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Design and Irradiation of a Molten Salt Corrosion Experiment in The Ohio State University Research Reactor

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OAK RIDGE NATIONAL LABORATORY
MANAGED BY UT-BATTELLE FOR THE US DEPARTMENT OF ENERGY

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Reactor and Nuclear Systems Division

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NOMENCLATURE

MCNP	Monte Carlo N-Particle code
MSR	molten salt reactor
ORNL	Oak Ridge National Laboratory
OSU	The Ohio State University
OSURR	Ohio State University Research Reactor

1. INTRODUCTION

The primary containment system of any molten salt reactor (MSR) will be exposed to corrosive salt at high temperatures (>650°C)(McDuffee et al. 2018). Many MSR designs also use dissolved fuel, which introduces the challenge of fission product–induced corrosion. Even those designs that do not call for dissolved fuel might need to address the deleterious effects of dissolved fission products on the fuel containment system and potential particle failure.

The primary containment and all internal components should, for both design and safety purposes, (1) be resistant to salt corrosion, (2) maintain adequate strength under anticipated and off-normal conditions, and (3) be resistant to corrosion induced by entrained fission products. Some of these design issues can be managed without irradiation testing, but neutron-induced changes in structural materials and fission product–induced changes in the salt composition will inevitably modify the structural materials' behavior and can only be addressed with irradiation testing.

Irradiation testing in support of the MSR development will include a wide variety of experimental vehicles including static capsules, natural convection loops, and pumped loops under both low-flux and high-flux conditions. This document summarizes the design and irradiation of a low-flux, static capsule that was irradiated in the Ohio State University Research Reactor (OSURR) in 2018. The salt used for this experiment is a eutectic mixture of $KCl\text{-}MgCl_2$, which is a common composition for use as an out-of-core coolant and should have corrosive properties similar to other chloride salts such as $NaCl\text{-}MgCl_2$. Two alloys were chosen for this study: Alloy N, a low-chromium nickel-base alloy developed for use with molten salts, and 316 stainless steel, a common candidate material for MSRs due to its extensive use in nuclear systems. An experimental temperature of 800°C was chosen to accelerate degradation during exposure.

2. GENERAL DESIGN

2.1 FUNCTIONAL REQUIREMENTS

This simple static capsule experiment required that the following key parameters and design conditions be met.

- *Containment*.—The salt and specimen should be contained in a weld-sealed capsule for the duration of the experiment and transport. No salt should be able to escape.

- *Temperature*.—The salt must be molten (426°C) to create the conditions necessary to cause corrosion. It should be noted that some salts disassociate when irradiated at low temperature (Briggs 1964). The temperature should be controlled and measured for the entire irradiation period.

- *Salt volume*.—There should be a sufficient amount of salt in the experiment so that corrosion products do not significantly change the salt composition, which would affect the specimen's corrosion rate. The salt volumes and specimen surface area used for the current experiment were taken from successful out-of-pile corrosion experiments being conducted at Oak Ridge National Laboratory (ORNL).

- *Materials*.—Materials should be carefully selected to ensure compatibility with high-temperature salt.

2.2 DESIGN DESCRIPTION

The experiment team took advantage of two key pieces of existing infrastructure to accelerate the testing program: (1) a proven molybdenum capsule successfully deployed under an ongoing corrosion testing program at ORNL, and (2) an existing furnace sized for the OSURR that can hold the existing molybdenum capsule without significant modification.

2.2.1 Specimen

The specimen to be held in the salt is a small rectangular shape measuring 6.4 mm × 1.59 mm × 12.7 mm. Before assembly, the specimen is attached by a small wire to the molybdenum container end cap. Figure 1 shows how the specimen was attached to the end cap.

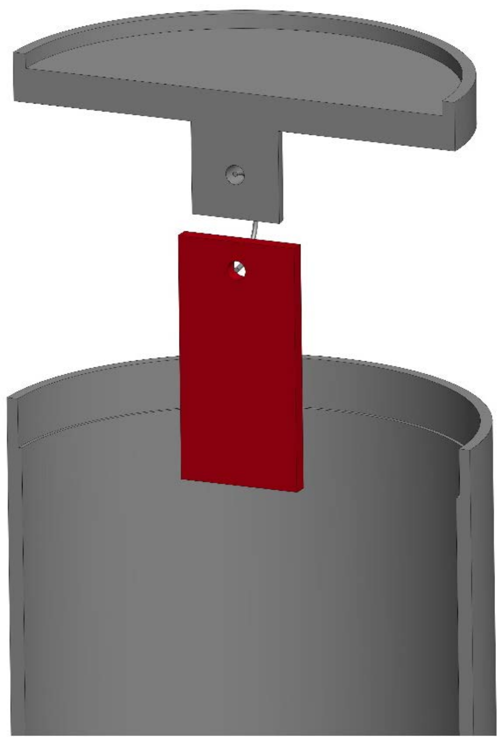

Figure 1. Corrosion specimen.

The nominal volumes for the specimen and wire were 127 mm^3 and 2 mm^3, respectively. The specimen mass depends on the material density, but assuming a density of 8 g/cm^3 (typical for stainless steel), the specimen mass was approximately 1 g. The molybdenum wire mass was approximately 0.02 g. The surface area of the specimen was approximately 112 mm^2.

2.2.2 Salt Capsule

The salt capsule housing consists of a small molybdenum container that is open on both ends. A solid end cap is welded to one end of the capsule. The container is then filled with solid salt chunks. The specimen is attached to the opposite end cap with wire, and the end cap is circumferentially welded to the other end of the capsule under vacuum conditions. Figure 2 shows the completed capsule assembly. Note that during irradiation the capsule is inverted so that the specimen is immersed in the liquid salt.

The volume of the housing and end caps is 6.36 cm^3. All parts are molybdenum, which has a nominal density of 10.22 g/cm^3, so the resulting mass is 65.0 g.

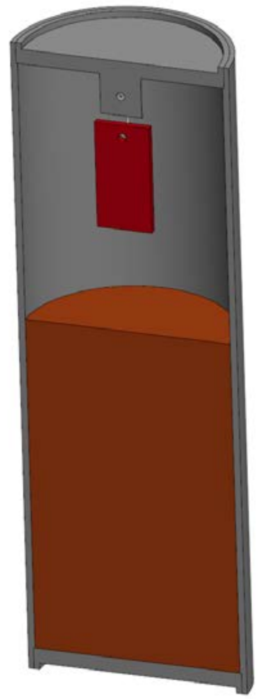

Figure 2. Salt container with salt and specimen.

2.2.3 Salt Capsule Primary Containment

Four capsule assemblies are stacked and loaded into the salt capsule primary containment. The assemblies consist of a simple titanium cylinder with a 50.8 mm outer diameter and 406 mm length fitted on each end with a plate that is 3.18 mm thick. The purpose of the primary containment is to provide a sealed environment with which to handle, move, irradiate, and ship the inner capsule assembly stack. Figure 3 shows the primary containment assembly, both with and without a capsule stack inserted. The capsule stack inside the primary containment is wrapped in Grafoil (not shown in Figure 3, but visible in the assembly pictures in Section 6), which serves as a diffusion buffer between the molybdenum and titanium and fills the gap between the stack and the containment to reduce mechanical interaction.

The combined volume of the cylinder and two end caps is 123.4 cm^3. Titanium has a density of 4.43 g/cm^3, so the total mass is approximately 547 g.

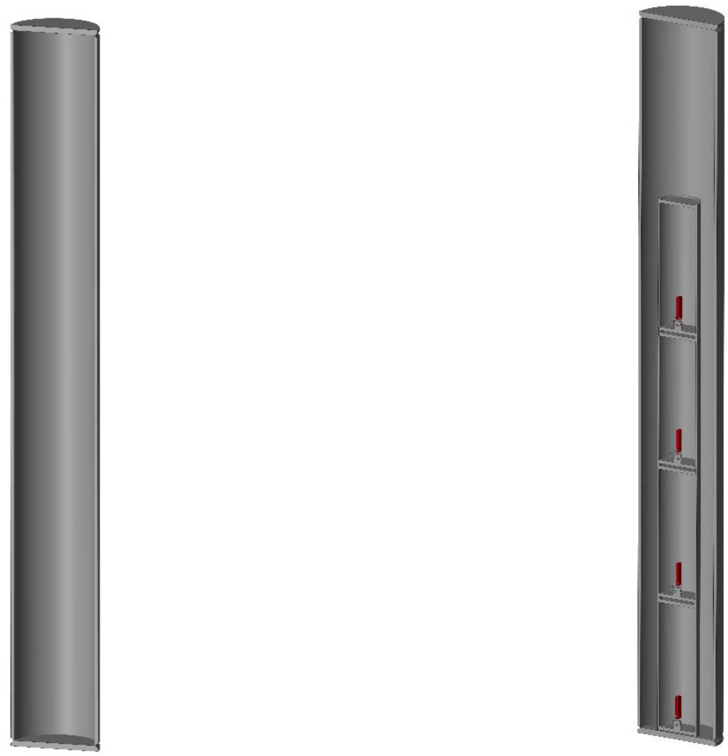

Figure 3. Primary containment assembly, shown (left) empty and (right) filled with four capsules.

2.2.4 Furnace and Insulators

This experiment used a Thermcraft VF-360-4-12-V-S furnace that has a 19.7 cm outer diameter and 53.34 cm height. The furnace uses a Kanthal A-1 Resistance wire coiled around the inner surface of the heated region. The insulation consists of Thermcraft 2300 vitreous aluminosilicate fibers (composition: 42% alumina, 56% silica, 2% other). The heater operates at a maximum power of 1800 W, a maximum voltage of 240 V, and a maximum temperature of 1100 °C. The heated length is 45.7 cm, and the heated inner diameter is 6.35 cm. Figure 4 shows the furnace used in this experiment inside the basket (described in Section 2.2.5) and a view through the center hole. The furnace volume is 9809.9 cm^3; its mass is 5.85 kg.

The furnace is capped top and bottom with Thermcraft 2300 insulators. The top insulator has a volume of 1366.6 cm^3 and a mass of 0.620 kg. The bottom insulator has a volume of 1505.5 cm^3 and a mass of 0.680 kg.

Figure 4. Experiment furnace.

2.2.5 Basket Assembly

The basket assembly—a metal framework with four tie rods (shown in Figure 4, left panel)—holds the experiment furnace and allows easy removal of all internal components from the secondary containment. The basket assembly is fabricated from grades 2 and 5 titanium alloys and has a total volume of 661.1 cm^3. The assembly's total mass is 3.065 kg.

2.2.6 Secondary Containment

The secondary containment includes an Al-6061 cylinder with a 228.6 mm outer diameter, 203.2 mm inner diameter, and 1532.5 mm length, as well as a solid Al-6061 lower end cap that is 15 mm thick. These two pieces together have a volume of 13.660 cm^3 and a total mass of 36.88 kg.

The upper lid assembly (see Figure 6) consists of a 6.4 mm thick stainless steel plate with a lifting lug welded to the top. Multiple pass-through holes in the lid allow for power, gas piping, and instrumentation. The lid assembly is attached to the main cylinder housing using 16 titanium 5/8 in. screws and sealed using a 3/32 in. O-ring.

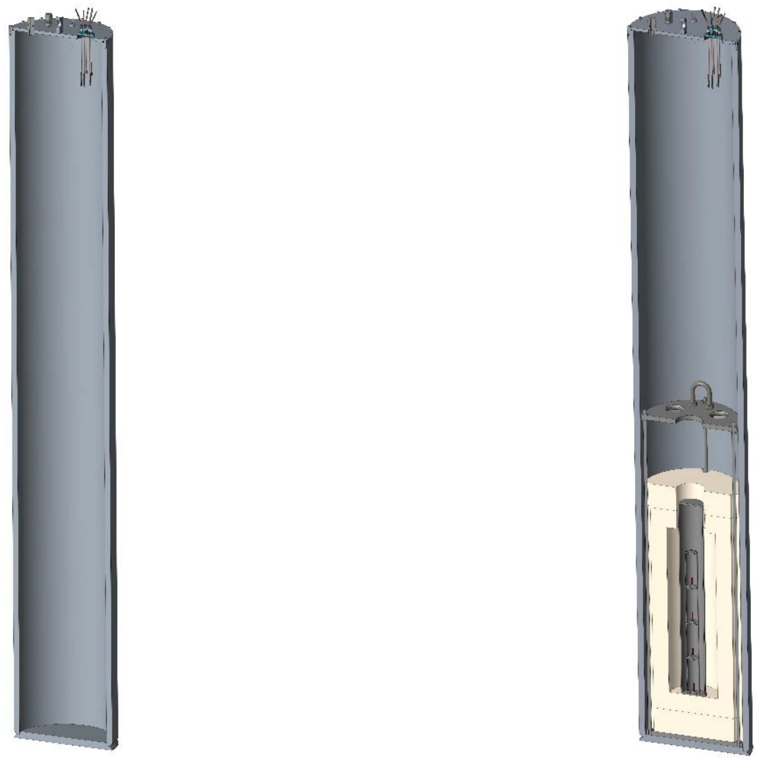

Figure 5. Secondary containment, shown (left) empty and (right) filled.

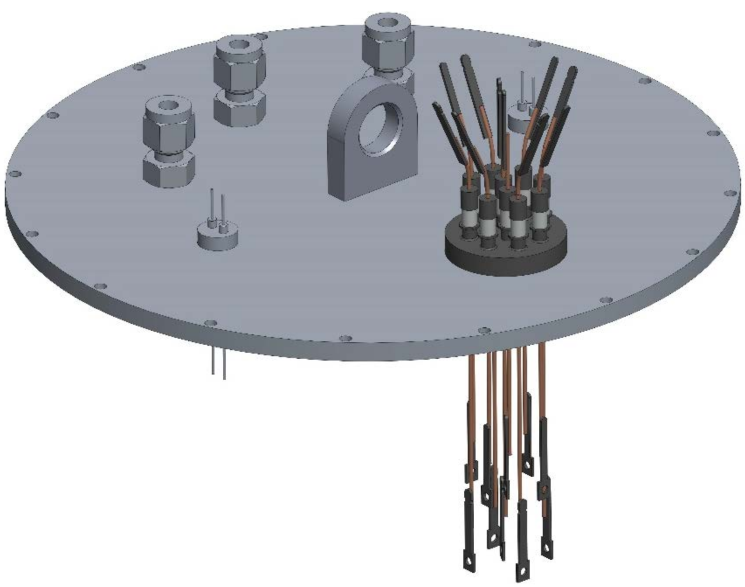

Figure 6. Secondary containment lid assembly.

The lid, lifting lug, tube fittings, and feedthroughs have a total volume of 274.06 cm^3 and a mass of 2.192 kg. The titanium screws each have a volume of 0.187 cm^3 and a mass of 0.82 g. All 16 screws have a total mass of 13.1 g.

2.2.7 Design Drawings

Table 1 provides the complete drawing list for this experiment.

Table 1. Drawing list for the molten salt corrosion experiment

Drawing no.	Title	Includes
S16-36-DETECTORB	Secondary containment bottom assembly	Secondary containment bottom cap Secondary containment tube Secondary containment bottom assembly
S16-37-DETECTORB	Secondary containment lid assembly	Secondary containment lid assembly Secondary containment lid plate Lifting lug Tube fittings Power feedthroughs Thermocouple feedthroughs
S16-42-DETECTORB	Basket assembly	Basket assembly Basket rods Basket bottom plate Basket top plate U-bolt Hex nuts
S16-43-DETECTORB	Insulator disks	Bottom cap Top cap Furnace bottom cap Furnace top disk
S18-05-SALT_CAP	OSU salt capsule experiment assembly	Salt capsule irradiation experiment
S18-03-SALT_CAP	OSU salt irradiation capsule part details and assembly	Salt irradiation capsule assembly Salt capsule housing Specimen Top end cap Perforated specimen holder Salt Wire
S18-32-SALT_CAP	OSU salt irradiation capsule primary containment details	Primary containment Tube End cap Lifting lug

2.2.8 Part Mass Summary

Table 2 summarizes the parts used in the experiment and their respective masses. The total experiment mass is 50.23 kg (111 lbm).

Table 2. Part mass summary

Part	Material	Part mass (g)	Experiment count	Total mass (g)
Specimen	Steel or nickel alloy	1.00	3*	3.00
Wire	Molybdenum	0.02	3*	0.06
Salt	$KCl-MgCl_2$	30.	3*	90.
Salt Capsule Housing	Molybdenum	65.	3*	195.
Primary Containment	Titanium alloy	547.	1	547.
Salt container wrap	Grafoil	94.71	1	94.71
Furnace	Alumina/silica	5850.	1	5850.
Furnace insulators	Alumina/silica	1300.	1	1300.
Basket assembly	Titanium alloy	3065	1	3065

Table 2. Part mass summary (continued)

Part	Material	Part mass (g)	Experiment count	Total mass (g)
Secondary containment	Al-6061	36,880	1	36900
Upper lid assembly	Stainless steel	2192	1	2190
Lid assembly screws	Titanium alloy	0.82	16	13.1
			Experiment total	50,230

** The design called for four capsules, but a weld failure during fabrication led to the removal of one of the capsules.*

3. OHIO STATE UNIVERSITY RESEARCH REACTOR

The OSURR is a pool-type reactor located at The Ohio State University (OSU) in Columbus, Ohio. It is licensed to operate at a thermal power of up to 0.5 MW with an active fueled length of 38 cm. The OSURR is operated using an on-demand schedule. This makes it ideally suited to short-term irradiations.

Three irradiation positions are available within the core grid, with each consisting of a dry tube that extends from the top of the pool to a position in the core grid. Moveable vertical dry tubes with 17.8 cm and 25.4 cm outer diameters are available for placing experiments at or near the core boundary. Table 3 provides a summary of the size and neutron flux for these irradiation positions.

Table 3. Characteristics of selected irradiation positions in the OSURR

Irradiation positions	Inner Diameter (cm)	Thermal flux (10^{13} n/cm^2·s)	Fast flux (1 MeV eq.) (10^{13} n/cm^2·s)
Central Irradiation Facility	3.30	1.4	0.47
Auxiliary Irradiation Facility	6.22	0.45	0.26
Peripheral Irradiation Facility	6.35	0.31	0.12
17.8 cm dry tube	16.51	0.11	0.02
25.4 cm dry tube	24.13	0.081	0.016

The 25.4 cm diameter dry tube was chosen as the irradiation position for this experiment. The large diameter allows for larger-scale experiments. The relatively low neutron flux is not conducive to high-fluence experiments but may be an ideal test bed for assessing cartridge loop technology in a facility more easily accessible than a national laboratory facility. The OSURR can accept fueled salt, but their post-irradiation handling limits may restrict the fuel loading and burnup to relatively low levels (The Ohio State University College of Engineering 2018). Although licensed to operate at 500 kWth, experiments in the 25.4 cm diameter dry tube are limited to reactor power below 250 kWth due to radiation streaming up the tube and limited shielding used at the pool top.

4. SALT AND SPECIMEN SELECTION

The salt used for this experiment is a eutectic mixture of KCl-$MgCl_2$ in a 68:32 molar ratio. This is a common composition for use as an out-of-core coolant and should have corrosive properties similar to other chloride salts such as NaCl-$MgCl_2$. Due to the short duration of exposure, an aggressive salt containing ~1% H_2O was chosen to maximize the severity of attack during the limited exposure to ensure measurable alloy degradation. However, if the effect of irradiation is small, it is possible that the effect of impurity-based corrosion will overwhelm the effect of irradiation on alloy degradation. For this reason, a second, less aggressive salt containing <30 ppm H_2O was also chosen.

Two alloys were chosen for this study: Alloy N, a low-chromium nickel-base alloy developed for use with molten salts, and 316 stainless steel, a common candidate material for MSRs due to its extensive use in nuclear systems. An experimental temperature of 800°C was chosen to accelerate degradation during exposure. Table 4 provides the experimental matrix. Half of the samples were separated for irradiation, and the other half will receive the same temperature treatment without irradiation.

Table 4. Salt/specimen experiment matrix

Capsule ID	Sample ID	H_2O impurity	Salt weight (g)	Specimen	Irradiated?
1	OSU-6-D-I	~1%	30.05	316 SS	YES
2	OSU-N-D-I	~1%	29.99	Hastelloy N	YES
3	OSU-6-D-U	~1%	29.96	316 SS	NO
4	OSU-N-D-U	~1%	30.06	Hastelloy N	NO
5	OSU-6-C-I	<30 ppm	30.02	316 SS	YES
7	OSU-6-C-U	<30 ppm	30.06	316 SS	NO

Note: SS=stainless steel

5. ACTIVITY AND DOSE ESTIMATE

An activation analysis of the experiment was conducted to determine when the experiment could be handled and ultimately shipped back to ORNL. The activation analysis was completed using the ORIGEN code within the SCALE software package (Oak Ridge National Laboratory 2011). ORIGEN calculates the activation products (including mass and activity) after irradiation with user-specified mass, composition, neutron flux energy spectrum, irradiation time, and decay time. Monte Carlo N-Particle code (MCNP) (X-5 Monte Carlo Team 2003) simulations were performed using a model of the OSURR to determine the neutron flux energy spectrum in the 25.4 cm dry tube. The ORIGEN simulations were completed for 3 days of irradiation at a reactor power of 250 kW and 7 hours of irradiation per day.

Decay time between successive days of irradiation was included in the simulations, as was additional decay time after the third day of irradiation. The atomic masses of the individual experimental components were input into the ORIGEN code in two separate categories. The salt capsules, salt, and specimens were categorized together, while the rest of the components were analyzed separately. These groupings were created such that an appropriate shipping date for the salt capsules could be determined. All components except for the lid of the secondary containment were assumed to receive the centerline total neutron flux value of $7.12 \cdot 10^{11}$ n/cm^2/s within the dry tube for 250 kW reactor power. The secondary containment lid, which sits roughly 1.2 m above the core centerline, received a neutron flux of $5.87 \cdot 10^{9}$ n/cm^2/sec based on MCNP calculations using a model of the OSURR core.

Table 5 presents the results of the activation analysis, using 238 group cross sections from ENDF/B-VII.1 (Brookhaven National Laboratory 2018) and JEFF-3.0/A (Nuclear Energy Agency 2018). The activity for each element was output by ORIGEN. The activities were then used to calculate the gamma dose rate for the experiment using gamma ray dose conversion factors (Unger and Trubey 1982) as shown in Table 6. The dominant activity and dose contribution of the salt capsules results from ^{51}Cr, ^{99}Mo, and ^{99m}Te. The dominant activity and dose contribution of the supporting experimental components results from ^{51}Cr, ^{59}Fe, and ^{65}Zn. An initial large activity and dose immediately follow irradiation due to ^{28}Al produced in the secondary containment, but decays quickly with a half-life of 2.2 min.

Table 5. Post-irradiation total activity (Ci)

Activity	Other components	Salt capsule and specimens	Total
Day 1 irradiation	1.99E+03	2.63E+01	2.02E+03
Decay	6.90E+00	6.06E–01	7.51E+00
Day 2 irradiation	2.00E+03	2.68E+01	2.02E+03
Decay	8.70E+00	8.70E–01	9.57E+00
Day 3 irradiation	2.00E+03	2.70E+01	2.02E+03
Day 4 morning	9.32E+00	1.01E+00	1.03E+01
Day 5 morning	2.71E+00	4.90E–01	3.20E+00
Day 6 morning	1.12E+00	2.88E–01	1.40E+00
Day 7 morning	6.70E–01	1.99E–01	8.69E–01
Day 8 morning	5.38E–01	1.52E–01	6.91E–01

Table 6 shows that, on the morning of Day 8, the experiment was within OSU's handling limits for tool manipulation (dose rate of 100 mrem/hr at 1 m). The salt capsule experiment remained in the dry tube for four full weeks following irradiation. The experiment was then relocated to another temporary storage location for one day before being re-packaged and shipped back to ORNL.

Table 6. Post-irradiation dose rate (Rem/hr at 1 m)

Activity	Other components	Salt capsule and specimens	Total
Day 1 irradiation	1.72E+03	1.86E+01	1.74E+03
Decay	1.91E+00	9.39E-01	2.84E+00
Day 2 irradiation	1.72E+03	1.92E+01	1.74E+03
Decay	2.26E+00	1.26E+00	3.51E+00
Day 3 irradiation	1.73E+03	1.95E+01	1.74E+03
Day 4 morning	2.37E+00	1.37E+00	3.73E+00
Day 5 morning	4.98E–01	4.66E–01	9.64E–01
Day 6 morning	1.61E–01	1.66E–01	3.27E–01
Day 7 morning	6.14E–02	6.42E–02	1.26E–01
Day 8 morning	3.12E–02	2.86E–02	5.97E–02

6. CAPSULE ASSEMBLY

As described in Section 4, each molybdenum container was filled with approximately 30 g of solid salt in a glove box and then seal-welded in a vacuum. Figure 7 shows the three seal-welded containers selected for irradiation.

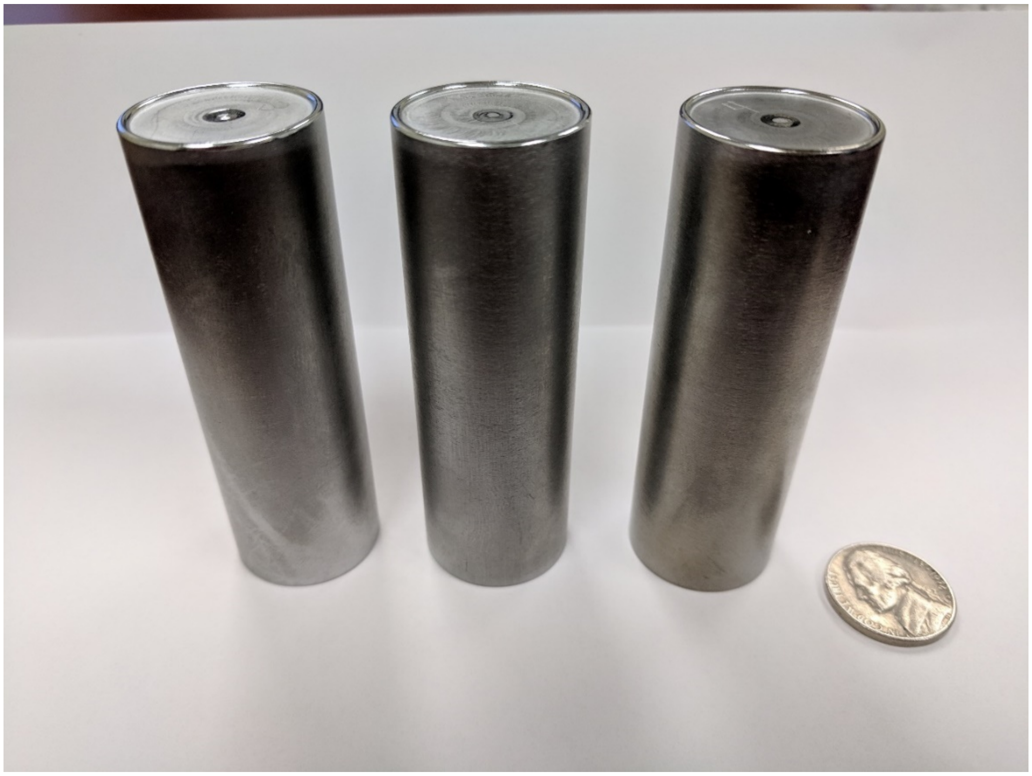

Figure 7. Salt capsules after welding.

Figure 8 shows the layout of the three capsules, along with the primary containment vessel and peripheral parts. The capsule stack is wrapped in Grafoil, which serves as a diffusion buffer between the molybdenum and titanium and fills the gap between the stack and the containment to reduce mechanical interaction.

Figure 8. Primary containment assembly including vessels and peripheral parts.

After welding in argon and conducting a weld inspection, the primary containment assembly was leak tested and radiographed. Figure 9 shows the results of radiography performed on the primary containment post-welding to ensure that the capsule stack was located properly. The salt density is too low to be seen, but the molybdenum capsules, end cap, and specimens are clearly visible. Note that the capsule stack was inverted from that shown in the radiograph.

After welding and weld qualification of the primary containment, three thermocouples were welded to the side of the primary containment, as shown in Figure 10.

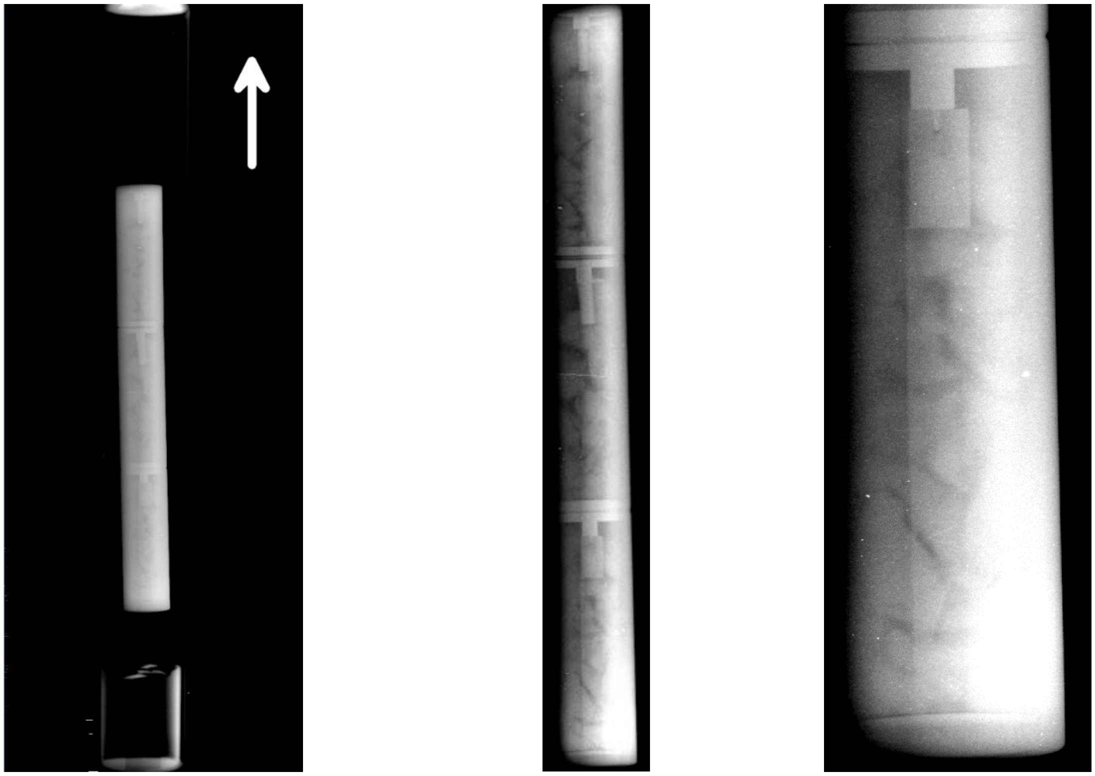

Figure 9. Results of radiography performed on the (left) secondary containment, (middle) capsule stack, and (right) single container.

At OSU, the primary containment, with thermocouples attached, was placed into the furnace and then into the secondary containment. The major pieces to the assembly are shown in the left panel of Figure 10. The secondary containment was then put under vacuum and filled with argon (middle panel). Once purged, the entire assembly was placed into the 25.4 cm dry tube in the reactor (right panel).

Figure 10. Final experiment assembly onsite at OSU.

7. OPERATION

The OSURR began withdrawing control rods at 7:37 a.m. on Tuesday, August 14, 2018, and reached 250 kW at 8:42 a.m. The reactor shut down for the day at 3:42 p.m. after 7 hours of continuous operation. The reactor followed a similar pattern over the next 2 days, operating for a total of 6,000 kW h. Before each day's ascent to 250 kW, the experiment was brought to a steady-state temperature of 800°C, as measured by the three thermocouples attached to the primary containment's outer surface. Figure 11 shows the experiment loaded in the dry tube under irradiation. Figure 12 shows the combined temperature profile for the 3 operating days.

Figure 11. Experiment under irradiation at the OSURR.

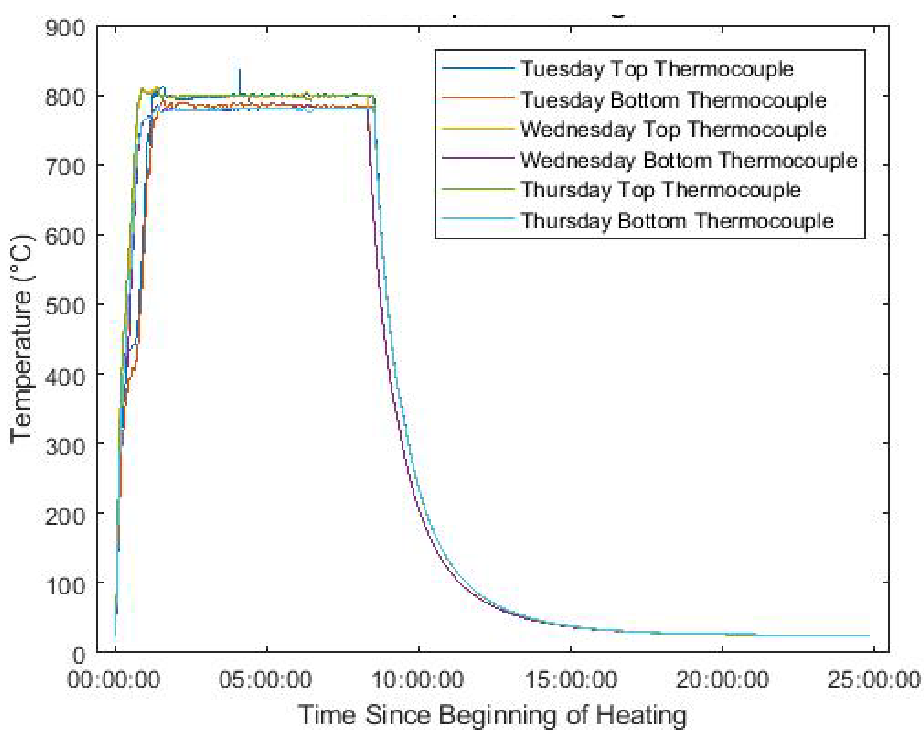

Figure 12. Experiment temperature profile overall operating days.

8. SUMMARY AND CONCLUSIONS

This report summarizes the design and irradiation of a low-neutron-flux, static capsule irradiated at the OSURR in 2018. A eutectic salt mixture of KCl-$MgCl_2$ in a 68:32 molar ratio was placed in each of three molybdenum capsules, along with either an Alloy N or 316 stainless steel specimen. Due to the short duration of exposure, an aggressive salt containing ~1% H_2O was chosen for two capsules to maximize the severity of attack during the limited exposure time. A less aggressive salt containing <30 ppm H_2O was used in the remaining capsule. All three capsules were irradiated at 800°C for a total of 6,000 kW h. The experiment has been shipped back to ORNL, and the specimens will be evaluated in fiscal year 2019.

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