Oak Ridge National Laboratory

Module 10: Operating Experience

David E. Holcomb, Ph.D.

Nuclear Energy and Fuel Cycles Division

November, 2022

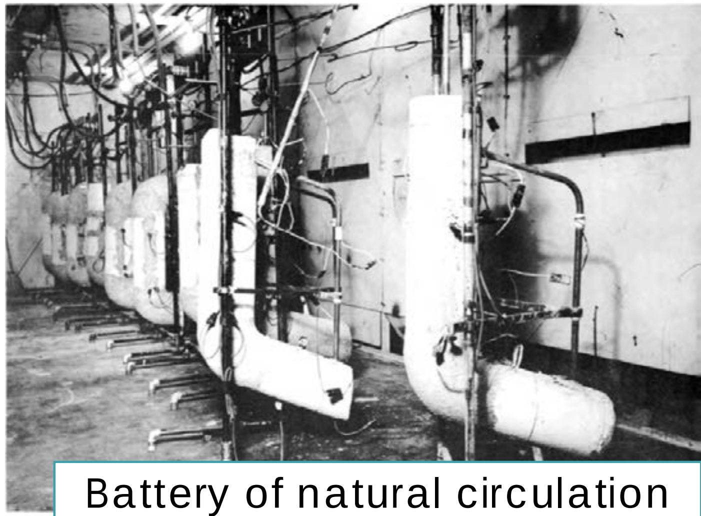
ORNL is managed by UT-Battelle, LLC for the US Department of Energy

U.S. DEPARTMENT OF ENERGY

ORNL Operated Many Fuel and Coolant Salt Test Facilities from the 1950s to the 1970s

- 2 operating MSRs
- 4 critical assemblies
  - Cold Aircraft Reactor Experiment (ARE), both cold and hot (liquid salt) Aircraft Reactor Test (ART), and a hot critical for the Pratt & Whitney Aircraft Reactor (PWAR)
- Multiple* (about a dozen) in-pile fuel salt loops at three test reactors
  - Materials Test Reactor
  - Low Intensity Test Reactor (both horizontal and vertical loops)
  - Oak Ridge Research (ORR) Reactor
- Numerous in-pile corrosion test capsules as well as high dose proton beam, electron, and gamma irradiations
- Many ex-core natural circulation test loops

ORR In-Pile Fuel Salt Loop



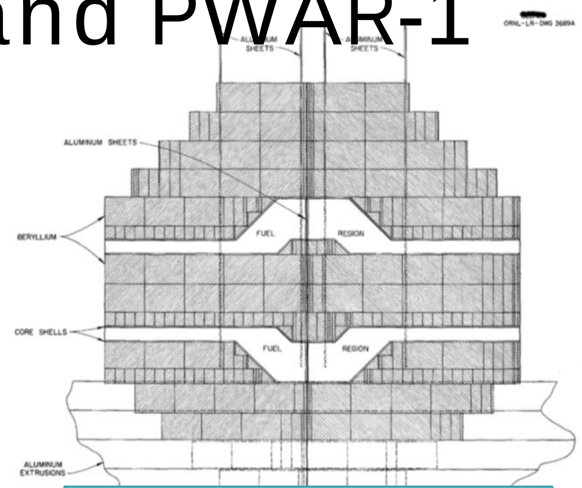
Battery of natural circulation loops as of 1957

*Extensive re-use of components between loops makes number of distinct loops unclear

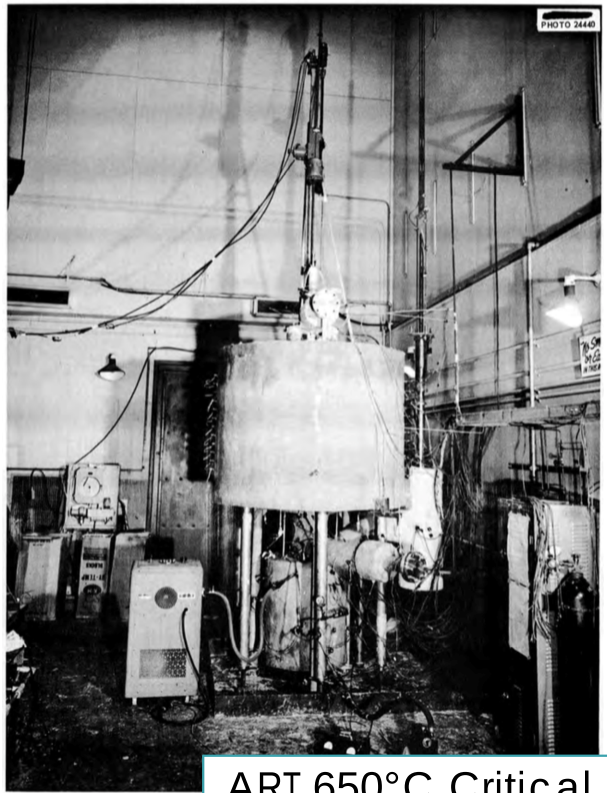
Oak Ridge National Laboratory

Critical Tests Were Performed for ARE, ART, and PWAR-1

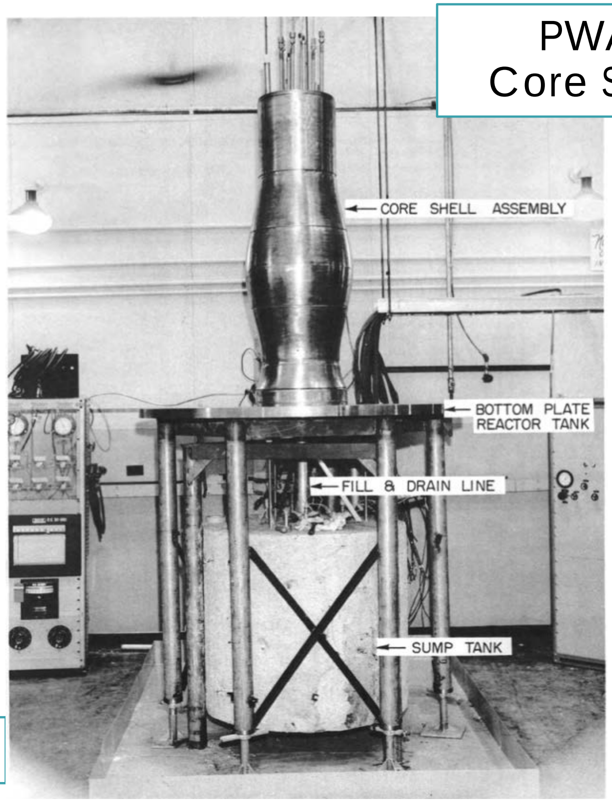
- ARE employed two room temperature system mock-ups
- Both hot and cold critical assemblies were employed for ART
  - Liquid salt test was to observe temperature reactivity coefficient
- PWAR-1 critical resembled ART high temperature critical



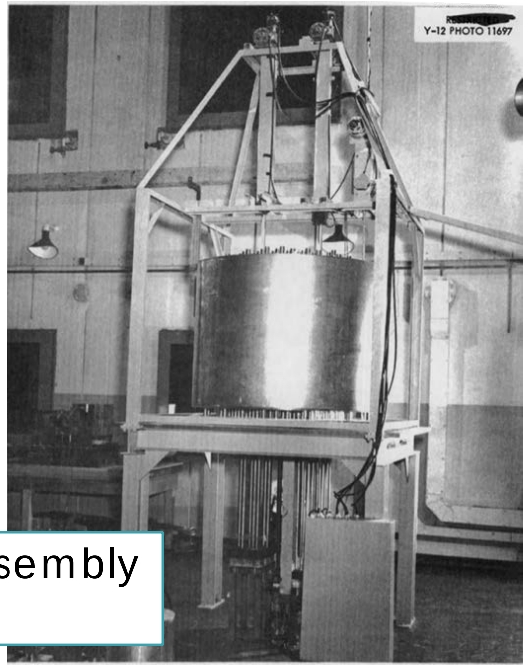
ART Room Temperature Critical Assembly



ART 650°C Critical Assembly



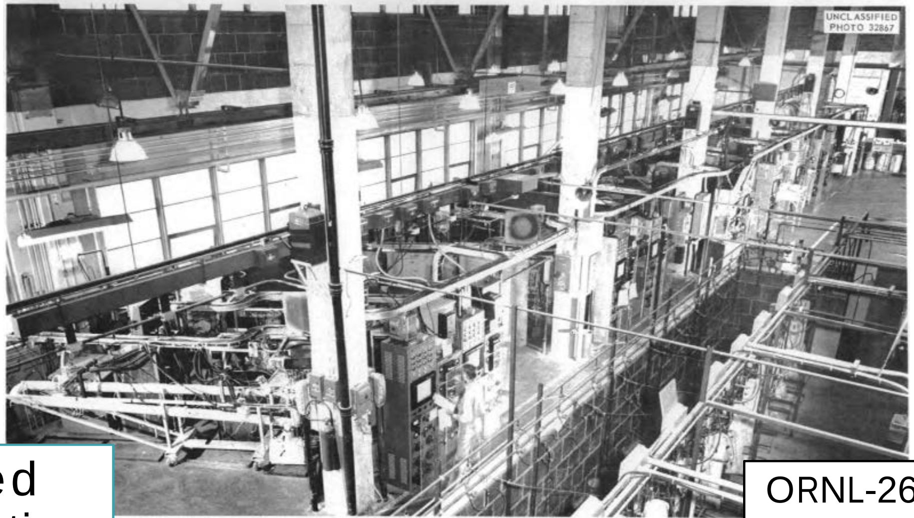
PWAR-1 677 °C Core Shell Assembly



First ARE Critical Assembly Structure

ORNL Employed an Edisonian Approach to Corrosion Studies

- 24 molten salt forced convection loops had completed operation by August 1961
  - 9 Inconel 600
  - 15 Alloy N
  - 4 loops operated 20,000 h
  - 704°C hot leg temperature typical
- Also operated refractory metal loops to evaluate non-salt coolants
  - Nb-1Zr / Pb loop



Forced convection loops

ORNL-2684

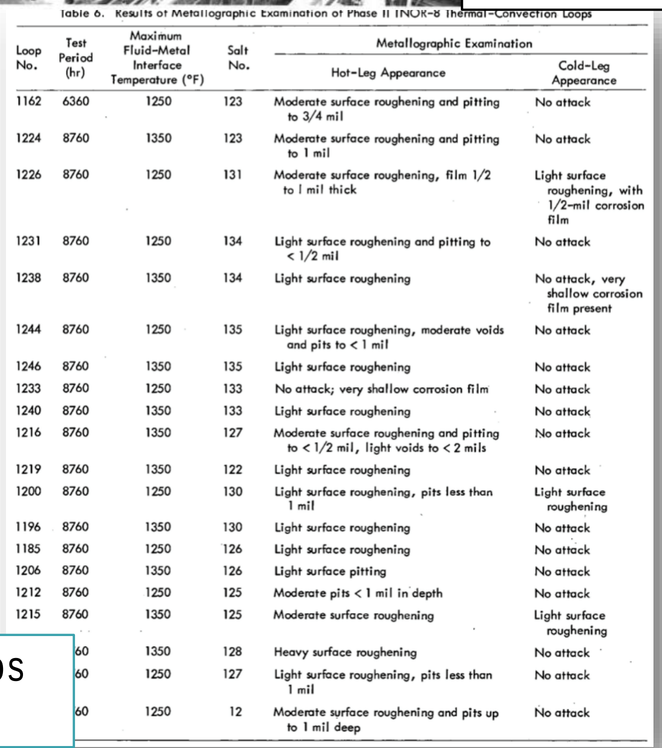


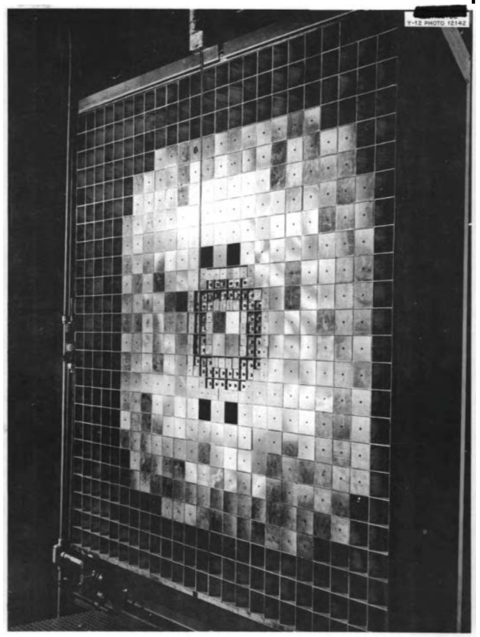
Table 6. Results of Metallographic Examination of Phase II INOR-8 Thermal-Convection Loops

Loop No.	Test Period (hr)	Maximum Fluid-Metal Interface Temperature (°F)	Salt No.	Metallographic Examination: Hot-Leg Appearance	Metallographic Examination: Cold-Leg Appearance
1162	6360	1250	123	Moderate surface roughening and pitting to 3/4 mil	No attack
1224	8760	1350	123	Moderate surface roughening and pitting to 1 mil	No attack
1226	8760	1250	131	Moderate surface roughening, film 1/2 to 1 mil thick	Light surface roughening, with 1/2-mil corrosion film
1231	8760	1250	134	Light surface roughening and pitting to < 1/2 mil	No attack
1238	8760	1350	134	Light surface roughening	No attack, very shallow corrosion film present
1244	8760	1250	135	Light surface roughening, moderate voids and pits to < 1 mil	No attack
1246	8760	1350	135	Light surface roughening	No attack
1233	8760	1250	133	No attack; very shallow corrosion film	No attack
1240	8760	1350	133	Light surface roughening	No attack
1216	8760	1350	127	Moderate surface roughening and pitting to < 1/2 mil, light voids to < 2 mils	No attack
1219	8760	1350	122	Light surface roughening	No attack
1200	8760	1250	130	Light surface roughening, pits less than 1 mil	Light surface roughening
1196	8760	1350	130	Light surface roughening	No attack
1185	8760	1250	126	Light surface roughening	No attack
1206	8760	1350	126	Light surface pitting	No attack
1212	8760	1250	125	Moderate pits < 1 mil in depth	No attack
1215	8760	1350	125	Moderate surface roughening	Light surface roughening
[illegible]	[illegible]	1350	128	Heavy surface roughening	No attack
[illegible]	[illegible]	1250	127	Light surface roughening, pits less than 1 mil	No attack
[illegible]	[illegible]	1250	12	Moderate surface roughening and pits up to 1 mil deep	No attack

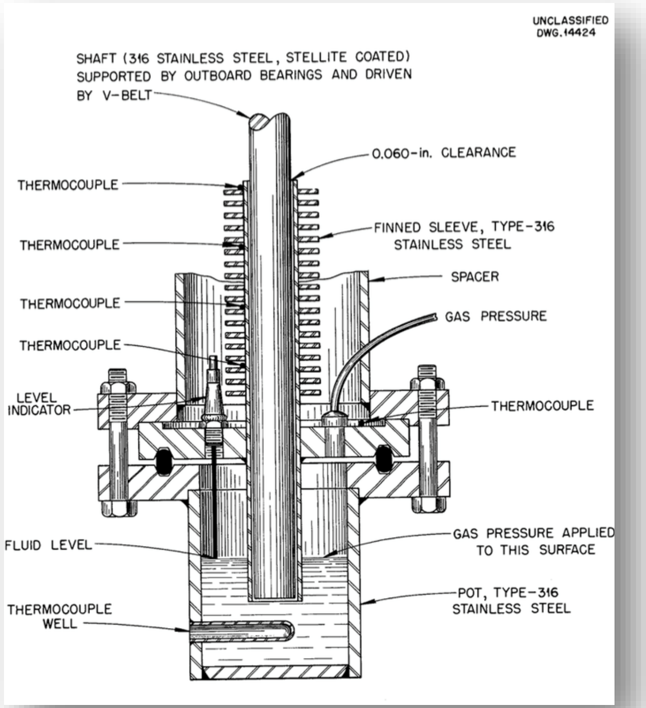
Alloy N natural convection test loops reported on in the first half of 1964

ARE Was Designed and Operated Prior to the Development of Computer Models

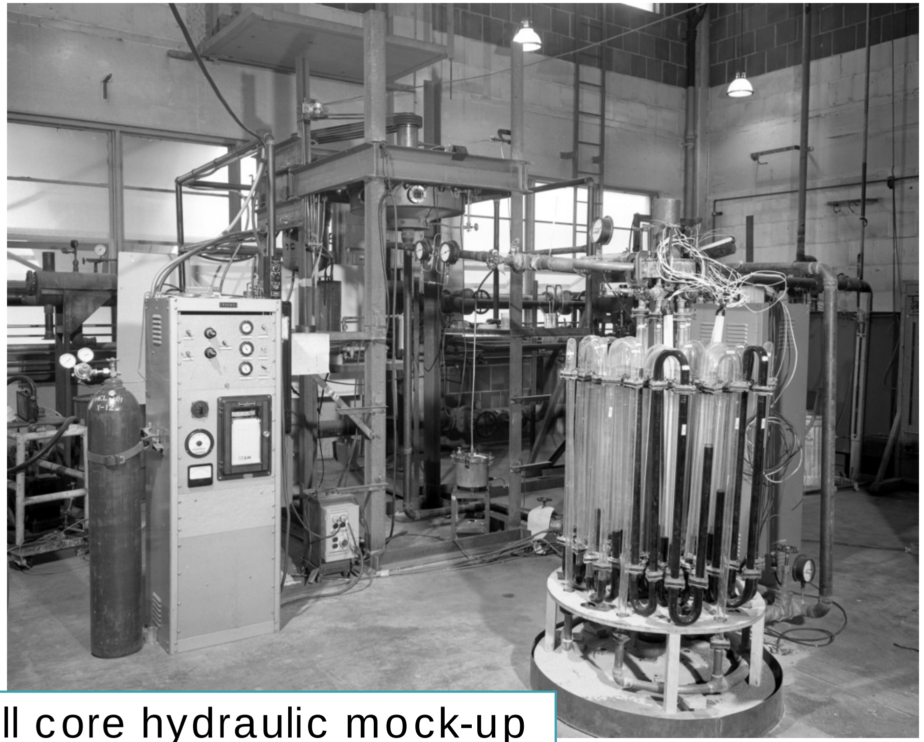
- Hydraulic design was performed using a mock-up
  - Both water and simulant fluids (water - glycerin and 1-1, 2-2 bromoethane) employed
- Components were classically designed and proven experimentally
- Reactor physics were based on hand calculations validated by critical experiments
  - ARE's first low temperature critical experiment assembly started in June 1952 and went critical August 21, 1952



Second ARE critical experiment



Freeze seal tester

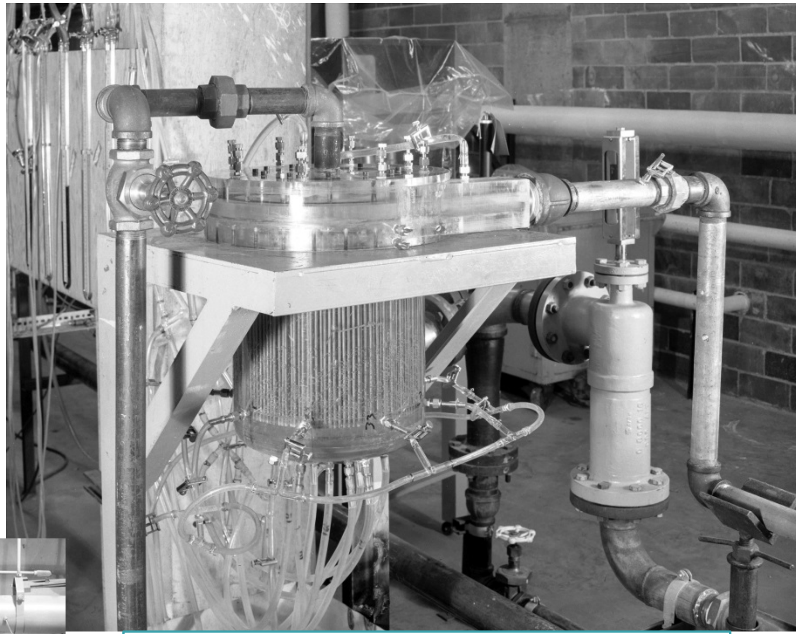


Full core hydraulic mock-up

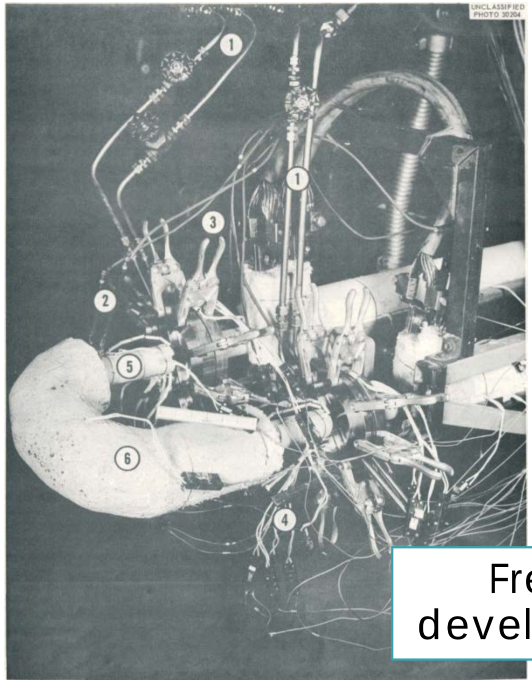
OAK RIDGE National Laboratory

MSRE's Design Was Developed and Validated Through Extensive Physical Modeling

- 1/5 scale core hydraulic model matched Reynolds number between water and fuel salt
- Full scale core model first run with water then with simulant fluid to match Reynolds numbers
- Specialized test rigs were developed for all components



1/5 scale MSRE core model

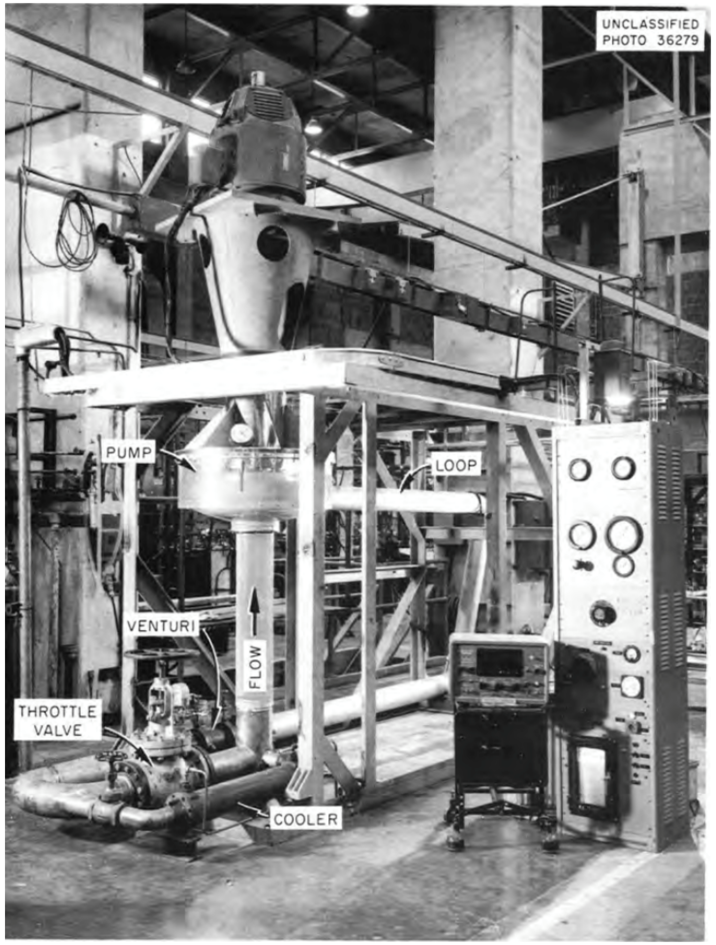


Freeze flange development test rig

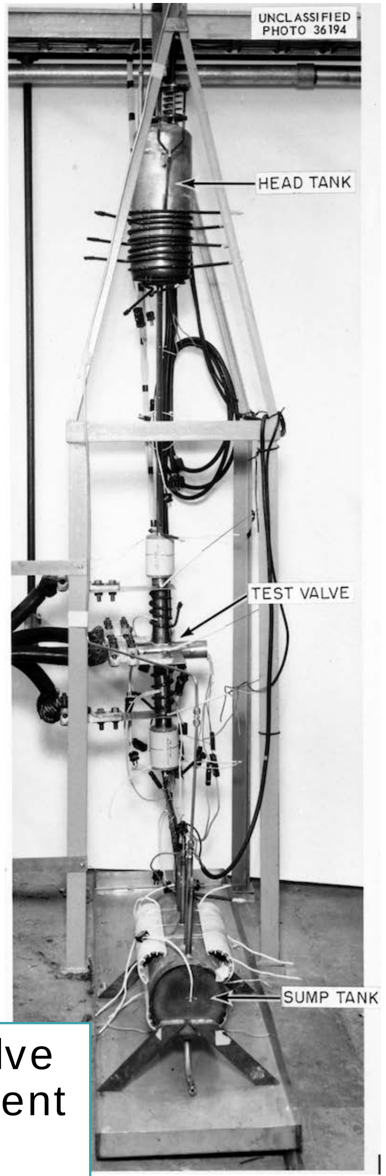
Full scale MSRE core model

MSRE's Components Were Developed Through Classical Design Methods and Validated Through Extensive Testing

- Primary heat exchanger modeling was generally successful (ORNL CF-61-4-1)
  - Water flow testing of primary heat exchanger resulted in removing the four outer U-tubes to decrease shell (fuel salt) side pressure drop
  - Fuel salt thermal conductivity data employed in model was sufficiently off to decrease the capability from 10 to 8 MW
- Pump design evolved from ARE designs
  - Limitations arose from available materials
- Freeze valves had multiple design options evaluated experimentally



MSRE fuel pump water test stand



Freeze valve development test rig

MSRE Operated Remarkably Successfully for a First of a Kind Reactor

- First criticality to conclusion of nuclear operation spanned 4.5 years
  - Salt operations began 9 months prior to criticality

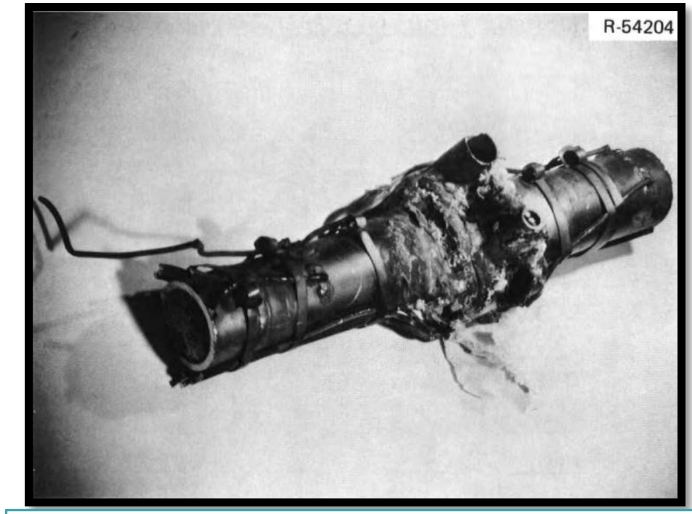
Full power	13,172 h ^{235}U 9,005 h ^{233}U 4,167 h
Fuel salt circulation time	21,788 h
Coolant salt circulation time	26,076 h
Availability during planned reliability testing period (final 15 months with ^{235}U)	86%
Availability during final runs with ^{235}U	98.6%
Availability during final runs with ^{233}U	99.9%

So far the Molten Salt Reactor Experiment has operated successfully and has earned a reputation for reliability.
USAEC Chairman Glenn T. Seaborg

Source: ORNL-TM-3039

MSRE Did Encounter Issues During Operation

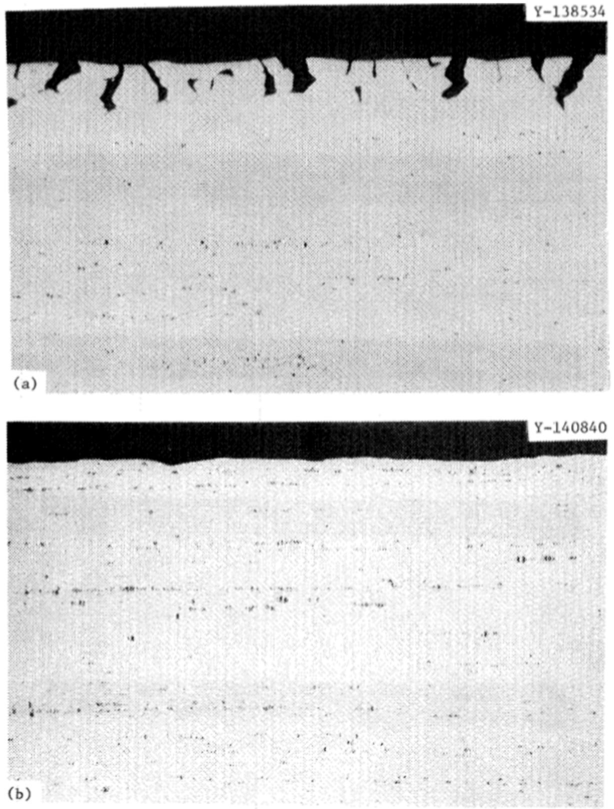
- Reactor vessel progressively embrittled due to neutron damage
- Drain tank isolation freeze valve cracked during its final cycle due to a field modification
  - Stiffening the air-cooling housing prevented pipe flexing
  - Xenon, iodine, krypton, and noble metals detected in reactor cell
- Small, continuous leak of lubricating oil into fuel pump causing foaming
  - Periodic plugging of offgas system
  - Fixed with larger filter
- Pump-entrained gas caused sporadic (about 10 times/h) increases in reactor power (~5–10%) for a few seconds
- Control rod failed scram test due to snagging on thimble
  - Experimental ‘rod-jogger’ stuck control rod in out position during a pseudo random binary sequence test
  - Power level ramped up then decreased without intervention



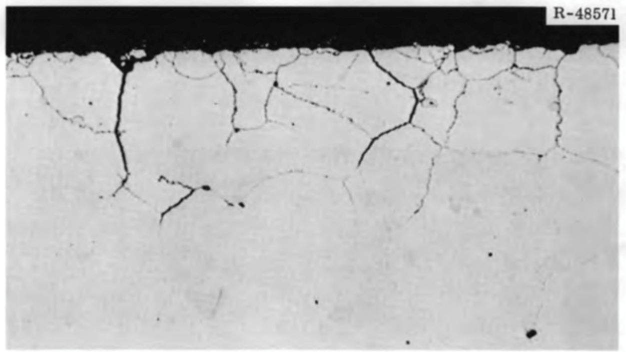
Bottom of cracked freeze valve

Salt-Wetted Alloy N Surfaces in MSRE Exhibited Tellurium-Assisted Surface Cracking

- Would be unacceptable for multi-decade lifetimes for thin-walled components
- Tensile testing of Alloy N surveillance specimens from the MSRE produced cracks in the grain boundaries connecting to the salt-exposed surfaces containing tellurium
- Intergranular embrittlement can be reduced by adding 1–2% niobium to Alloy N or by maintaining the salt in reducing conditions

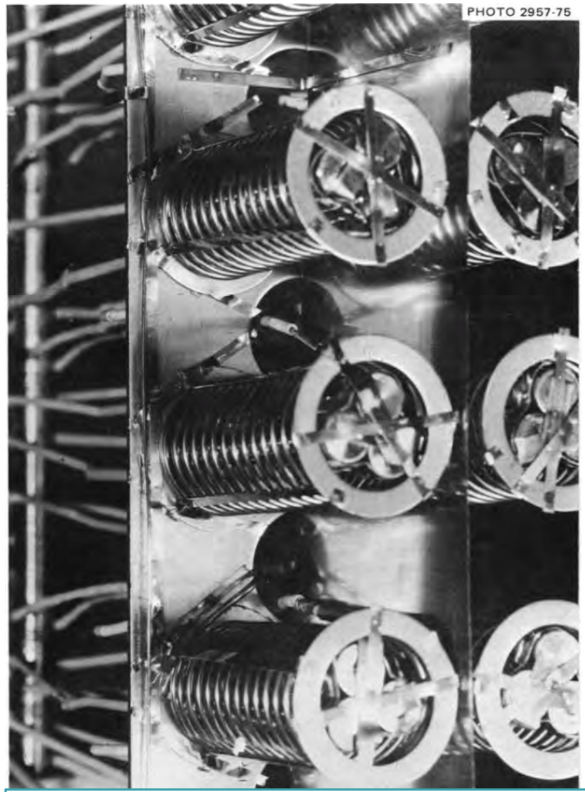


Alloy N exposed to MSRE fuel salt (500 h, 700°C) containing tellurium (a) oxidizing, (b) reducing – 100x



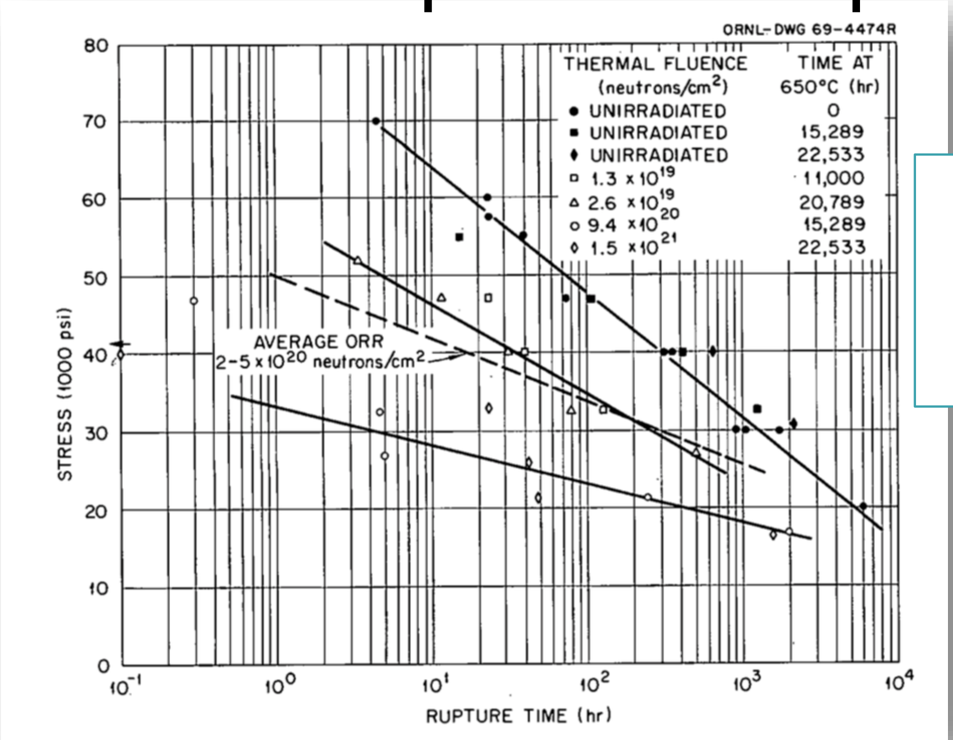
Typical microstructure of Alloy N after exposure to MSRE core for 22,533 h at 650°C – 500x

Niobium-Modified Alloy N Was Developed in Response to MSRE Embrittlement



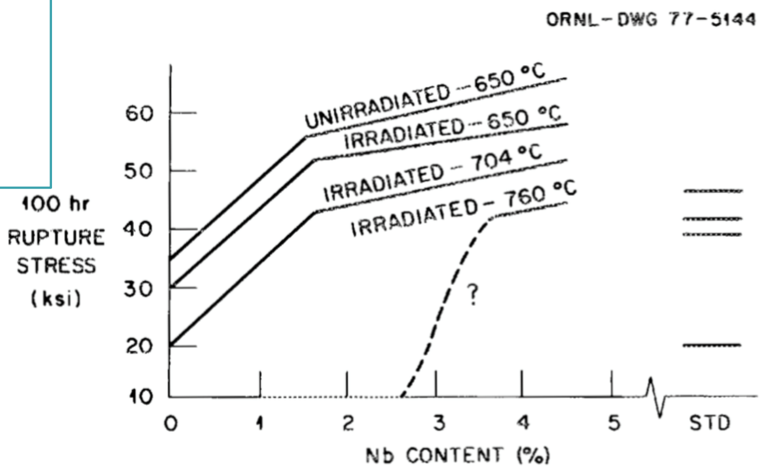
Cluster of modified Alloy N creep specimens prior to irradiation

All niobium-modified Alloy N specimens irradiated at 650°C had rupture lives in excess of those of standard unirradiated Alloy N



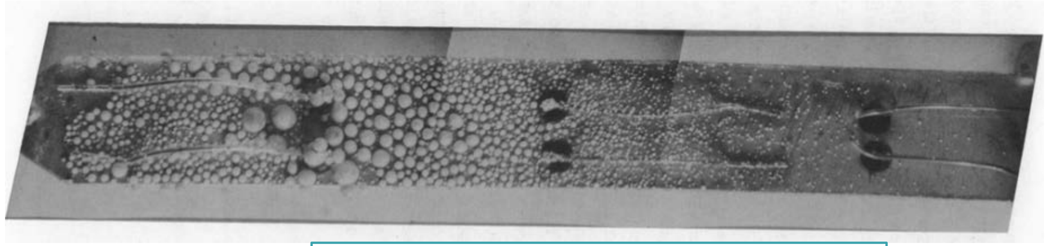
Stress rupture properties of MSRE surveillance specimens

Influence of Nb on stress rupture properties



Spray, Mist, Bubbles, and Foam Were All Present in MSRE Pump Bowl

- Fuel pump bowl incorporated spray ring (15.9 L/s – 4% recirculation from pump discharge) to disengage xenon
- Spray produced mist of salt droplets, which drifted to offgas line (few g/month) necessitating periodic cleanout
- Jets drove bubbles into pump salt pool
  - Changed density of salt shifting bubbler level measurements
- Salt frothed into overflow line (enhanced by hydrocarbons from pump oil leak)
  - Larger salt overflow following beryllium additions
- Small changes in pump speed or salt properties changed volume fraction of gas in core (0.02–0.7%)



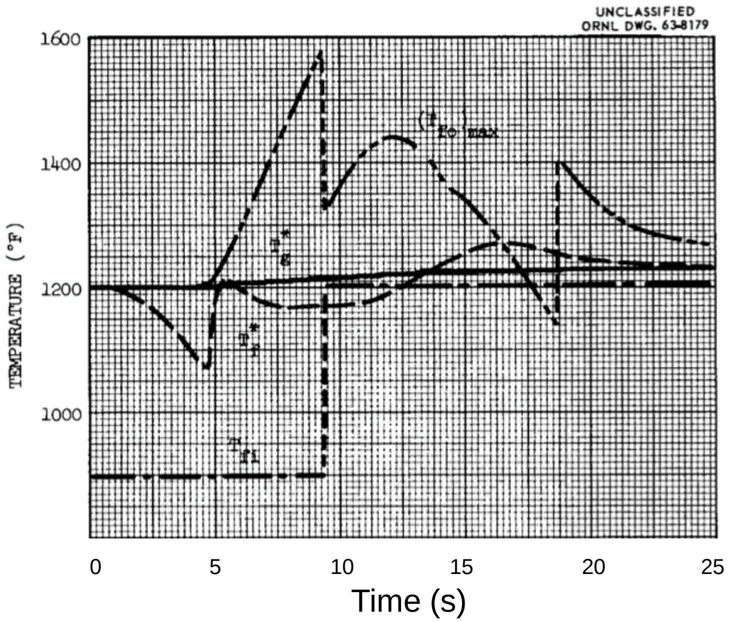
Salt droplets on metal strip exposed to MSRE pump bowl for 10 hours

MSRE Filling and Startup Procedures Were Straightforward

- UF_4-LiF salt added to barren carrier salt for initial startup
- 10^9 n/s source employed for approach to critical
  - Inherent neutron sources not sufficient
  - Movable BF_3 chambers for startup range flux measurements
- Fuel system purged with helium to lower cover gas oxygen to < 100 ppm
- Molten salts moved pneumatically
- System was preheated to 650°C prior to filling
- General procedure steps
- Thaw freeze valves, fill coolant loop, start coolant pump
- Thaw freeze valve, fill fuel loop with flush salt, start fuel pump
- Drain flush salt, empty overflow tank, and freeze freeze valve
- Thaw freeze valve, run rod drop and safety tests
- Fill fuel loop with fuel salt, freeze drain valve, start fuel pump

MSRE Designers Employed Computational Models to Solve Coupled Neutron and Fuel Salt Transport Equations

- MURGATROYD code logic was developed and validated for Aqueous Homogeneous Reactor design
  - Extended to provide separate graphite heat capacity
  - Single point, single energy group, seven delayed neutron precursor groups
  - Employed for both design and safety calculations
  - Beta effective was used based upon the fraction of the time fuel is in the core
- ZORCH code developed that includes axial spatial dependence in fuel and graphite temperature to more accurately represent transient responses
  - Shows that no damage would be anticipated even for unrealistic transients
  - Maximum fuel temperature anticipated ~850 °C (< 5 seconds) for unprotected cold slug addition

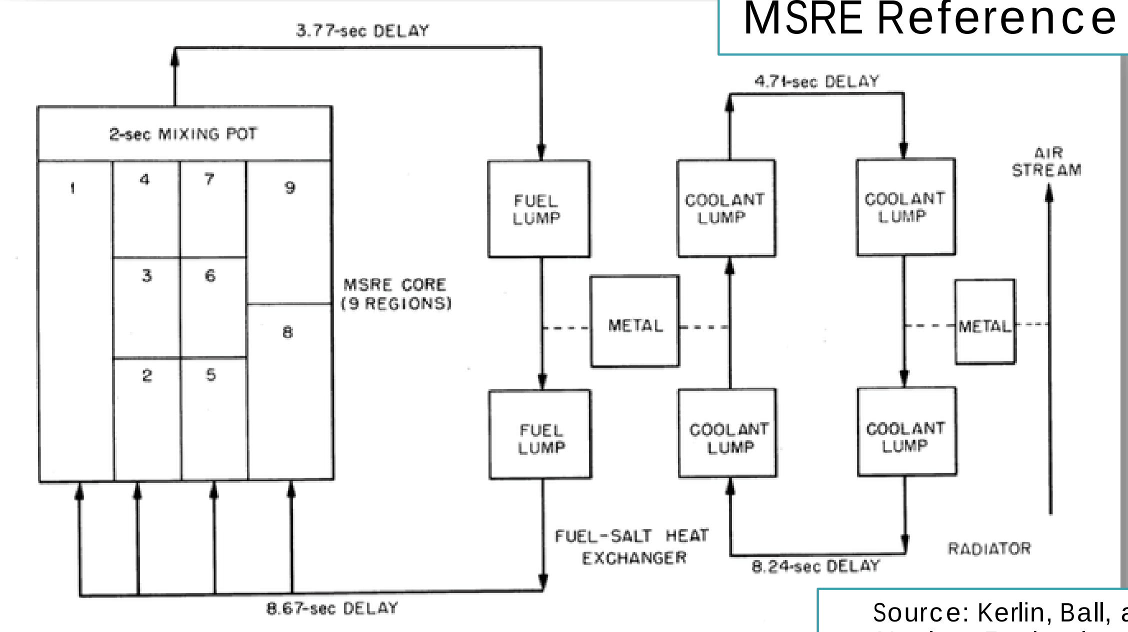


ZORCH temperature prediction for unprotected cold slug accident

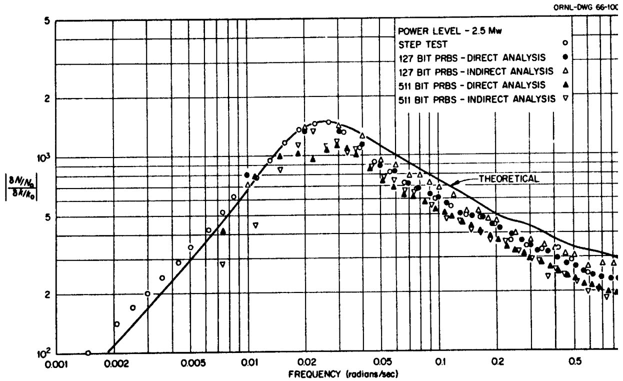
Dynamic Stability Tested at Low Power Before Full-Power Operations Began

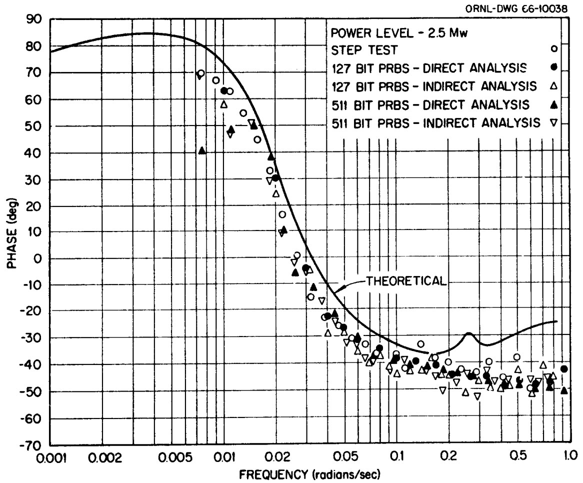
- Dynamic plant model predicted stable operation – which was confirmed using low power testing
  - 44th - order system matrix with 4 time delays for heat convection and 6 time delays for precursor circulation
  - Solved with MATEXP Code
- Main conclusion – system has no operational stability problems and its dynamic characteristics were as predicted

MSRE Reference Model

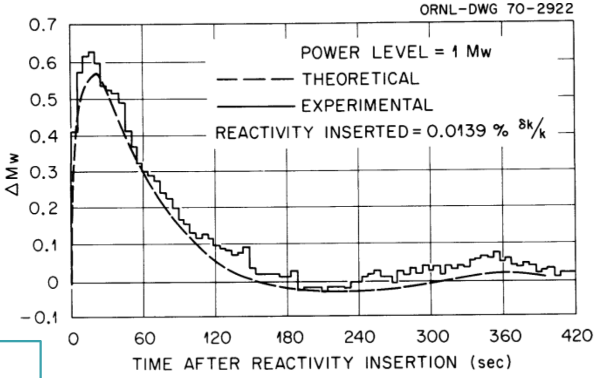


Source: Kerlin, Ball, and Steffy Nuclear Technology 10, 1971





ORNL-TM-1647



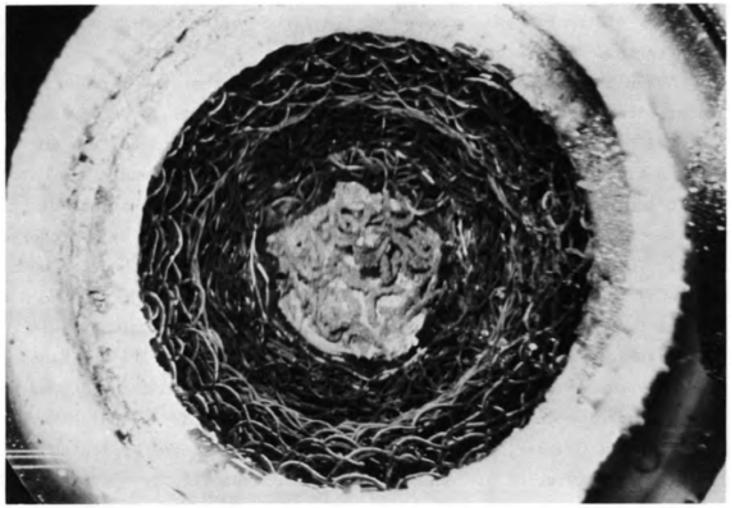
ORNL-TM-2997

MSRE Operating Power Was Determined by Heat Balance Measurements

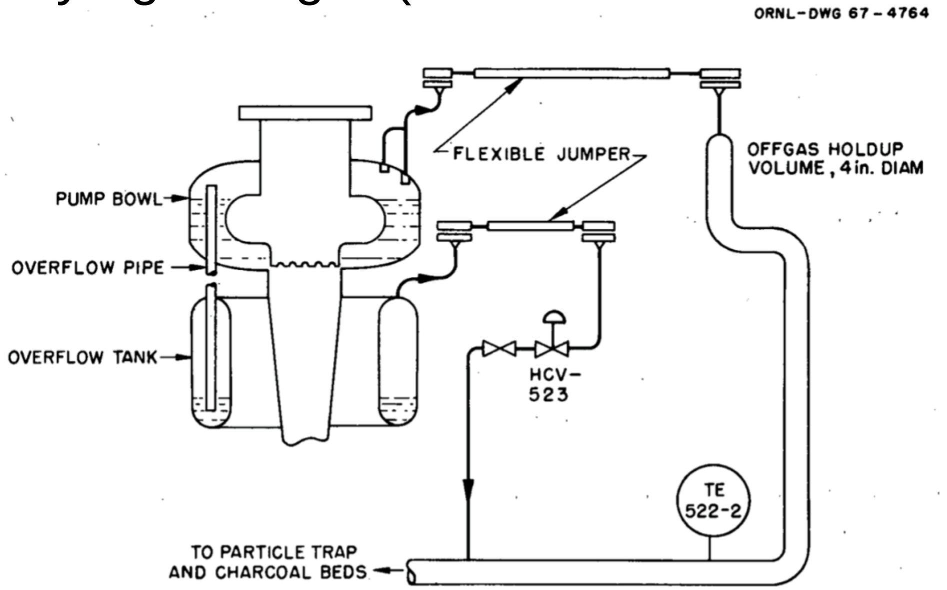
- Heat balance measurements indicated full power of 8.0 MW
- Isotopic changes in U and Pu in the fuel salt indicate a 7–10% lower full power (7.34 MW)
  - Appears that the coolant salt flow measurement was primary source of error
- Heat transfer capabilities of both fuel to coolant salt and coolant salt to air radiator were below initial design predictions
  - Erroneous physical property data (primarily thermal conductivity) used in primary heat exchanger
  - Improper selection of air film temperature was source of error in radiator design
- No decrease in heat exchanger performance over course of operations

Offgas System Posed Challenges Due to Plugging Exacerbated by Oil Leak

- Lubricating oil leaking from pump seal caused issues with filters, check valves, and control valves
- Hydrocarbons tended to have gaseous fission products stick to them and in turn deposit on the particle filters thus clogging the system
- Problem was substantially reduced by employing a larger (15 versus 10 cm diameter), redesigned particle trap



MSRE Mark 1 Offgas Particle Trap

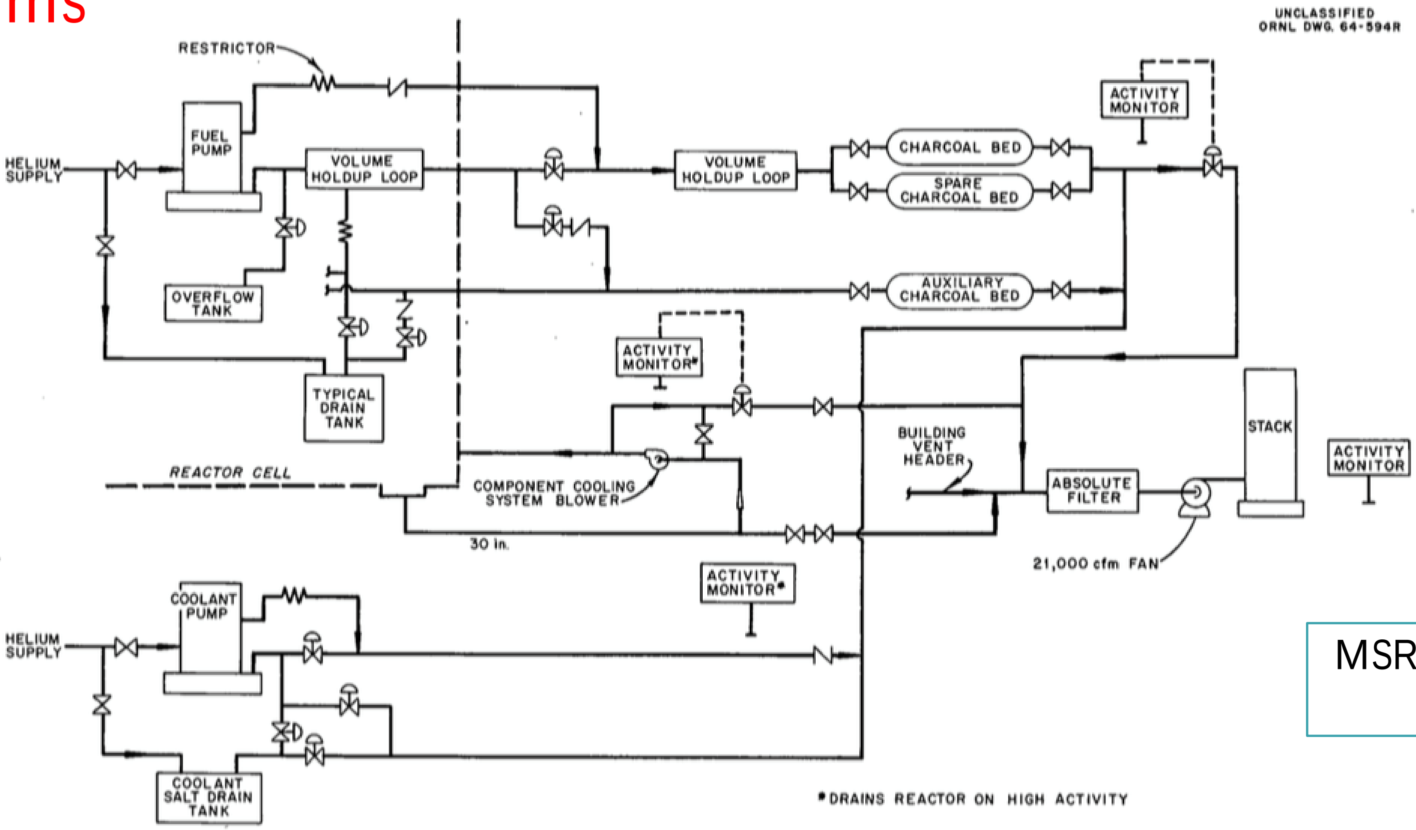


Offgas Piping Near Pump and Overflow Tank

Source: ORNL-TM-3039

Charcoal Bed Entrance Plugged Periodically

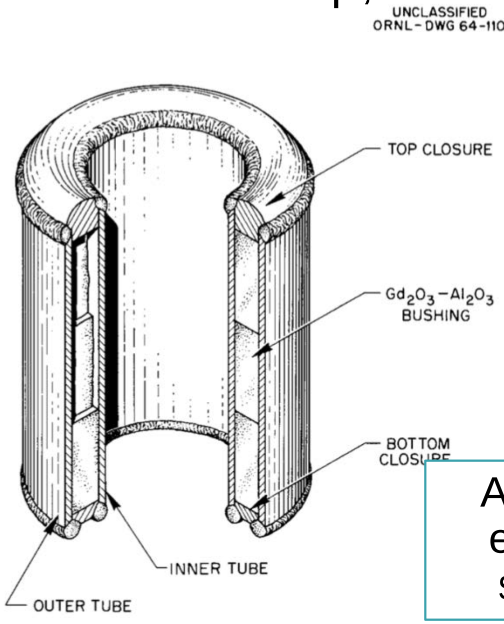
- Appears to also have been related to hydrocarbon vapors
  - More solid decay products are borne in the holdup volume between particle filter and the entrance to charcoal bed
- Key recommendation: Avoid use of hydrocarbon lubrication in all salt-connected systems



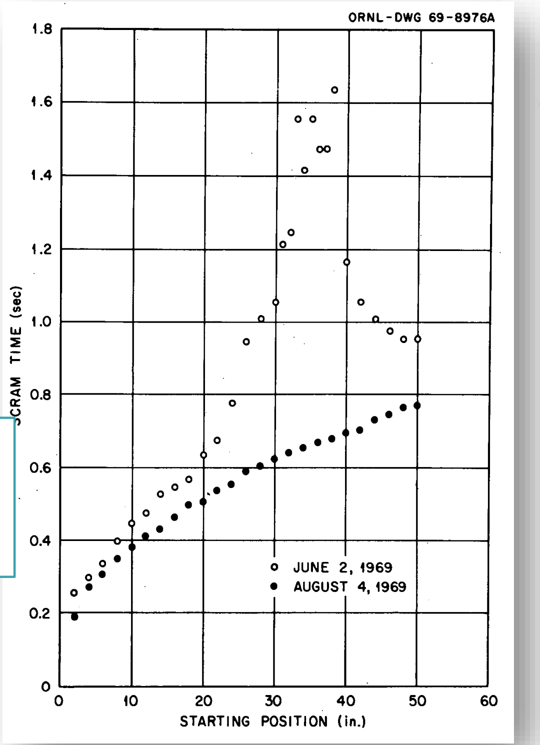
MSRE Cover Gas Flow Schematic

Absorber-Based Control System Generally Performed Well

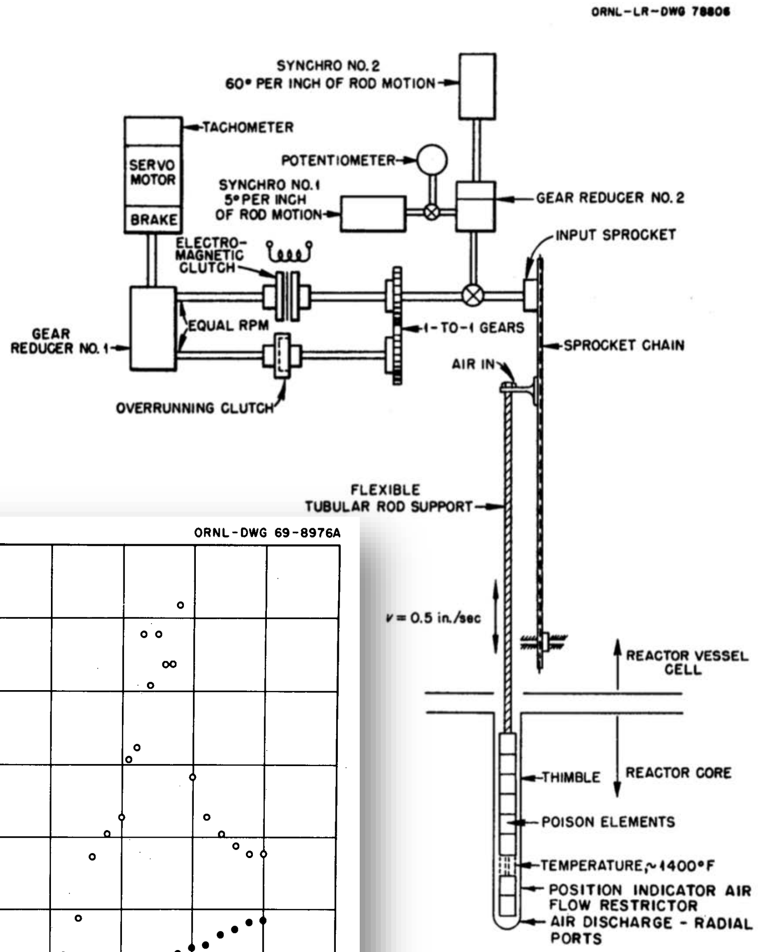
- Over 3000 scram tests performed with only one failure
  - Rod 3 stuck at 35 inches in channel on June 1, 1969
- Mechanical wear was resulting in progressively longer drop times
  - Rods were used to shift power levels, to compensate for fission product buildup, and for fueled shutdown



Absorber element section



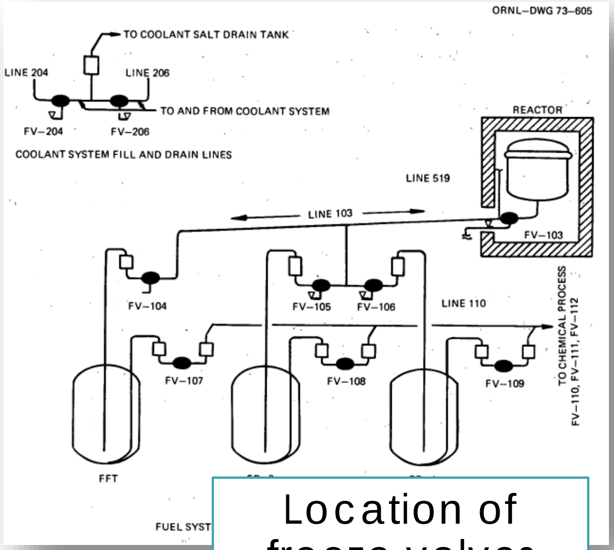
Stuck element and normal drop



Absorber control schematic

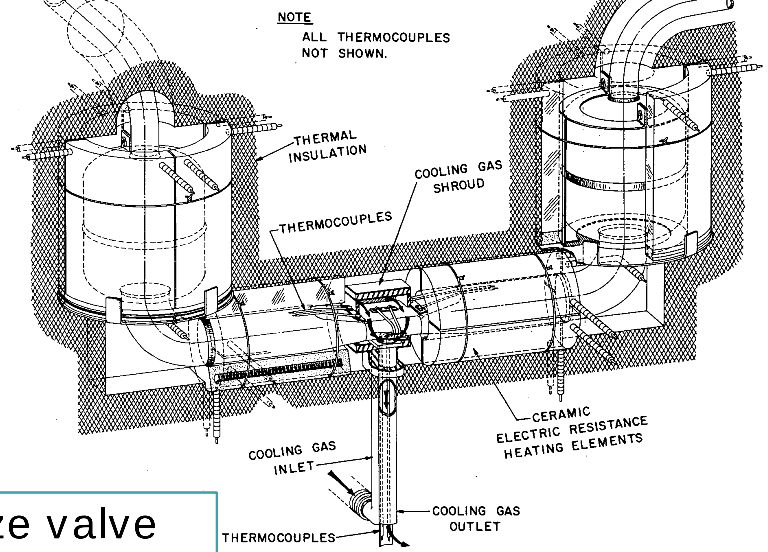
Freeze Valve Operations Improved Over Time

- 12 freeze valves located throughout the plant
- Freeze valves preferred since reliable mechanical closure valve unavailable
  - Development began in 1960
  - Operations not hampered by “slow” response and lack of “off-on” functionality
- Three operational modes
  - Deep frozen: heaters adjusted to maintain 400–500°F without cooling air
  - Thawed: heaters adjusted to maintain 1200°F without cooling air
  - Frozen: Heaters remained in thawed condition but cooling gas flow adjusted to hold just frozen to allow for rapid thaw
- Main fuel salt drain valve (FV-103) kept in frozen condition while operating and required to open in 15 minutes
  - One unscheduled drain caused by setpoint drift and untested operating procedure
- While initially some issues with incomplete freezing, extensive valve testing eventually resulted in improved operating procedure



Location of freeze valves

Source: ORNL-TM-3039

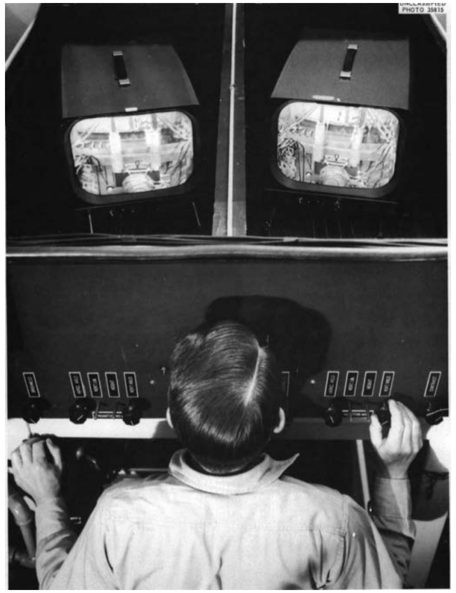


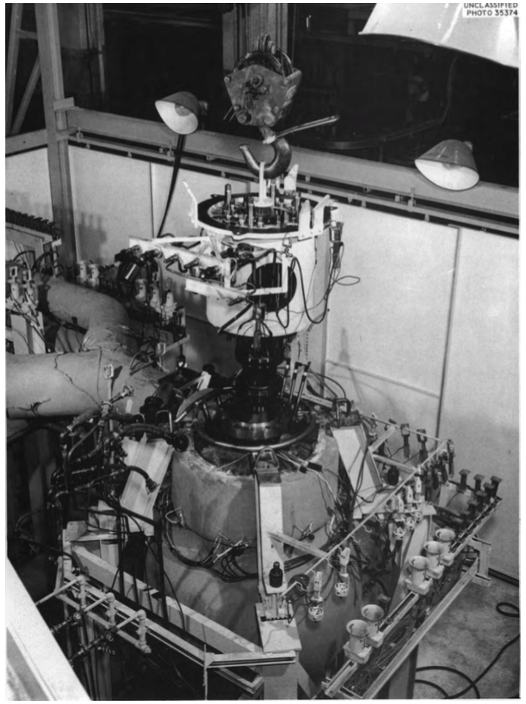
Freeze valve schematic

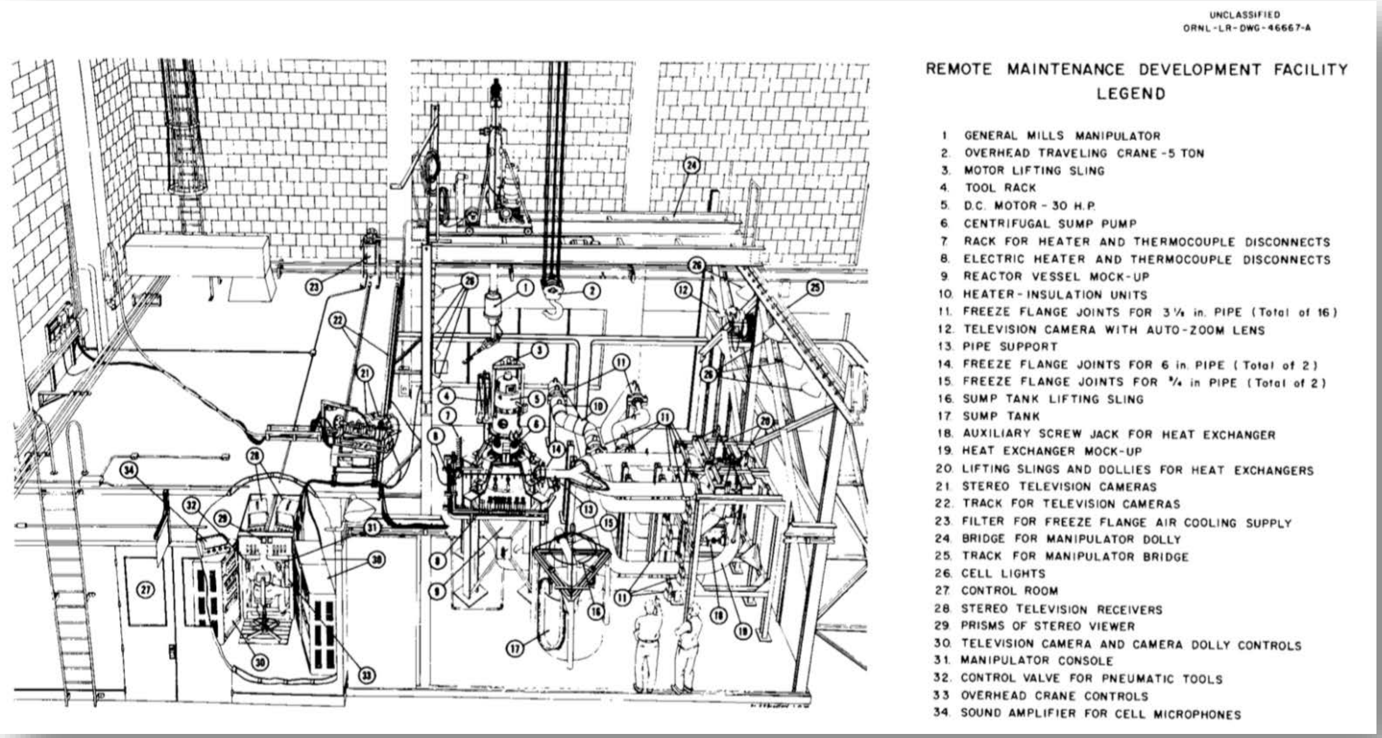
OAK RIDGE National Laboratory

Extensive Remote Maintenance Planning and Demonstration

- Remote maintenance mock-up facility created
  - 650°C mock-up of 20 MWt MSR
  - https://youtu.be/uHT-w2x6dDg
  - Tools, techniques, and procedures for replacing all major components including heat exchangers, fuel pumps, reactor core vessel, pipe preheaters, and piping sections developed and demonstrated



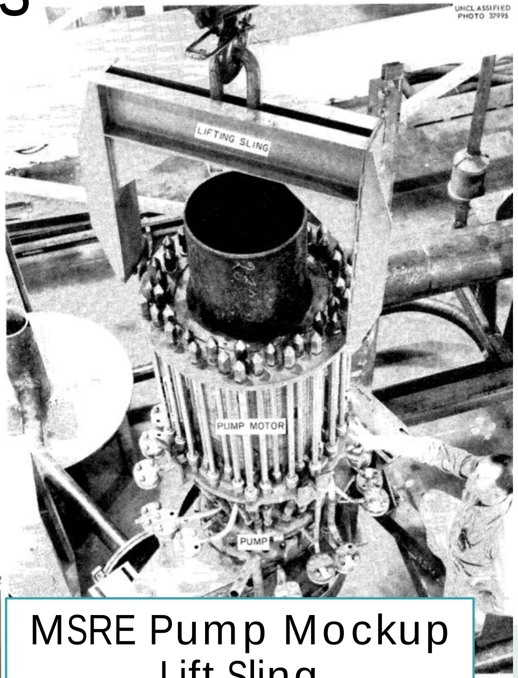




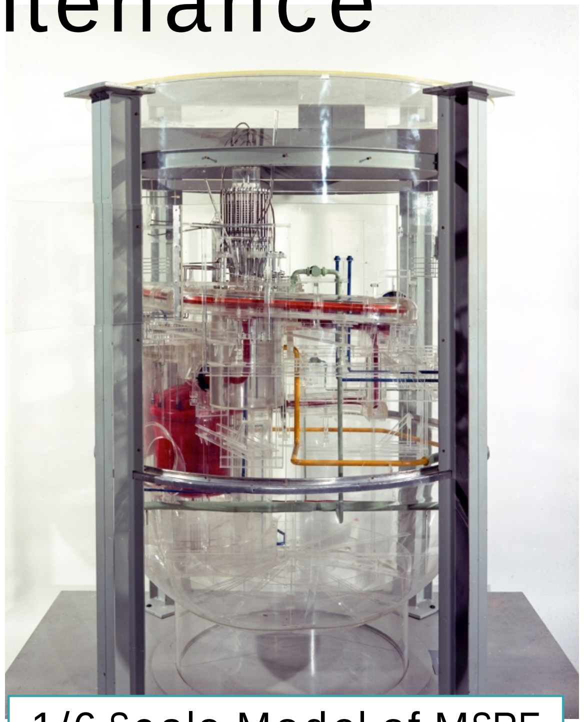
MSRE Also Included Substantial Remote Maintenance and Planning Support Facilities

Core Top While Drained Through Fisheye Lens

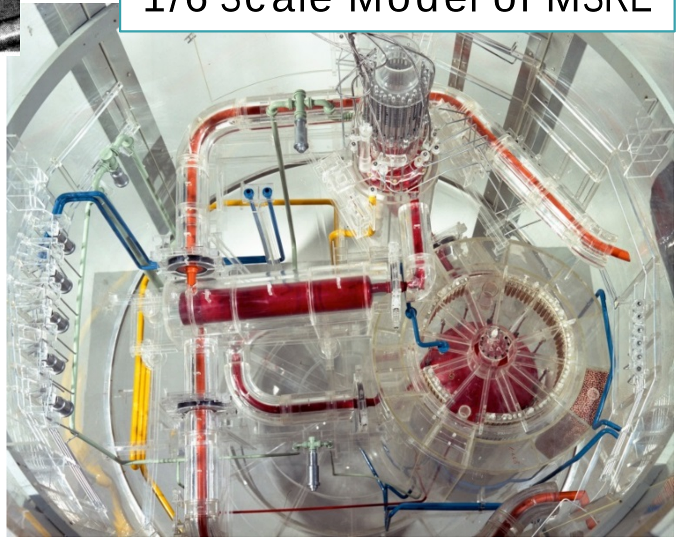
Fuel Pump Rotary Element Removed Following Run 3



MSRE Pump Mockup Lift Sling



1/6 Scale Model of MSRE



OAK RIDGE National Laboratory

MSRE Had Limited Core Structural Integrity Challenges

- Upper end of 6 or 7 graphite blocks from the periphery of the core had broken off sometime during operation
  - Likely explanation is that during cooldown salt was trapped between the top Hastelloy N retaining ring and the periphery graphite froze
  - Further thermal contraction of ring produced bending load
  - Pieces trapped by outlet screen
  - No significant perturbations to operations
- No damage to straightening vanes (most delicate flow control component) detected during operation

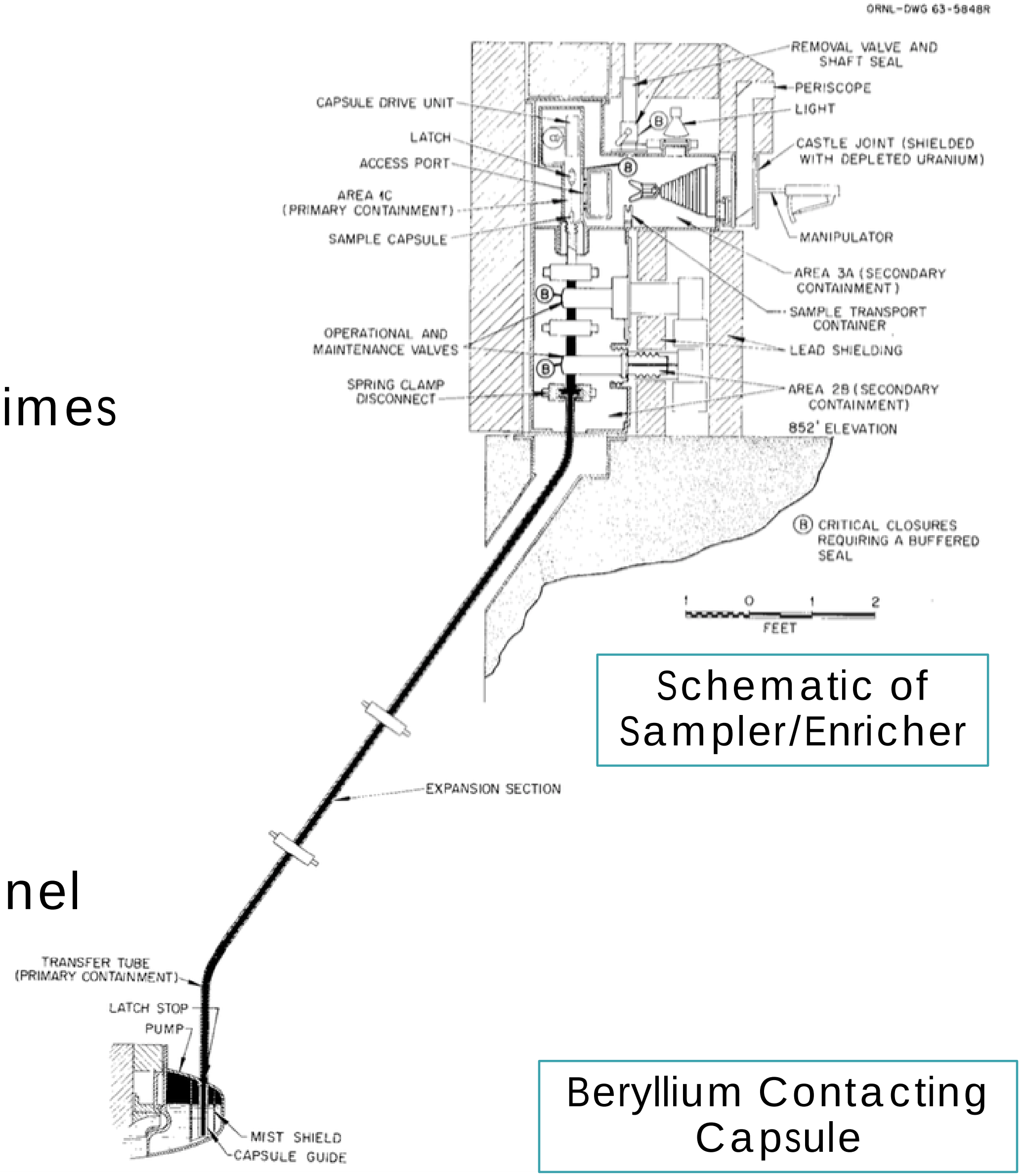
Welding flow straightening vanes on lower vessel head

Broken piece of graphite on top of MSRE core

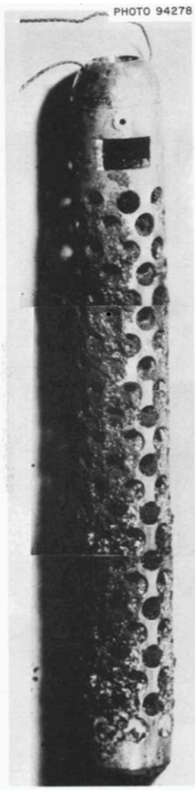
OAK RIDGE National Laboratory

Sampler-Enricher Employed to Remove Fuel and Add Fissile Material to Pump Bowl

- Could add poison in emergency situation
- Also added redox adjustment capsules
- Required maintenance/repair a number of times
- Capsules dropped a few times sometimes recovered with a magnet
- Significant maintenance activity to recover dropped capsules and latches
- Employed a two-barrier design/operational requirement to avoid contamination/personnel exposure
  - Had three barriers, two were always closed



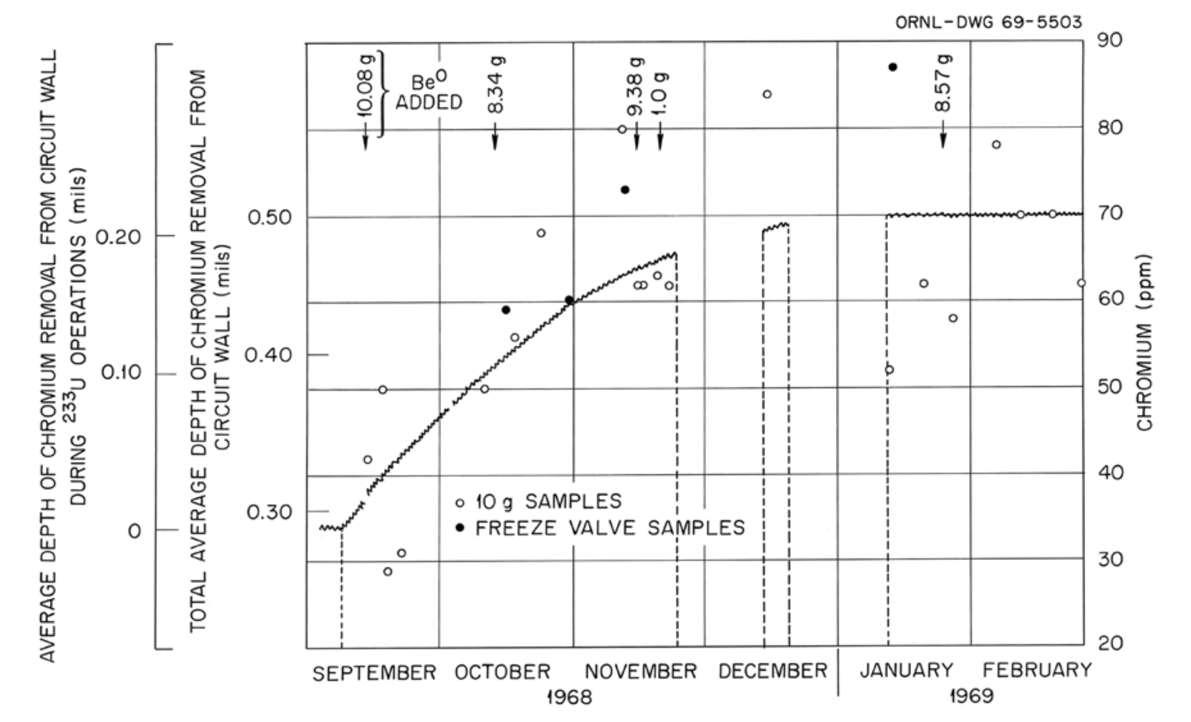
Schematic of Sampler/Enricher



Beryllium Contacting Capsule

Fuel Salt Redox Chemistry Was More Difficult to Control Using 233U

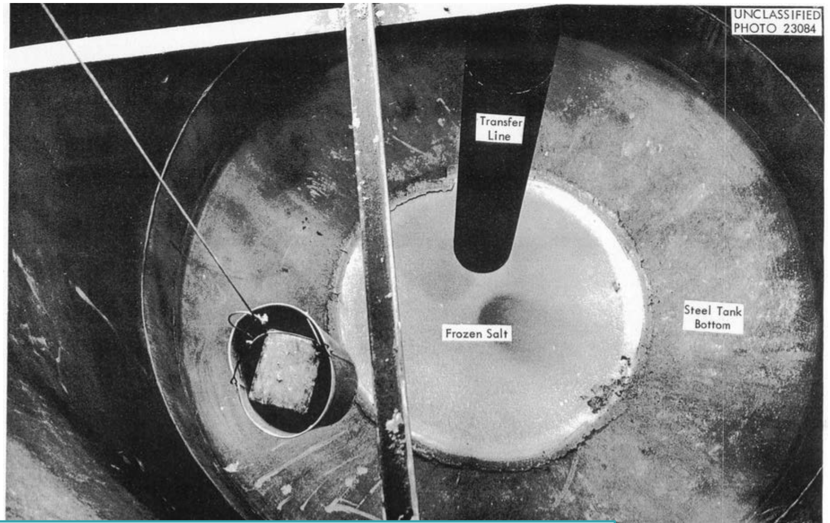
- Fissile material was changed from $^{235,238}UF_4$ (33% ^{235}U) to $^{233}UF_4$
  - Total amount of uranium in the ^{235}U fuel exceeded that contained in the ^{233}U fuel by sixfold
- Operation with ^{233}U fuel showed pronounced changes in fission product and corrosion chemistry as variations of the concentration of UF_3 in the fuel salt were made
  - Persisted until 28.80 g of Be were added
- After opening for maintenance the fuel salt became oxidizing with respect to the MSRE fuel salt containment for both ^{235}U and ^{233}U operation



Corrosion in MSRE Fuel Circuit Following ^{233}U Changeover

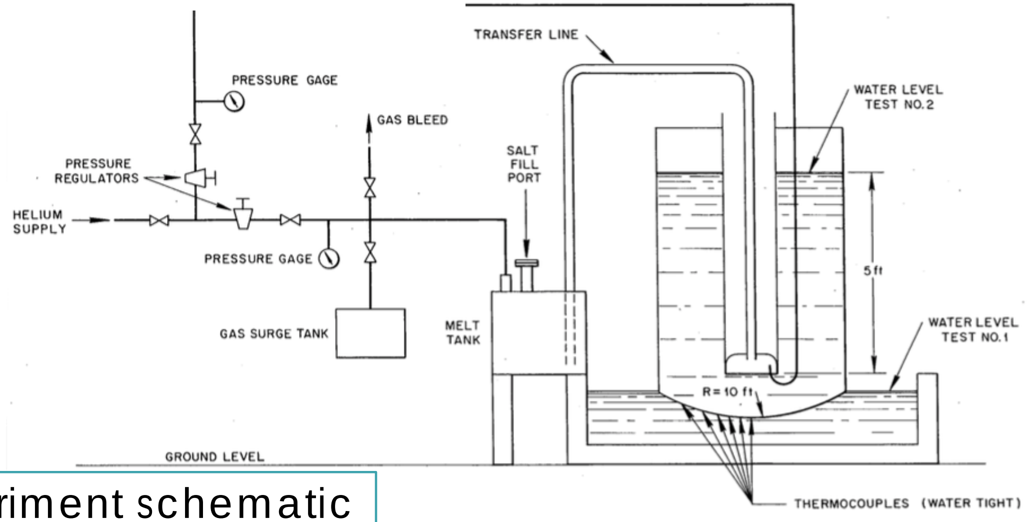
Potential for Significant Water Interaction Was Investigated Through Core Dump Tests

- Dump salt onto bottom of water cooled lower head to assess decay heat removal
- Dump irradiated fuel salt into water pool to assess radionuclide escape
- Test 1 – decay heat removal
  - Water adjacent to center of tank boiled briskly, but not violently enough to shake tank
- Test 2 – radionuclide release
  - 229 kg of unirradiated fluoride (11.5 NaF-46.5%LiF-42KF mol%) salt (including 2 kg of UF_4) heated to 816°C – water to 85°C
  - 99 grams irradiated Na_2UF_6 added – 15–30 mCi (~6 days after irradiation)
  - Salt forced into water tank in 45–50 seconds via helium overpressure
  - Violent boiling in tank – negligible amount of steam reached surface
  - No radioactivity was detected outside of the water



Interior of steel tank after test #1

ORNL CF-55-2-100



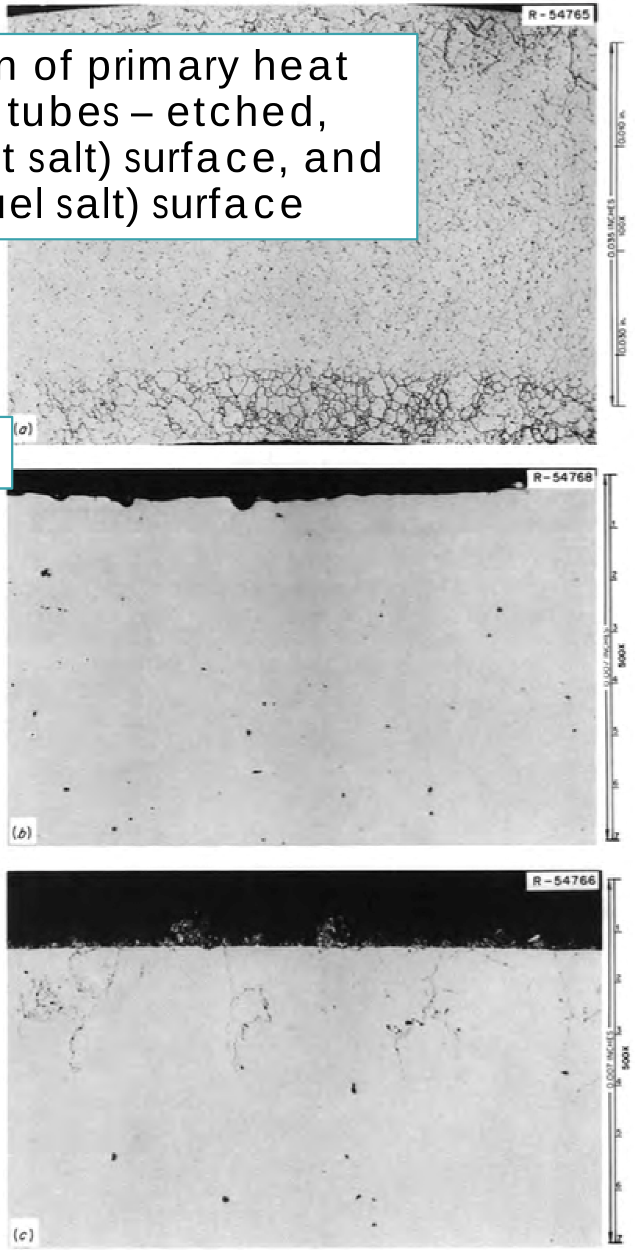
Experiment schematic

MSRE Heat Exchanger Tubes Were Cut Out for Post Operation Examination

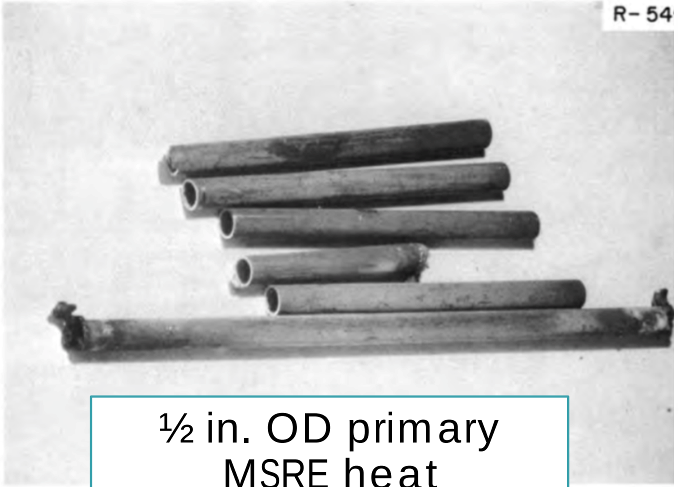
(a) Cross-section of primary heat exchanger tubes – etched, (b) inside (coolant salt) surface, and (c) outside (fuel salt) surface

- Fuel salt side exhibits characteristic minor tellurium corrosion
- Coolant salt side remains in good condition

(a)

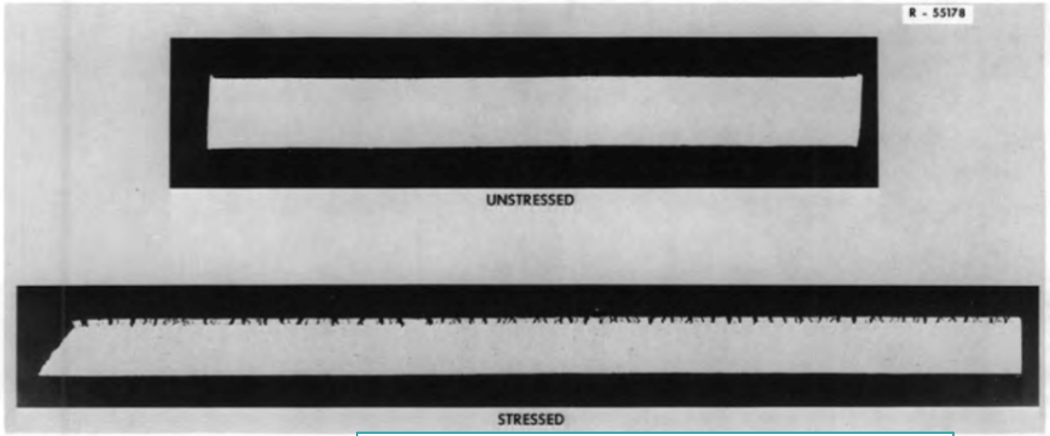


(c)



½ in. OD primary MSRE heat exchanger tubes

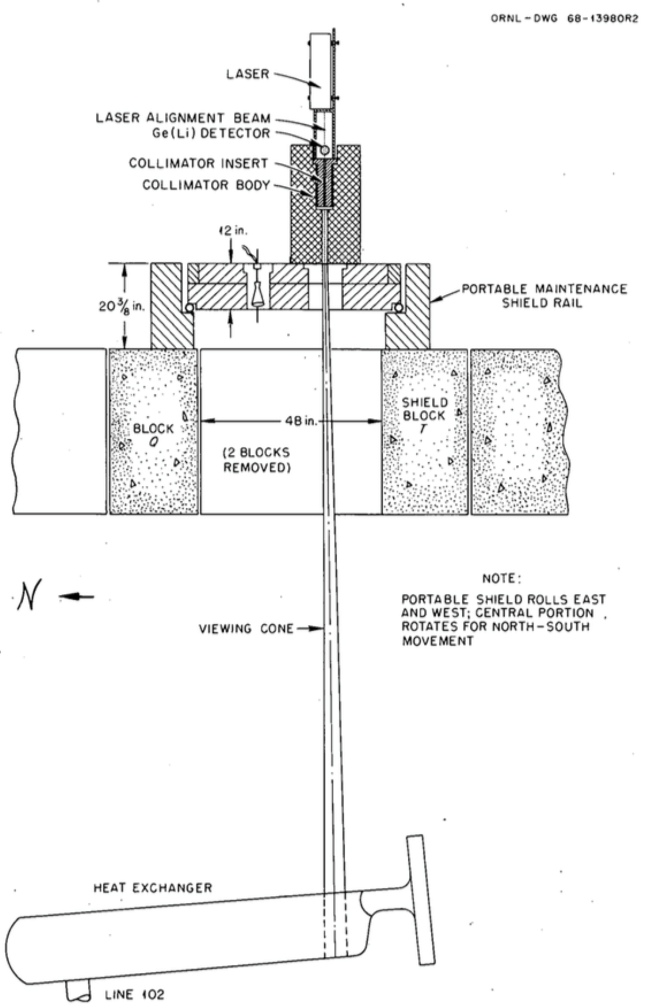
(b)



Unstressed and stressed fuel salt surface

Fission Product Distribution Was Tracked Throughout MSRE Operation

- MSRE fission products are distributed among fuel salt, metal and graphite surfaces, and cover gas system
  - Significant amounts were removed by fluorination during changeover to ^{233}U
- Substantial amount of fission product distribution data was generated from gamma scanning (ORNL-TM-3151) , post shutdown material sampling (ORNL-4865), and from pump bowl salt samples (ORNL-4658)



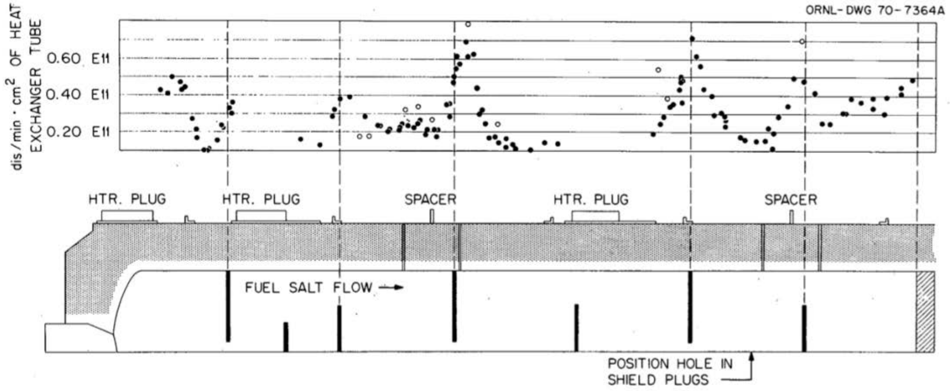


Fig. 7.23. Activity of ^{103}Ru at reactor shutdown on November 2, 1969, in the MSRE heat exchanger.

Primary Heat Exchanger Temperature Can Be of Concern Following Salt Draining Accidents

- Substantial portion of heat producing noble metal fission products may plate onto heat exchanger tube walls
  - Main competing process is plating out onto filters
- Following both fuel and coolant salt draining, fission product heating will cause temperature tube wall temperature to rise
- Peak temperature will depend on heat exchanger design details
  - Well insulated shell may result in unacceptable peak temperatures
  - MSBR design employed an inner shell to facilitate tube bundle replacement – significantly raised anticipated peak temperatures

ORNL-TM-3145, *Thermal Radiation Transfer of Afterheat in MSBR Heat Exchangers*

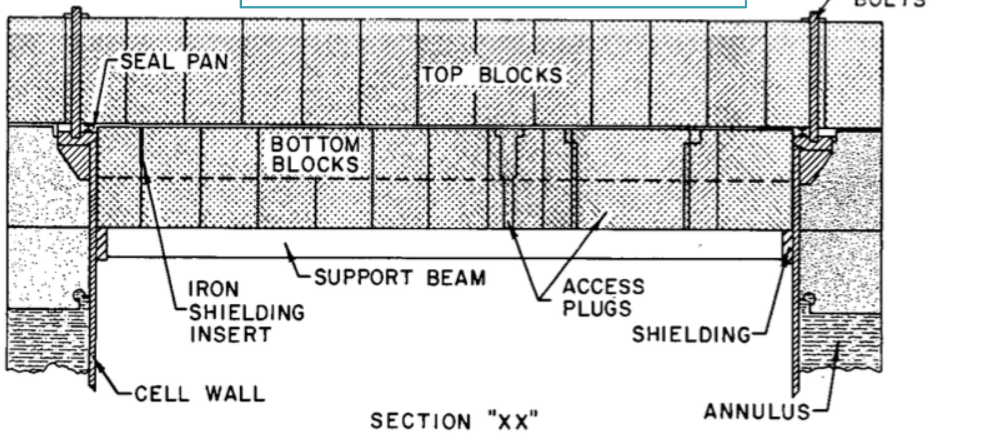
Instrumentation Had a Number of Issues; However, None Was Related to Salt Contact

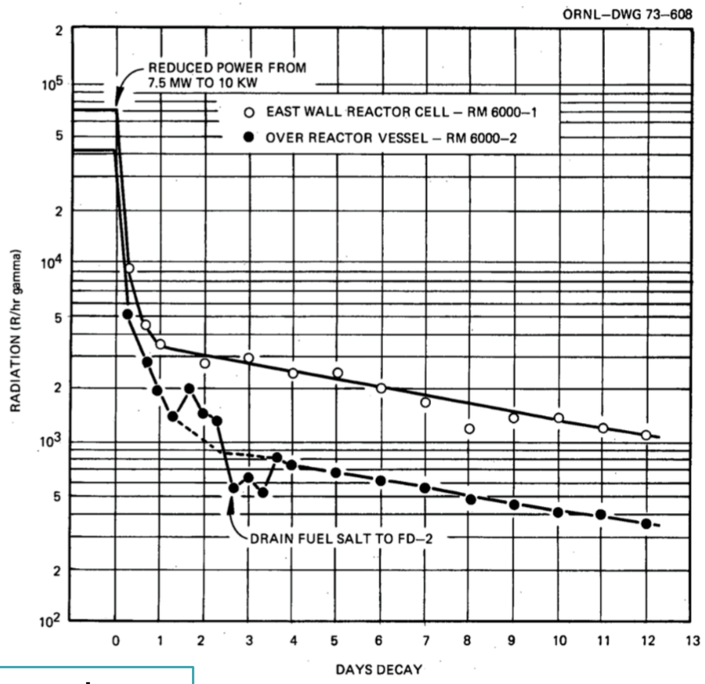
- Largely the instrumentation did not touch salt
- Instrumentation problems were typical of first-of-a-kind process plant deployments
  - Moisture ingress into fission chambers
  - Vibration sensitive switches on high temperature trip channels
  - Air leakage through insulation around differential pressure impulse line caused rapid temperature decrease whenever radiator air flow increased
  - 60 cycle noise pick on plant thermocouples
  - Oxidation of lead wires to pressure gauges due to high temperatures caused failures
  - Zero and span shifts occurred in flowmeter electronics

MSRE Had Extensive Biological Shielding

- MSRE biological shield was designed so that the dose rate would not exceed 2.5 mrem/h during normal operation at any point on the shield exterior located in an unlimited access area
- Gamma radiation levels in the reactor cell
  - 40,000 to 70,000 R/h at full power
  - 3,000 to 5,000 R/h upon a shutdown and drain

Upper Shielding Blocks



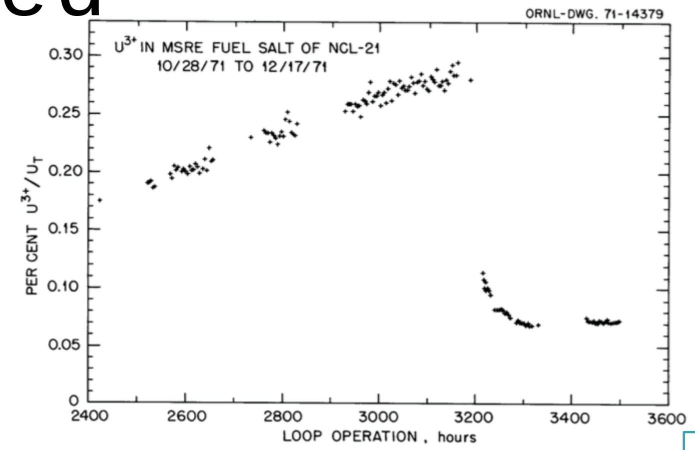


In-cell gamma dose

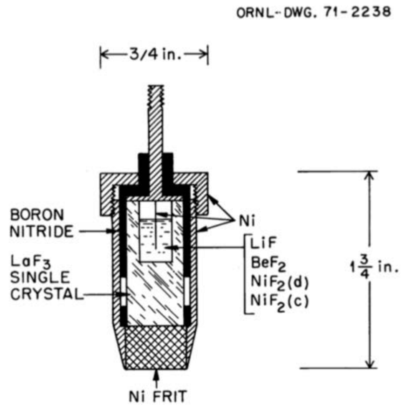
Optical and Electrochemical Fuel Salt Characterization Instrumentation Was Developed

- Spectrophotometer was employed to observe fission product content in MSRE fuel salt
- Electrochemical measurements showed the redox condition of nonradioactive MSRE fuel salt surrogate

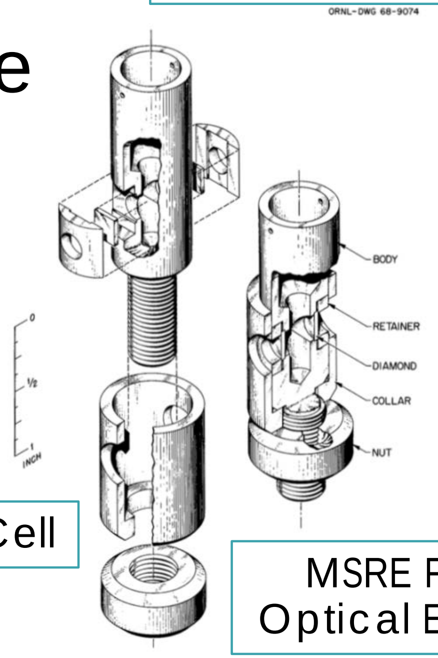
Source: ORNL-4812



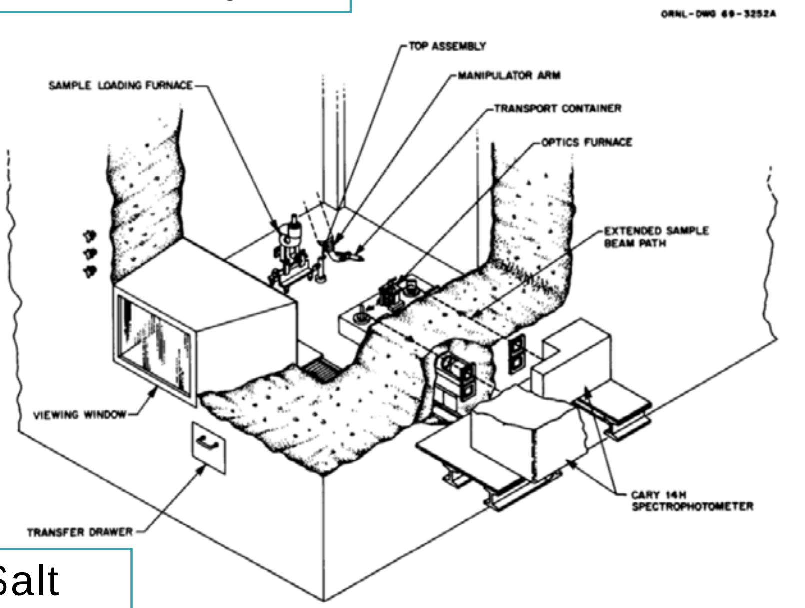
Electrochemical Measurement of U^{3+} in Flowing Fuel Salt Surrogate



Reference Electrode



Diamond Window Cell

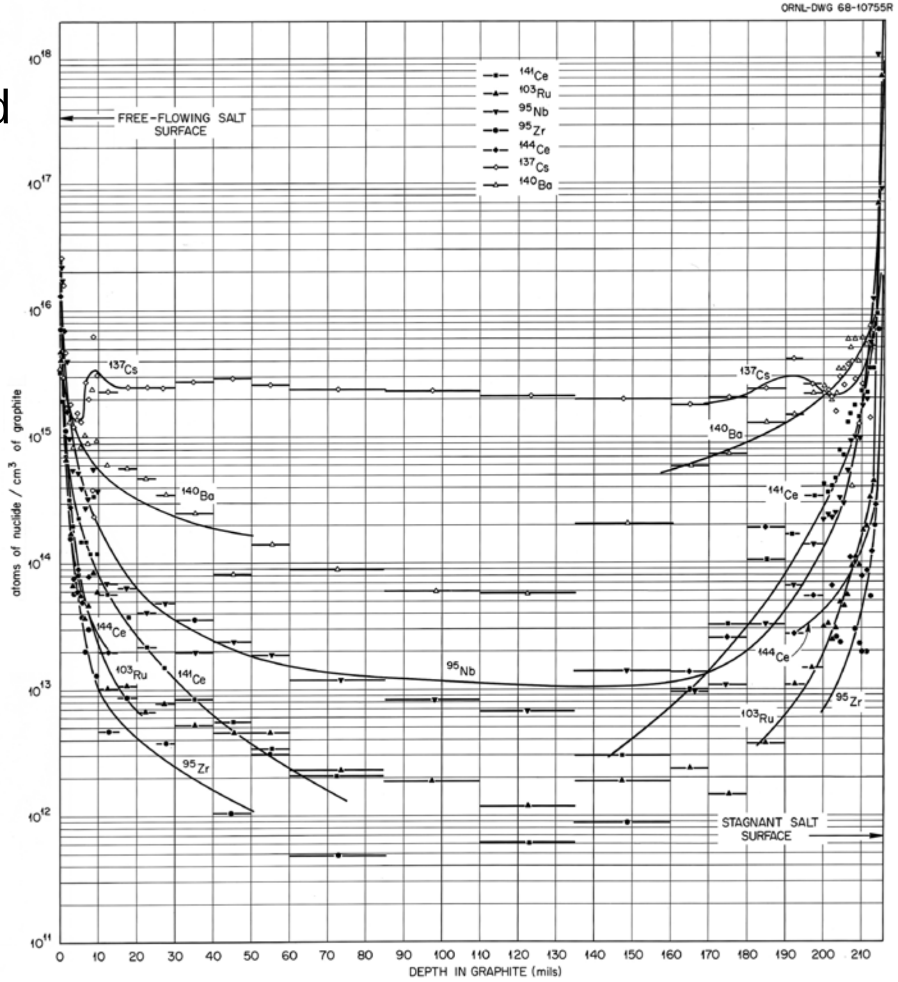


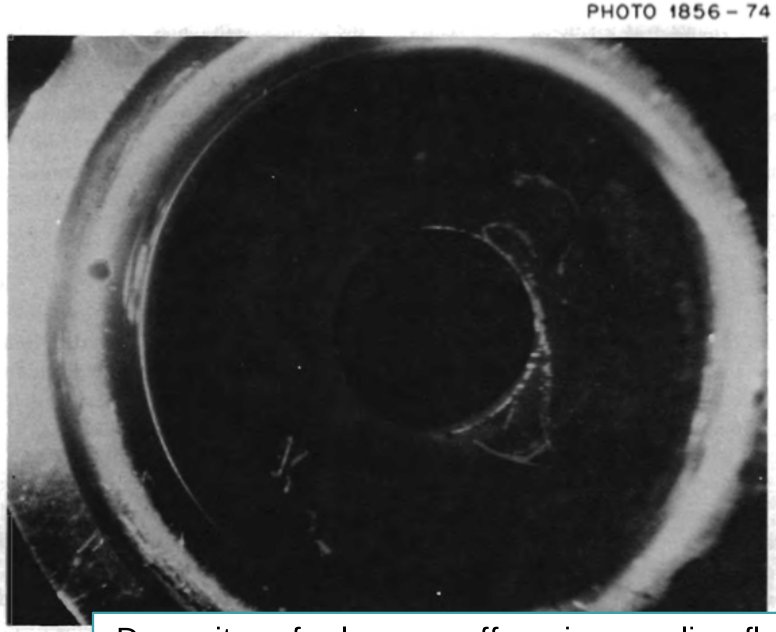
MSRE Fuel Salt Optical Evaluation

Graphite and Metal Surfaces Retained Fission Products

- Rb, Cs, Sr, Ba, Y, Zr, and lanthanides all form stable fluorides that are salt soluble – found in salt samples
  - Iodine generally remains in salt
- Nb, Mo, Tc, Ru, Ag, Sb, and Te do not form stable fluorides and are found ubiquitously on systems surfaces
  - Deposits on and in graphite were a combination of shallow (< 250 µm) salt penetration and deeper penetration by gaseous fission products

Fission product penetration into graphite stringer





Deposit on fuel pump offgas jumper line flange

Source: ORNL-4865

Alloy N is Readily Formable and Joinable into Complex Structures

- Forming techniques for austenitic stainless steels can generally be followed with a small increase in temperature
  - ORNL-TM-1854 provides an overview of Alloy N forming techniques employed at MSRE
  - ORNL-TM-5920 provides an overview of the metallurgy knowledge at the close of the MSBR program
  - ORNL and Haynes International jointly reviewed the available data for employing Hastelloy® N at MSRs
- Information necessary to generate a high-temperature ASME Section III Code Case (CC 1315-3) was generated by mid-1960s
  - Continues to be approved under Section VIII, Division 1 up to 704°C
- Melting and casting were carried out using conventional practices for nickel alloys
- Alloy N is readily weldable and brazeable using conventional techniques
- Gas-tungsten arc welding employed throughout MSRE
  - Filler metal has same basic composition
- Hot-cracking has been observed in both during welding and forming operations
  - Insufficient hot ductility to prevent cracks forming when the material contains residual stresses due to prior working

Long Term Used Fuel Salt Storage Methods and Issues

- Below ~200°C recombination becomes slower than radiolysis
  - UF_6 is volatile
- MSRE's fuel salts were left to cool for decades following shutdown
  - Periodically reheated – promoted UF_6 motion (goal was to recombine F_2 into UF_4)
  - Resulted in fissile material deposition in ventilation system filters
  - Uranium is readily removed from fuel salts through fluorination – process/facilities used for ^{233}U changeover
  - Disposal alternatives were documented shortly following shutdown
- Result was a $10s of millions cleanup effort in the 1990s–2000s
- Salt waste forms and processing technologies have been studied during and since MSBR program
  - Stability and durability are primary safety issues

Historic MSR Program Provided Substantial Experience for to Support Future MSRs

- Very positive reactor operating experience
  - Adequate solutions to materials and operational challenges were demonstrated
  - Required instrumentation and components were demonstrated
- Little information was generated for chloride salts
- Extensive experimental base provides confidence that fluoride salt interactions and operations are adequately understood
  - Remaining issues for thermal spectrum fluoride salts are in system scale-up and modernization (i.e., automation for maintenance)