Molten Salt Reactor PROGRAM

Argonne NATIONAL LABORATORY

EXPERIMENTS TO ADVANCE AND VALIDATE ACCIDENT PROGRESSION MODELS FOR MSRs

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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

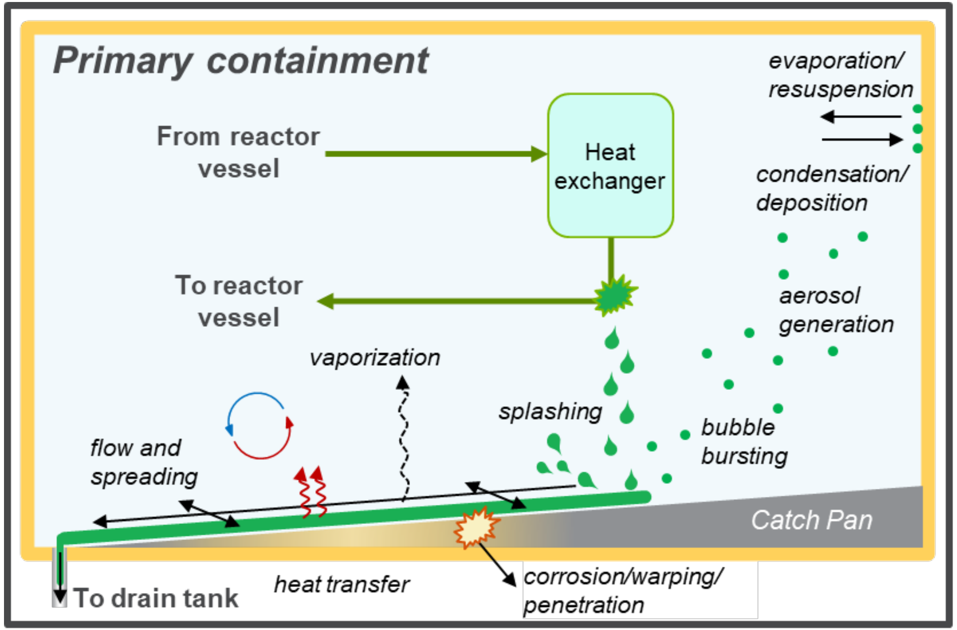
Objective

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

MOLTEN SALT SPILL ACCIDENT

- Common postulated accident scenario for many MSR concepts involves a rupture within the primary loop that leads to hot fuel salt spilling onto the primary containment floor

For MSR: salt spilling onto primary containment floor



Processes for which experimental data are needed to develop and validate models

- Spreading and flowing
  - On containment floor and through tubing into drain tank
- Heat transfer
  - By convection, conduction, and radiation
- Interactions with structural materials
  - Warping and corrosion
- Vaporization and condensation
- Aerosol and splatter formation
  - Due to splashing, spraying, bubble bursting, and vapor nucleation

EXPERIMENTAL APPROACH

Modular laboratory tests were designed and conducted to:

- Develop methods to quantify individual processes important to accident progression modeling
- Generate data that can be used in individual process models
- Identify key factors and priorities for model development and measurements in future integrated tests

Test descriptions:

1. Heat transfer of molten salt (as a static pool and on a stainless steel catch pan)
2. Spreading and freezing of molten salt on a sloped stainless steel catch pan
3. Splashing, splatter formation, and aerosol generation from spilled molten salt

Salt compositions used in tests:

- Eutectic NaCl-UCl_3 (66-34 mol %)
- Eutectic NaCl-UCl_3 doped with cesium and iodine as surrogate fission products (0.9 mol % CsCl, 0.099 mol % CsI)

SIGNIFICANCE OF MOLTEN SALT SPREADING AND HEAT TRANSFER TO CONSEQUENCES OF MOLTEN SALT SPILL ACCIDENT

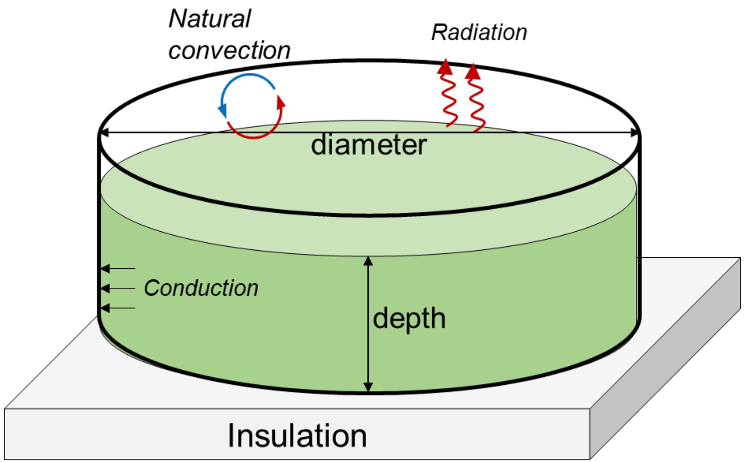
- Spreading determines the extent of radionuclide-bearing molten salt dispersal
- Spreading determines the surface area of salt in contact with the atmosphere (radionuclide vaporization)
- Spreading and heat transfer behavior determines the duration the salt surface stays molten (radionuclide vaporization)
- Spreading and heat transfer behavior determines the duration molten salt is in contact with catch pan (catch pan integrity)

Test methods were developed to:

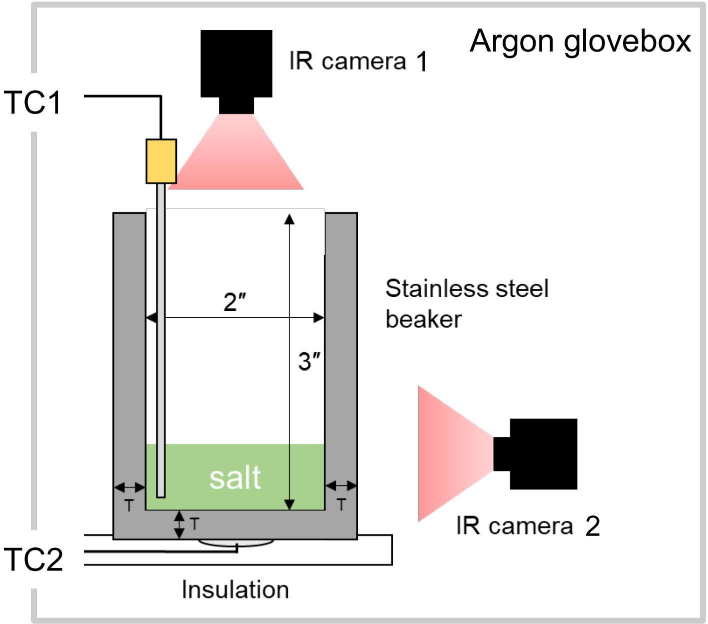
- Quantify the heat transfer behavior of static pools of molten salt
- Quantify the spreading behavior of molten salt flowing on a sloped stainless steel catch pan

HEAT TRANSFER FROM STATIC POOL OF MOLTEN SALT

Schematic of heat transfer model



Schematic of test setup



Measurements

- Temperature of salt surface (IR camera 1)
- Temperature of salt (immersed thermocouple TC1)
- Temperature of substrate underside (thermocouple TC2)
- Temperature of substrate wall (IR camera 2)
- Depth and mass of salt in beaker

Variables tested

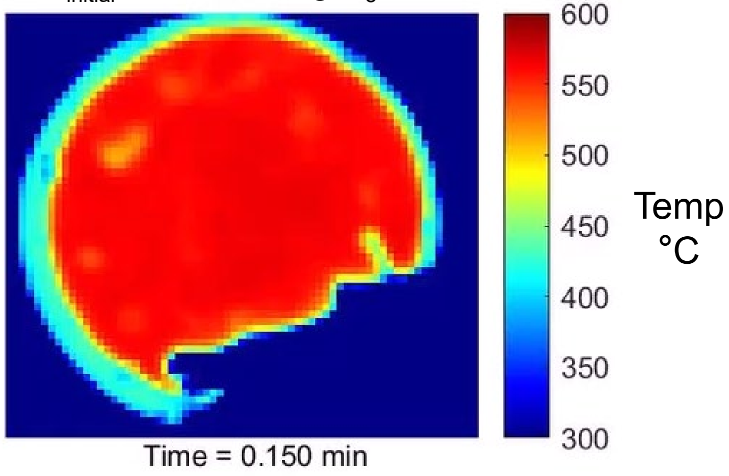
- Beaker wall & bottom thickness (¼″, ⅛″, 1⁄16″)
- Beaker thermal mass (proportional to wall thickness)

QUANTIFYING HEAT TRANSFER FROM FLiNaK AND EUTECTIC NaCl-UCl_3 FOR MODEL VALIDATION

- Temperature of the salt surface was measured by using an IR camera
- Salt near walls cools faster than salt near center
- FLiNaK retains heat more efficiently than eutectic NaCl-UCl_3 (on a per gram basis)

FLiNaK

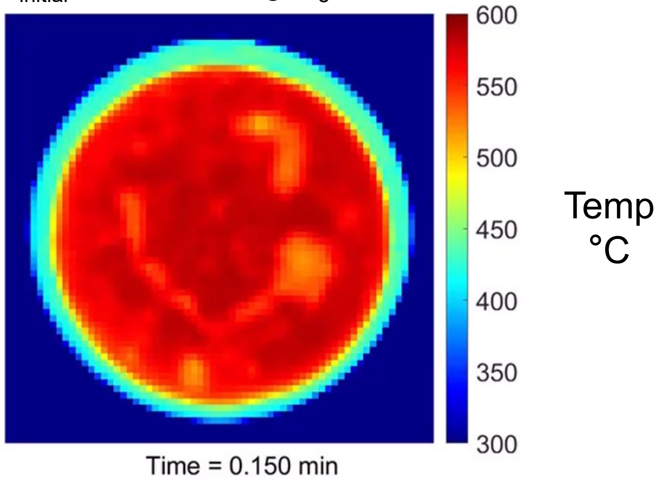
$T_{initial}$ = 650 °C, 60 g, ⅛″ wall beaker



$C_{p,liquid}$ = 1.72 J K^{-1} g^{-1}, ΔH_{fus} = 434 J g^{-1}

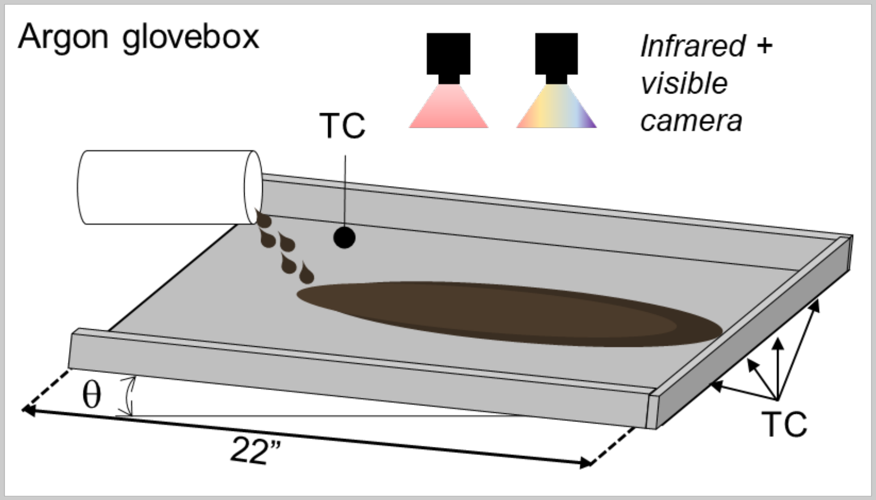
NaCl-UCl_3

$T_{initial}$ = 650 °C, 70 g, ⅛″ wall beaker



$C_{p, liquid}$ = 0.63 J K^{-1} g^{-1}, ΔH_{fus} = 158 J g^{-1}

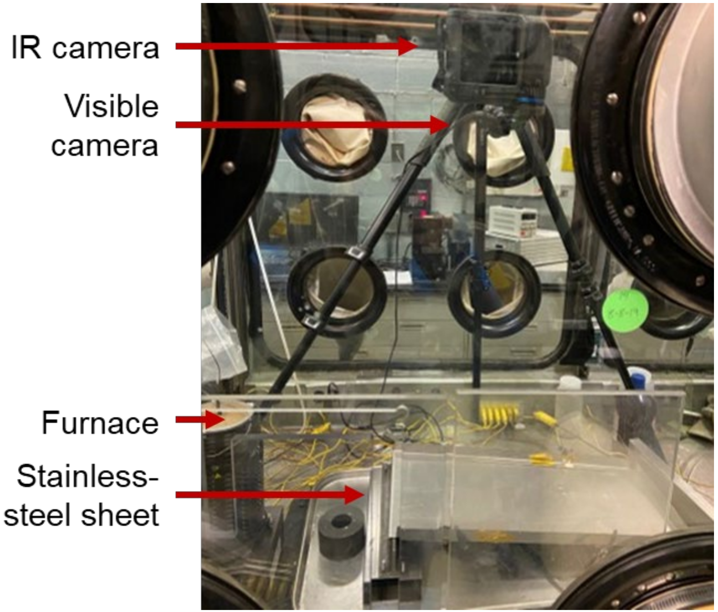
SPREADING AND HEAT TRANSFER ON SLOPED STAINLESS-STEEL CATCH PAN



Measurements

- Visible video of salt spreading
- Leading edge and covered area of salt on substrate vs. time (IR camera)
- Temperature of catch pan underside, catch pan surface, salt surface, and atmosphere
- Composition of frozen salt collected after spreading

Apparatus in glovebox



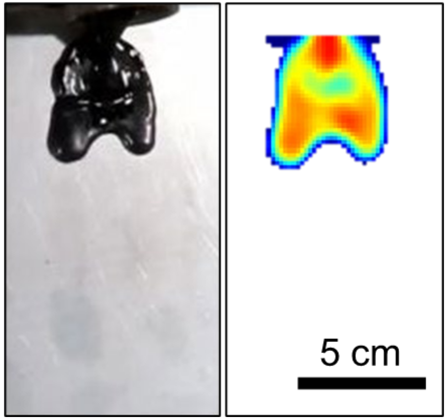
Variables tested

- Initial salt temperature (550 °C, 650 °C, 750 °C)
- Pour rate
- Pour mass

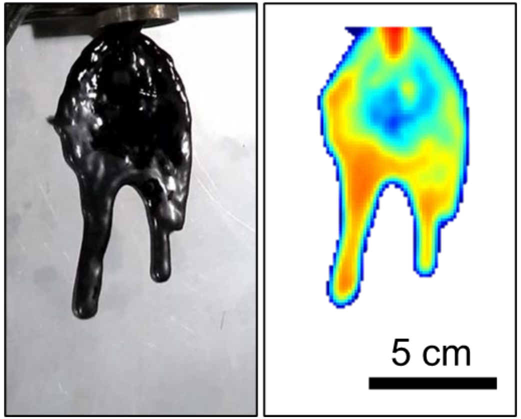
EUTECTIC $NaCl$-UCl_3 SPREADING ON STAINLESS-STEEL CATCH PAN

- Visible and IR video frames provide insight into the spreading and freezing behavior of eutectic $NaCl$-UCl_3
  - Initial conditions: mass = 65 g, temperature = 550 ° C, catch pan tilt angle = 2.5°
  - Salt appears to freeze on catch pan upon impact and forms a “salt dam”
  - Molten salt flows around the salt dam and spreads until it pools at the end of the catch pan
  - Spreading limited but not entirely hindered by freezing
  - Thin 1 mm crust formed on catch pan along flow path

Time after pour: 0.3 seconds



Time after pour: 0.7 seconds



Temp °C

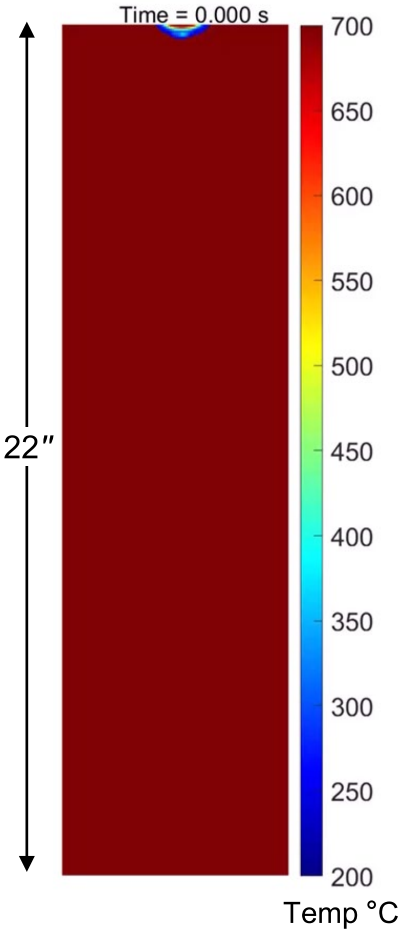
Time after pour: 14 seconds

1 mm-thick crust

QUANTIFYING MOLTEN SALT SPREADING

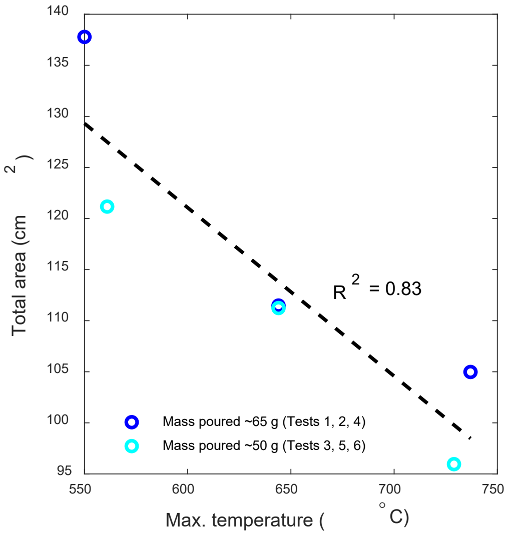
Process video frames to extract salt pixels for analysis (MATLAB)

- Convert to binary image to distinguish salt from catch pan
- Create separate image files of the salt and the catch pan
- Correct for estimated emissivity of salt



- Results from IR video analysis can be directly compared with results from spreading and heat transfer models
  - Leading edge vs. time
  - Area vs. time
  - Salt surface temperature at each pixel vs. time

Total covered area vs. initial salt temp.

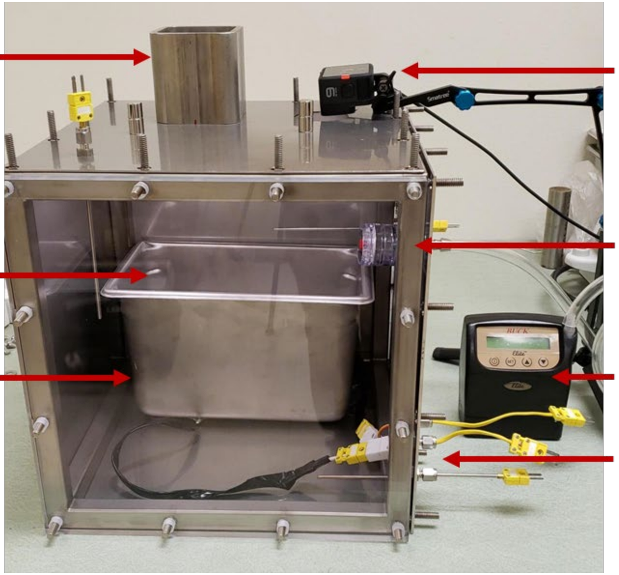


SPLATTER AND AEROSOL GENERATION FROM MOLTEN SALT SPLASHING

Significance to consequences of salt spill accident

- Determines dispersal of radionuclides (source term)
  - Radionuclide-bearing splatter
  - Radionuclide-bearing aerosols
- Determines human health hazard if radionuclide-bearing aerosols are within respirable size range

Placed in argon glovebox



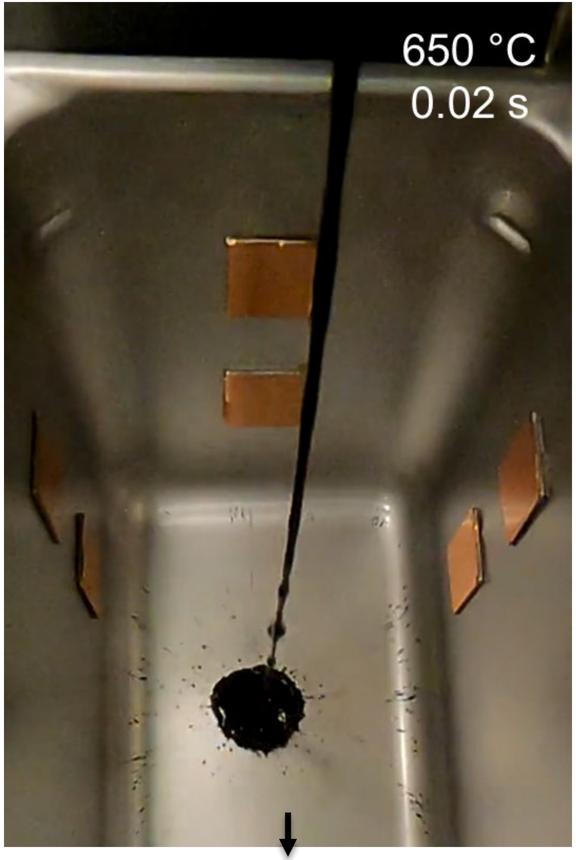
Measurements

- Visible video of molten salt splashing (slow motion)
- Splatter abundance, size distribution, and composition
- Temperature of atmosphere within spill containment box and at underside of catch pan
- Composition of aerosols collected on filters (0.45 µm pore size) by ICP-MS and collected on adhesive tape by SEM-EDS

Variables tested

- Initial salt temperature (650 °C and 800 °C)
- Presence of CsCl and CsI surrogate fission products

STILL FRAMES OF EUTECTIC NaCl-UCl_3 POURING INTO CATCH PAN



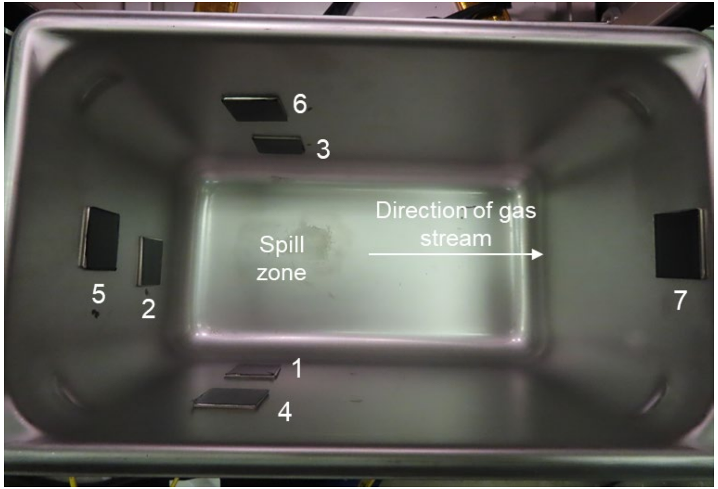
Aerosol collection filter

Aerosol collection filter

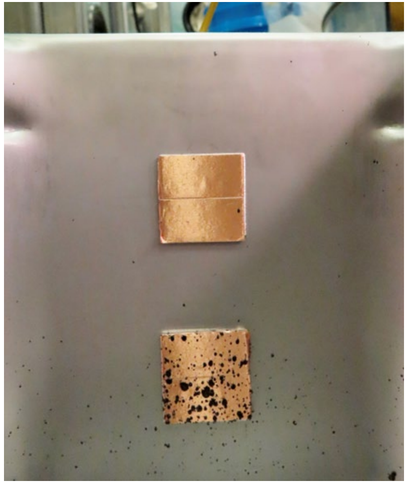
IMAGES OF SPLATTER COLLECTED ON COUPONS

- Coupons with adhesive were positioned on the catch pan walls near the spill zone at two heights above the catch pan floor to collect splatter for analysis
- One coupon with adhesive was positioned just below the filter that was sampling the spill containment box atmosphere to collect aerosol particles floating in the gas stream above the spilled salt pool

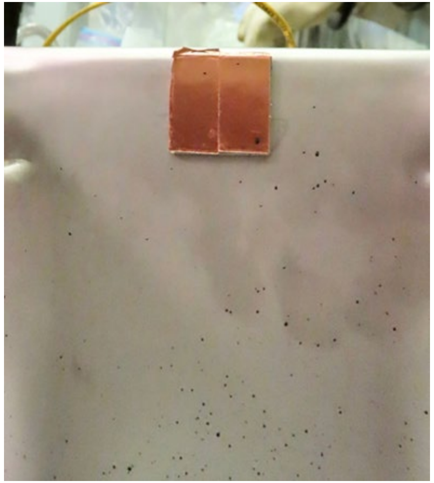
Coupon layout



Splatter on Coupons 2 and 5



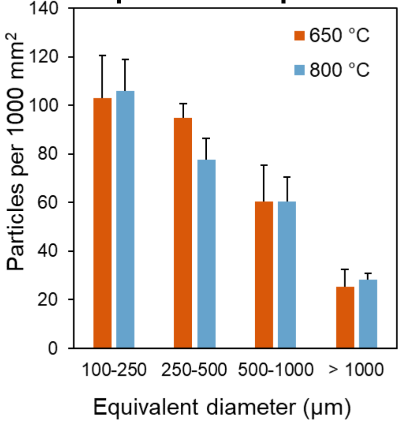
Splatter on Coupon 7



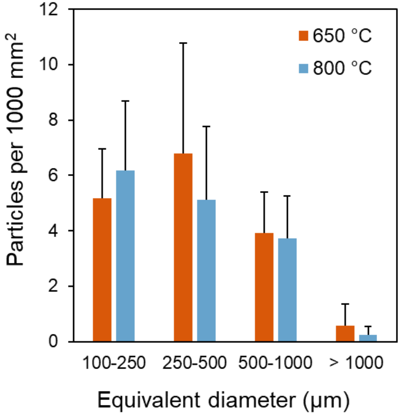
SPLATTER ABUNDANCE, SIZE DISTRIBUTION, AND COMPOSITION

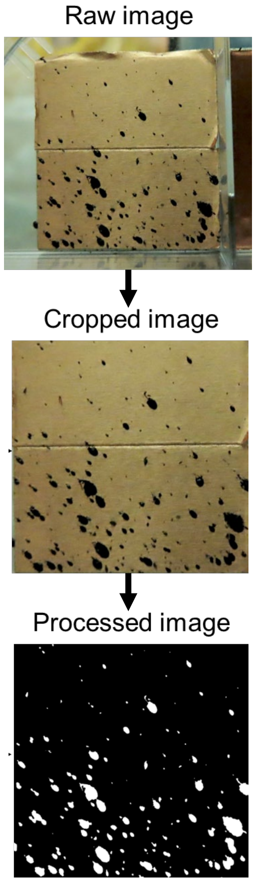
- Splatter generated due to splashing quantified by weight
  - Approximately 25% of the spilled salt mass formed splatter particles
  - Splatter abundance was not dependent on initial salt temperature
- Splatter abundance and size distribution on coupons quantified by image analysis techniques (detection limit: 100 μm)
  - Small particles (100 – 250 μm diameter) most abundant
  - Splatter size distribution was not dependent on initial salt temperature
- Splatter composition matches that of the bulk salt

Splatter on lower coupons near spill zone



Splatter on upper coupons near spill zone

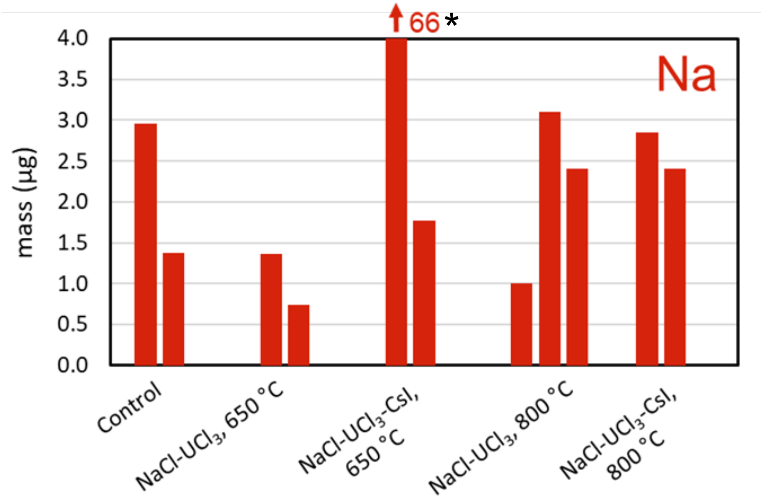


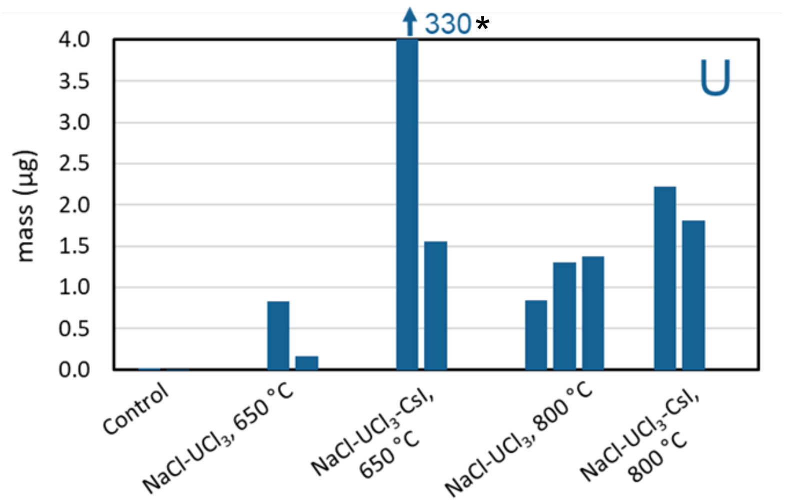


AEROSOL COMPOSITION

- Aerosol particles collected on PTFE filters (0.45 µm pore size) after each splashing test were washed off with water and the elemental composition of particles in wash solution was determined using ICP-MS
- Sodium was present on control filters at similar amounts as test filters
- Uranium-bearing aerosols detected on filters and amount appears to increase with initial salt temperature
- Iodine was not detected for any sample
- Small amounts of cesium were detected (0.01 – 0.04 µg) in some samples without a clear trend

Salt composition: eutectic NaCl-UCl_3 + 0.9 mol % CsCl + 0.099 mol % CsI



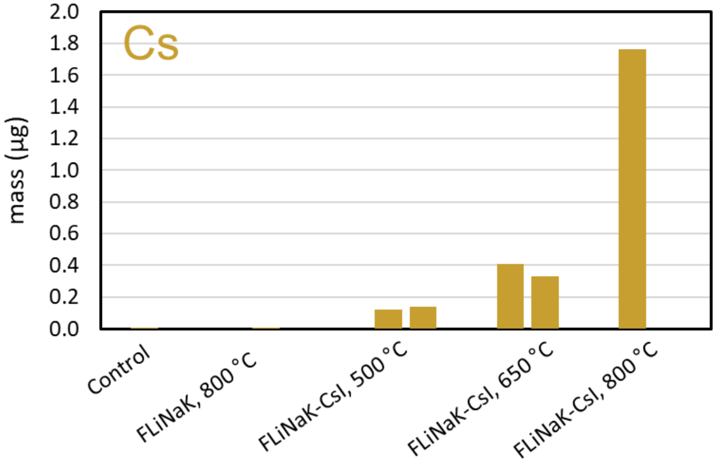


*Sample was contaminated by visible NaCl-UCl_3 splatter particle

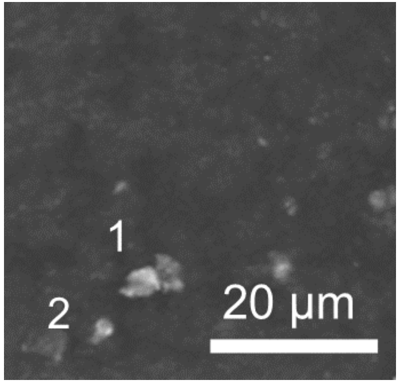
FLiNaK-Cs-I RESULTS FROM FY21

- Aerosol particles collected on PTFE filters (0.45 μm pore size) after each splashing test were washed off with water and the elemental composition of particles in wash solution was determined using ICP-MS
- Cesium-bearing aerosols detected on filters and amount increases with initial salt temperature
- Iodine (0.15 μg) is only detected on filter for test at an initial temperature of 800 °C
- CsI aerosol particles collected on adhesive were detected using SEM-EDS

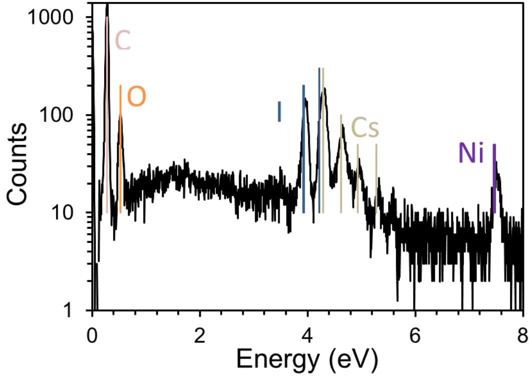
Salt composition: FLiNaK + 0.9 mol % CsF + 0.099 mol % CsI



Secondary electron image of particles on adhesive near filter



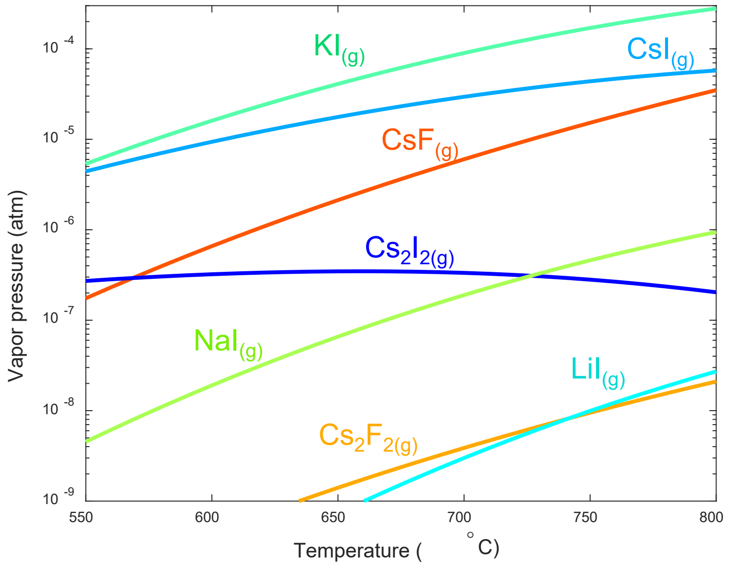
EDS spectrum of particle 1



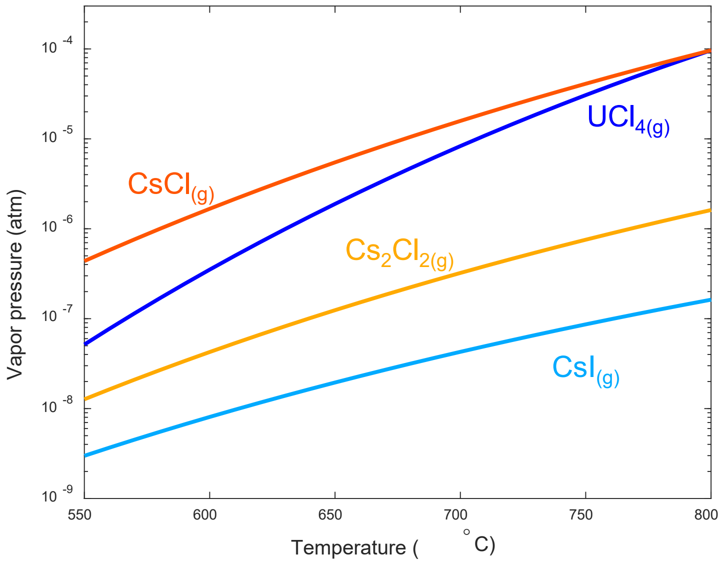
CALCULATED VAPOR PRESSURES OF RADIONUCLIDE SPECIES

- Calculated vapor pressures of key radionuclide species above eutectic NaCl-UCl_3 and FLiNaK using MSTDB-TC Version 2 and FactSage software
- Vapor pressures of iodine species much higher above FLiNaK than eutectic NaCl-UCl_3
- $UCl_{4(g)}$ is a significant vapor species above eutectic NaCl-UCl_3

FLiNaK + 0.9 mol % CsCl + 0.099 mol % CsI



Eutectic NaCl-UCl_3 + 0.9 mol % CsCl + 0.099 mol % CsI



CONCLUSIONS AND FUTURE WORK

- Methods developed for individual tests generate data that can be used to develop individual process models and these methods can be combined for integrated process testing
  - Heat transfer of static pools of molten salt
  - Spreading of molten salt on sloped catch pan
  - Splatter and aerosol generation
- Much more data are needed to understand the potential consequences of MSR accidents to supporting licensing efforts

Future work (Argonne FY23)

- Conduct integrated process tests at a larger scale using FLiNaK doped with surrogate fission products

Future work (outyears)

- Tests with complex salt compositions and environmental conditions
- Real-time monitoring of generated aerosols
- Engineering-scale tests

ACKNOWLEDGEMENTS

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  - Josh Jackson, Melissa Rose, Timothy Lichtenstein, Levi Gardner, Julia Kreuger, Bill Ebert, Vineeth Gattu, Evan Wu, Sarah Stariha, Yifen Tsai, Kristin DeAngeles, Mike Kalensky, Nicholas Condon, Mitchell Farmer, Shayan Shahbazi, David Grabaskas, and Matthew Bucknor
- Participants of the MSR campaign
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Results shown in this presentation are discussed in a publicly available report:

Thomas, S., and Jackson, J. (2022). "MSR Salt Spill Accident Testing Using Eutectic NaCl-UCl_3." Argonne National Laboratory Report. ANL/CFCT-22/32.

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