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Argonne NATIONAL LABORATORY

Molten Salt Reactor PROGRAM

Salt spill testing for MSR accident progression model validation

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Motivation and Objective

Motivation

- Analysis of the effects of postulated accidents on safety is required to obtain NRC license for new nuclear reactors
- There is a lack of experimental data on processes that determine the potential consequences of molten salt reactor (MSR) accidents
  - Experimental data is needed by vendors preparing for the licensing process
  - Experimental data is needed by modelers to guide and advance model development
- Common postulated accident scenario for many MSR concept involves a rupture within the primary loop that leads to hot fuel salt spilling onto the primary containment floor

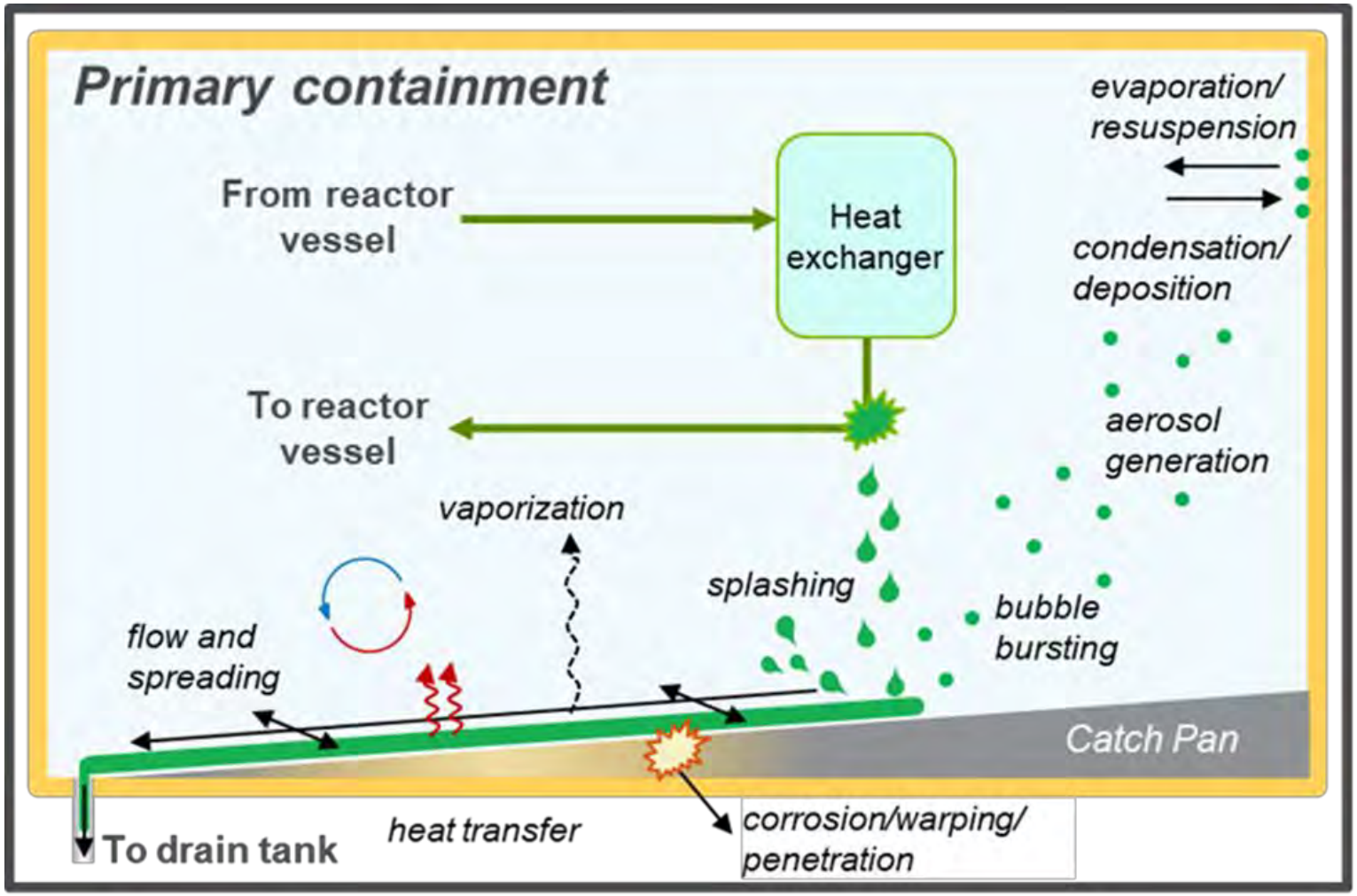
Objective

- To provide the experimental data that are needed to close identified gaps in mechanistic source term and accident progression models for postulated MSR salt spill accident scenarios.

Experimental data are needed to fill data gaps in key processes:

- Spreading and flowing behavior of the bulk salt
- Heat transfer between the salt and its surroundings
- Interactions between the salt and structural materials
- Vaporization and condensation of radionuclide species
- Formation of radionuclide-bearing aerosol and splatter particles

Molten salt spilling from primary loop onto containment floor



Pathway to generating experimental data for accident progression model development and validation

- Model simulations of postulated accident scenarios for MSRs will require experimental validation using datasets generated at a relevant scale

Salt spill scenario

Interaction with stakeholders
(model developers and MSR vendors)

Conduct individual process tests at laboratory scale	Conduct integrated process tests at laboratory scale	Design engineering-scale tests for integrated processes	Conduct-engineering-scale tests
• Identify and develop measurement techniques • Develop methods to simulate processes • Quantify dispersal behavior of spilled molten salt to fill data gaps in process models	• Apply coupled methods and measurement techniques • Measure processes simultaneously to fill data gaps on coupled processes in multi-physics codes	• Identify initial conditions of the spill and test variables • Design test apparatus • Identify safety considerations and design limitations	• Use representative fluoride and chloride salt compositions • Test geometries that facilitate model development • Adopt a modular design approach for maintenance and design flexibility

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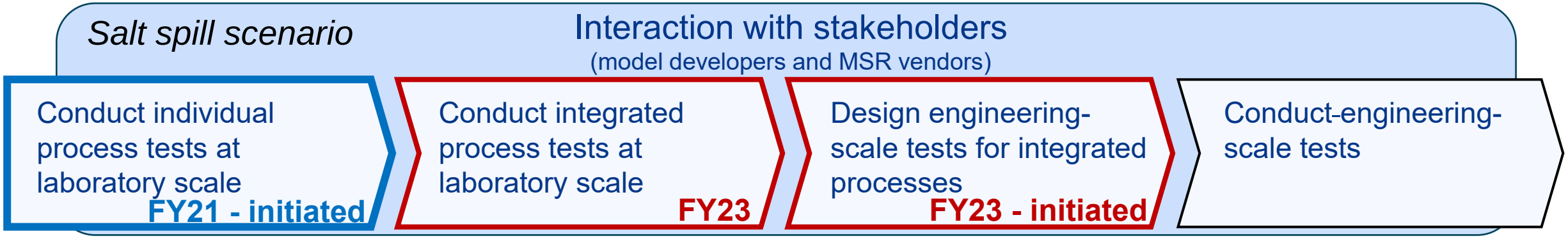

- Identify and develop measurement techniques
- Develop methods to simulate processes
- Quantify dispersal behavior of spilled molten salt to fill data gaps in process models

- Apply coupled methods and measurement techniques
- Measure processes simultaneously to fill data gaps on coupled processes in multi-physics codes

- Identify initial conditions of the spill and test variables
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- Use representative fluoride and chloride salt compositions
- Test geometries that facilitate model development
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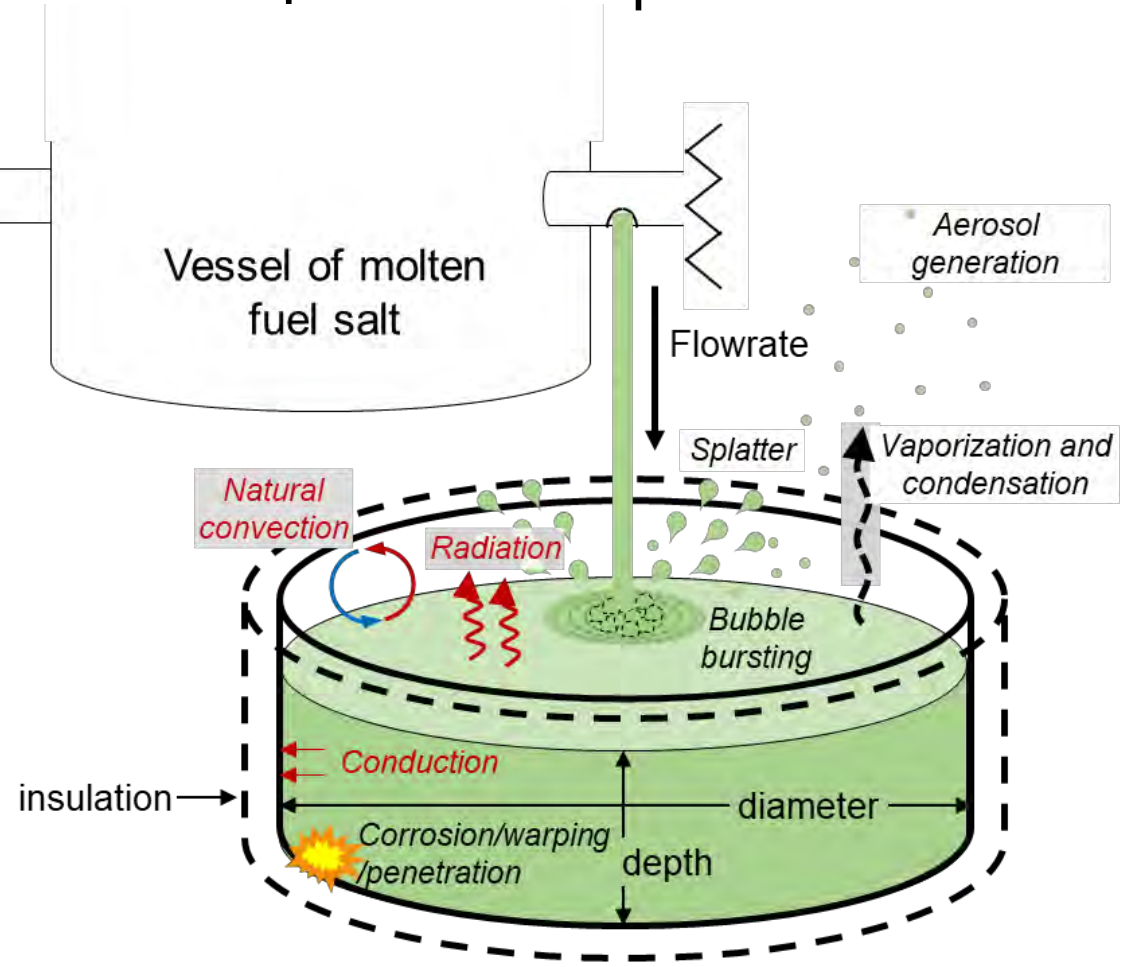
FY23 Experimental Approach

Conduct integrated process tests that:

- Employ proven methods developed during individual process testing
- Increase the amount of salt poured by up to a factor of 10 compared to previous tests
- Use salt compositions doped with multiple surrogate fission products

Schematic of FY23 spill scenario and relevant processes and parameters: Spilled salt forms static pool



FY23 Salt compositions

Salt compositions (mole %):

Component	Composition 1 (low burnup)	Composition 2 (high burnup)
FLiNaK	99.645	98.225
ZrF_4	0.05	0.25
Mo	0.05	0.25
NdF_3	0.05	0.25
CeF_3	0.05	0.25
CsF	0.05	0.25
CsI	0.005	0.025
SrF_2	0.05	0.25
Ru	0.05	0.25
Te[a]	0.005	0.025

[a] Added as Na_2Te

Pure FLiNaK

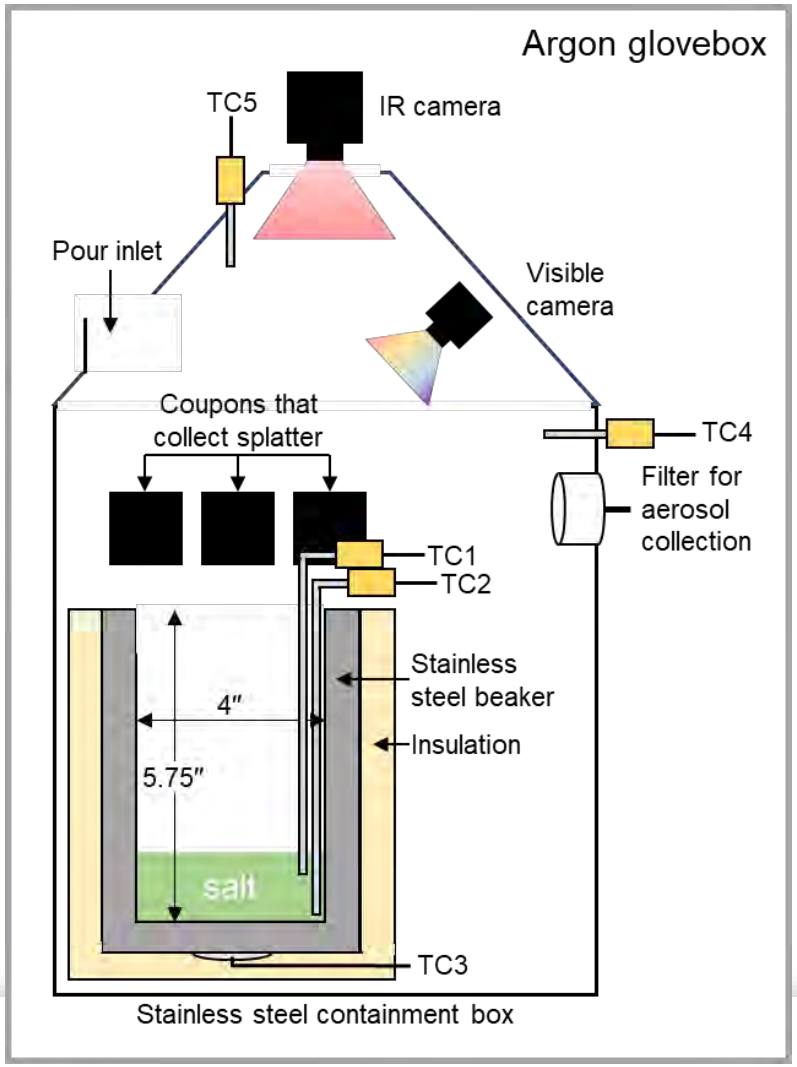
Doped FLiNaK (low burnup)

Doped FLiNaK (high burnup)

- Compositions were inspired by depletion calculation results for MSRE
- Thermophysical and thermochemical properties of doped FLiNaK compositions are being measured by Argonne team led by Melissa Rose (work package AT-23AN070501)

FY23 test setup, measurements, and variables

Schematic of test setup

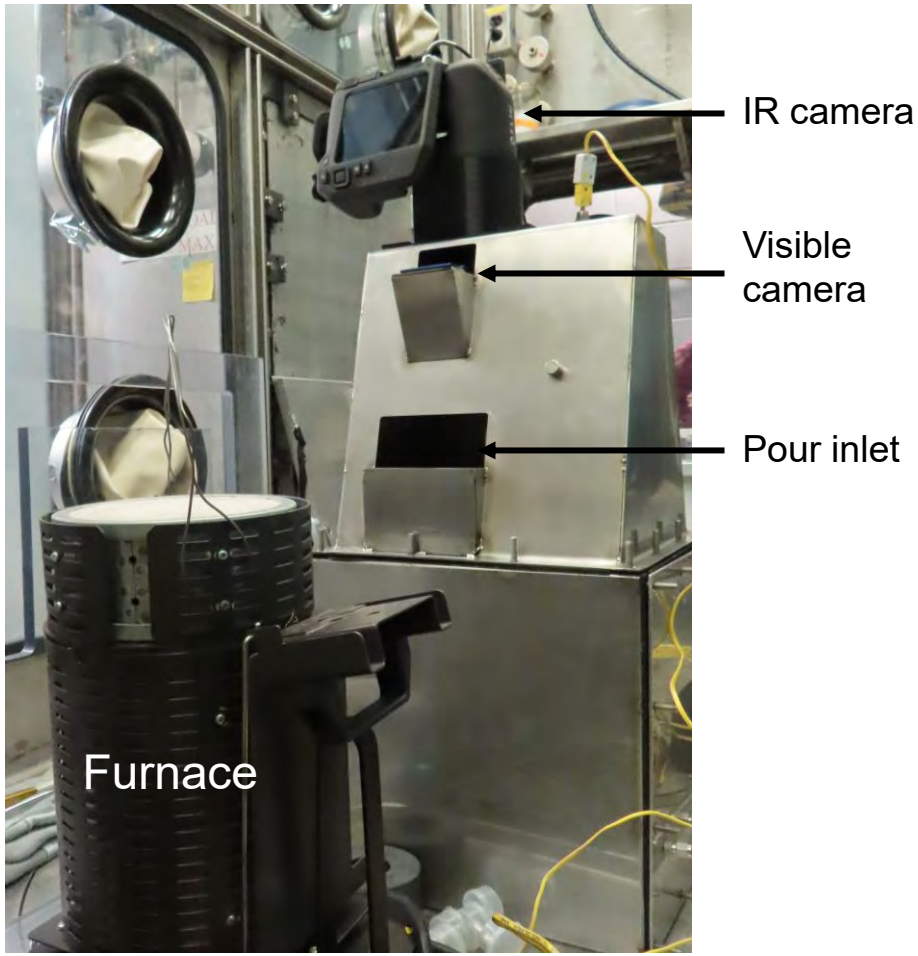


Measurements

- To quantify heat transfer from molten salt:
  - Temperature of salt and atmosphere at various locations
  - Depths and mass of frozen salt in catch pan
- To quantify dispersal of radionuclides:
  - Composition of frozen salt after the spill
  - Composition of splatter collected on adhesive
  - Composition of aerosols collected on PTFE filters
  - Composition of aerosols collected on adhesive (SEM-EDS)
  - Size of splatter and aerosol particles collected on adhesive

FY23 test setup, measurements, and variables

Setup in argon atmosphere glovebox



- Initial conditions of spill
  - Mass spilled: approximately 300 g
  - Drop height: 15″ (38 cm)
  - Pour duration approximately 5 seconds
- Variables
  - Salt composition (low and high burnup)
  - Initial salt temperature (650 °C, 750 °C, and 850 °C)

Slow motion video (0.05x) of doped FLiNaK (high burnup, 850 °C) spilling into catch pan

Quantifying heat transfer behavior of molten salt pool

- The temperature measurements of the catch pan underside, salt surface, and atmosphere at various locations within the test setup can be used to validate heat transfer models of static pools of spilled molten salt

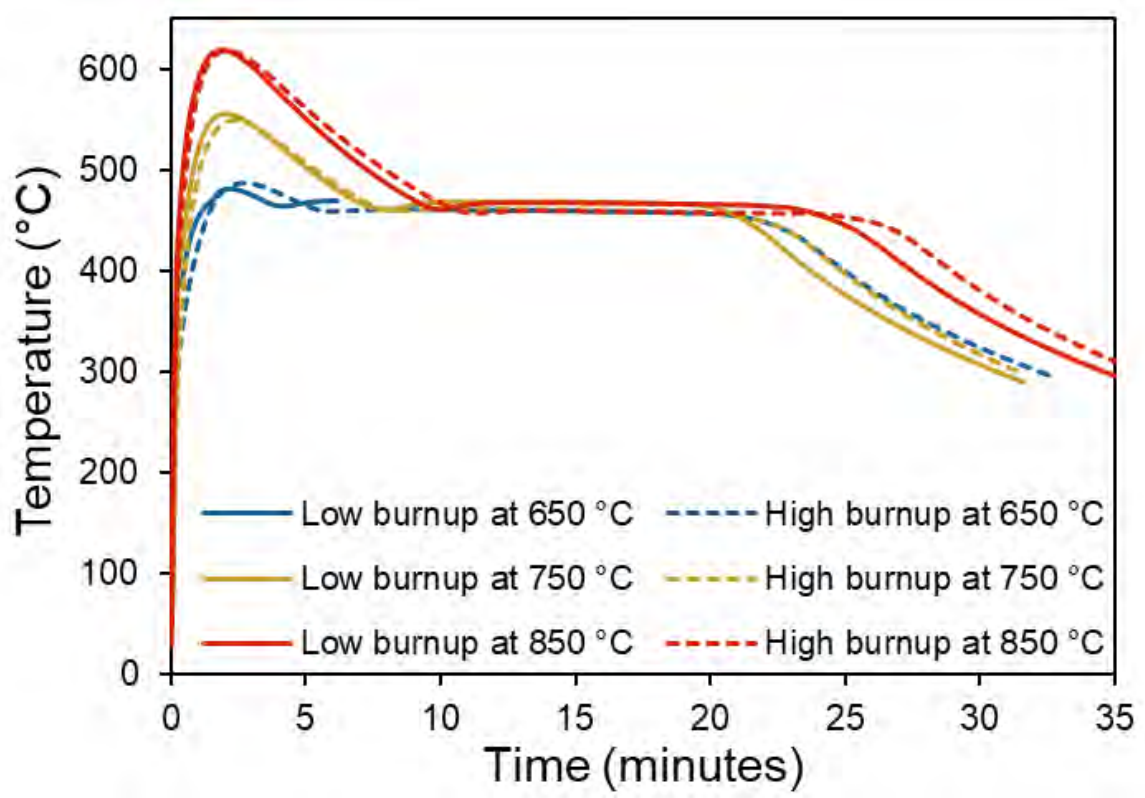
- Incorporating validated heat transfer models into accident progression simulations of salt spill accidents can reduce uncertainties around the consequences of a molten salt spill accident
  - Determines duration surface of spilled salt remains molten (radionuclide vaporization)
  - Determines duration molten salt is in contact with catch pan (catch pan integrity)
  - Informs decay heat removal strategy

Quantifying heat transfer behavior of molten salt pool

- Measured temperatures at underside of catch pan all plateau at the freezing temperature of the doped FLiNaK
- Phase transition temperature of the salt is given by the temperature plateau (between 457 °C and 468 °C, depending on initial temperature and salt composition)
- Voids observed in solidified salt after every test that was conducted

Void observed in solidified salt recovered from catch pan

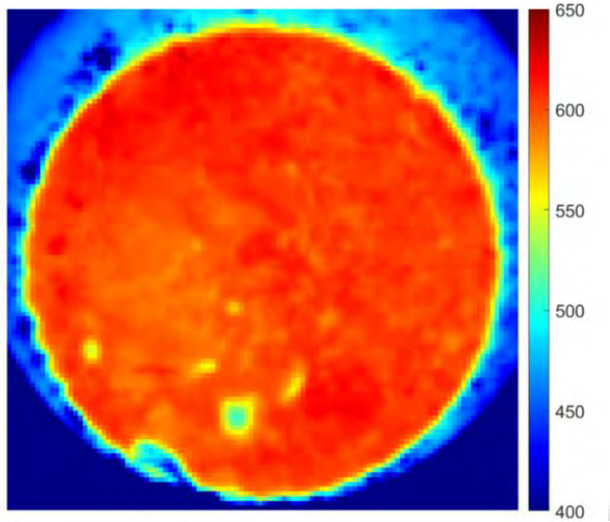
Temperature measured at underside of catch pan using surface thermocouple for six tests



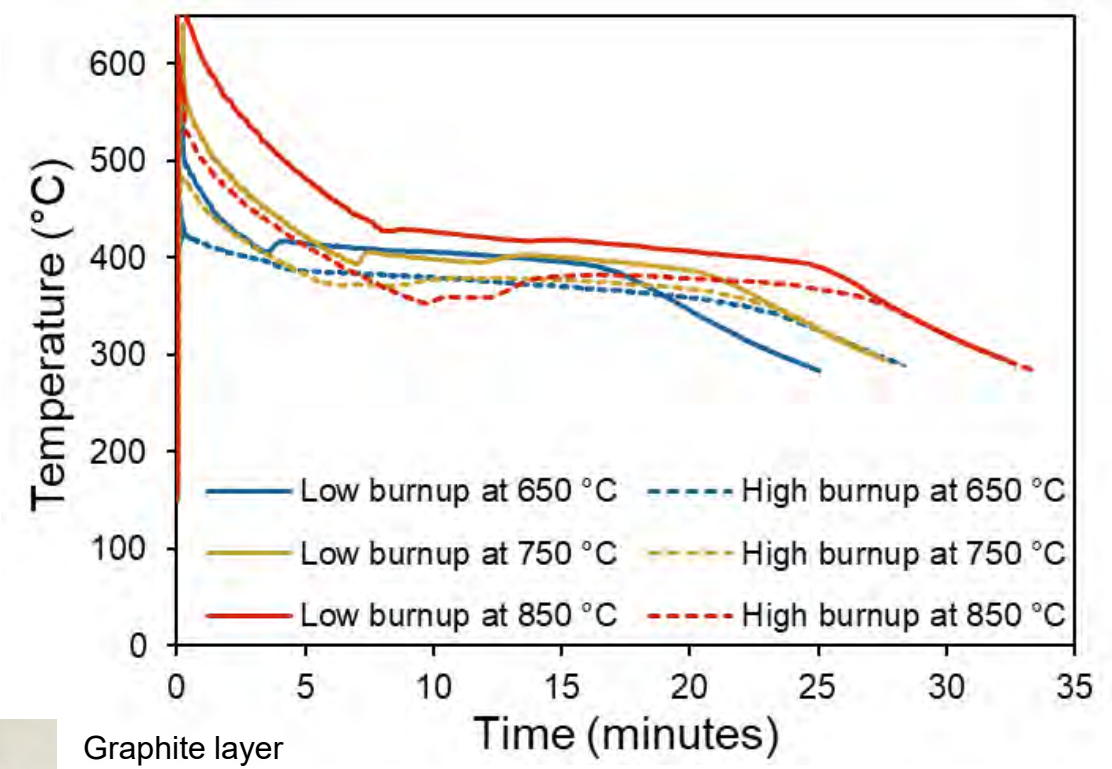
Quantifying heat transfer behavior of molten salt pool

- Determination of salt surface temperature measured by IR camera is complicated by unknown emissivity of each salt composition and potentially by graphite particles at salt surface
- Temperature is uniform across the molten salt surface

Surface temperature of doped FLiNaK (low burnup, 850 °C) in catch pan



Apparent salt surface temperature measured by using IR camera for six tests (not corrected for emissivity)



Graphite layer due to use of graphite crucibles

Quantifying surrogate fission product distribution among salt pool, splatter, and aerosol samples

- Measuring the dispersal behavior of radionuclides due to a salt spill accident determines:
  - The potential for radionuclide release beyond the containment structure (source term)
  - The potential for radionuclide-bearing splatter or condensation to deposit on structures and cause undesired heating due to radioactive decay

- Formation of radionuclide bearing aerosols during and after a salt spill is a particularly important accident consequence
  - Aerosols can remain suspended in atmosphere for prolonged periods
  - Radionuclide-bearing aerosols in the respirable size range are particularly hazardous to human health
  - Data gaps on formation mechanisms, quantity, size distribution, and composition of aerosols formed during salt spills

Quantifying surrogate fission product distribution among salt pool, splatter, and aerosol samples

- Measure elemental composition of bulk salt by using ICP-OES and ICP-MS (quantitative)
  - Elemental composition of representative sample of salt that remained in crucible
  - Elemental composition of representative sample of salt in catch pan
- Assess spatial distribution of elements in salt samples by using SEM-EDS (semi-quantitative)
  - Cross-section and edge samples taken of the pooled salt
  - Splatter particles collected on adhesive
- Determine composition of individual aerosol particles (< 20 μm diameter) collected on adhesive by using SEM-EDS (qualitative)
- Measure elemental composition of sample of global aerosol population collected on 0.45 μm filter by leaching filters and analyzing leachate by using ICP-MS (quantitative)

Summary of accomplishments and milestones

- Conducting integrated process tests using simulated irradiated fuel salt mixtures to provide experimental data on coupled processes for model development
  - Completed spill tests using two compositions of FLiNaK doped with 9 surrogate fission product reagents to simulate low burnup and high burnup fuel salt at multiple initial salt temperatures
  - Measured temperatures of catch pan underside, salt surface, and atmosphere over time for use in heat transfer models of spilled pool of molten salt
  - Collected samples of pooled salt, splatter, and aerosols for composition analyses to provide insight into radionuclide dispersal (analyses are in progress)
- Drafting report on pathway towards engineering-scale molten salt spill tests
  - Distribute report to stakeholders including accident progression and mechanistic source term modelers, MSR vendors, and participants of the MSR campaign for feedback and suggestions

Upcoming milestones – on schedule

Milestone Number	Title	Due
M4RD-23AN0602092	Roadmap report on pathway to engineering-scale tests	5/26/23
M3RD-23AN0602091	Report and data package of results from spill tests	9/15/23

Future work

- Continue to develop and confirm test methods and measurement techniques that provide validation datasets for individual process models and incorporate into engineering-scale tests, including those that emphasize:
  - Real-time monitoring of aerosol size distribution, concentration, and composition (aerodynamic particle sizer)
  - Salt compositions that contain actinides and surrogate fission products
  - Presence of humidity and oxygen in atmosphere
  - Automated spill device that allows for control of flow rate and flow velocity
- Design engineering scale salt spill tests with input from MSR campaign participants, modelers, and MSR developers

Thank you

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