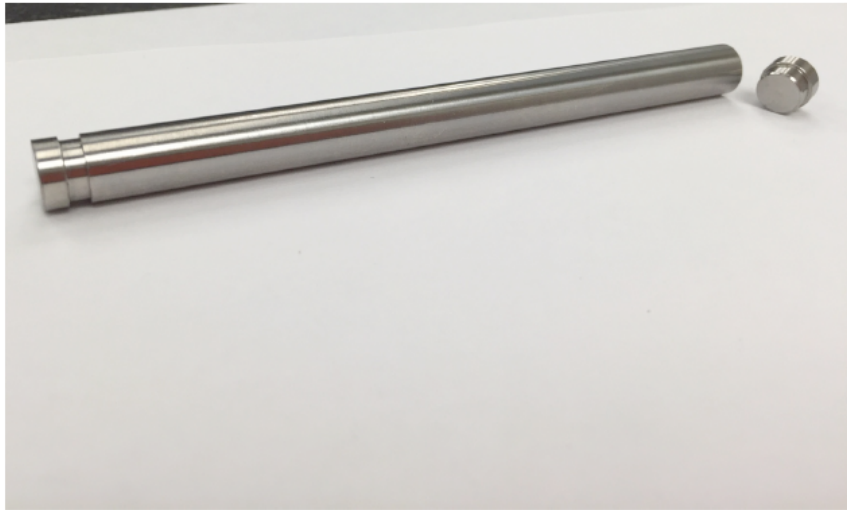


Fig. 9. 304L Stainless steel capsule containment