INL/MIS-23-73378-Revision-0

Understand and Predict Radiation-Induced Iodine Speciation, Chemistry, and Transport in High-Temperature Molten Salts

July 2023

Changing the World's Energy Future

Gregory Peter Holmbeck

Idaho National Laboratory

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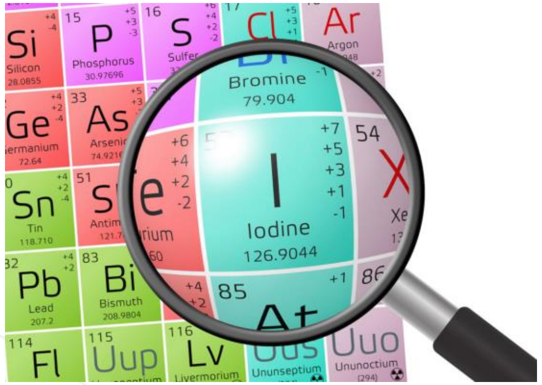
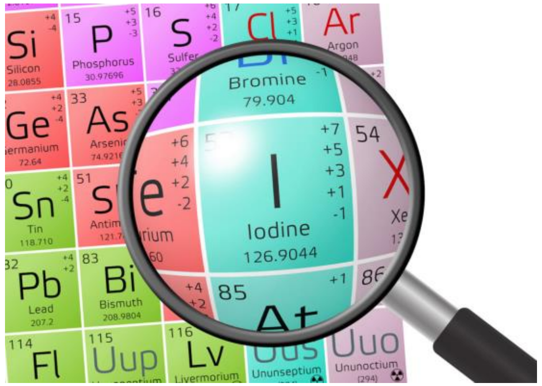


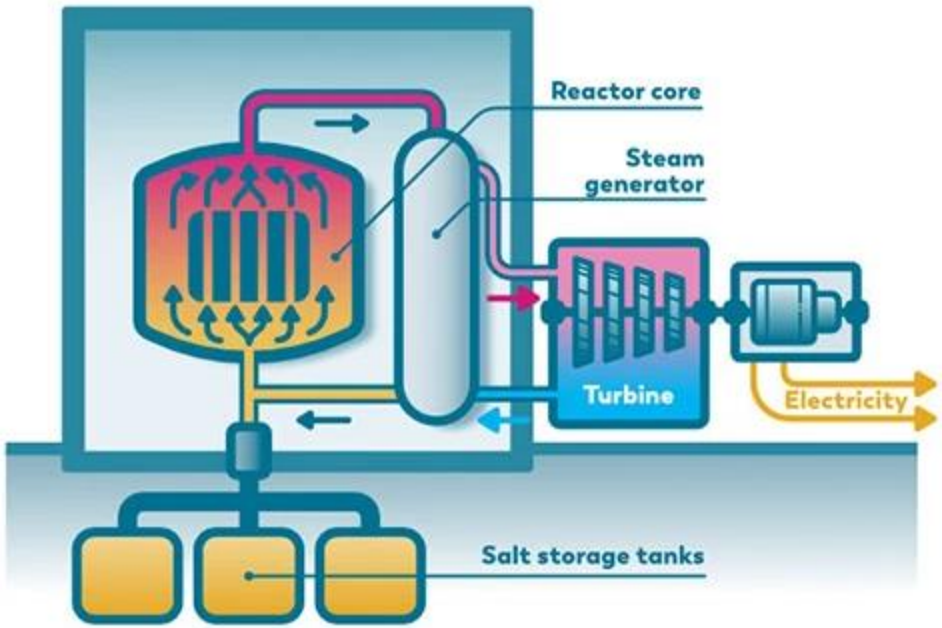
Understand and Predict Radiation-Induced Iodine Speciation, Chemistry, and Transport in High-Temperature Molten Salts

Brookhaven National Laboratory

Molten Salt Reactors and Fission Product Iodine

- Nuclear reactor technologies are an integral part of the future clean energy portfolio.
- **Iodine is a high-yield fission product** of concern for environmental release due to uptake in the human thyroid gland.
- Up to **70% of the iodine** generated in the 1975 Molten Salt Reactor Experiment **could not be accounted for**.





Retrieved from: https://www.ensuringnuclearperformance.com/

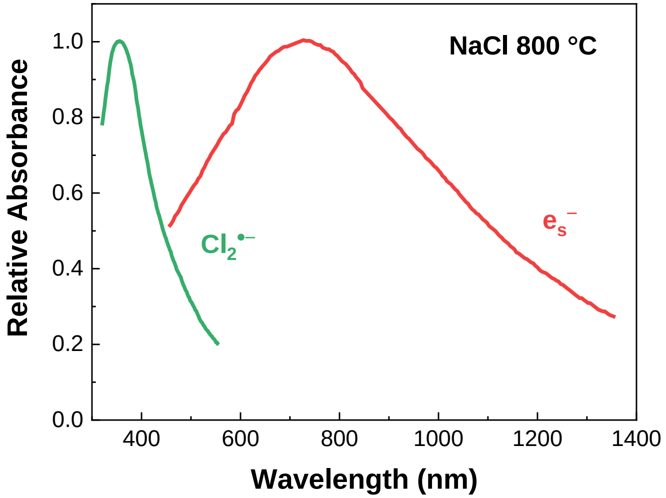
- Compere *et al.* Fission product behavior in the Molten Salt Reactor Experiment. ORNL-4865, **1975**.

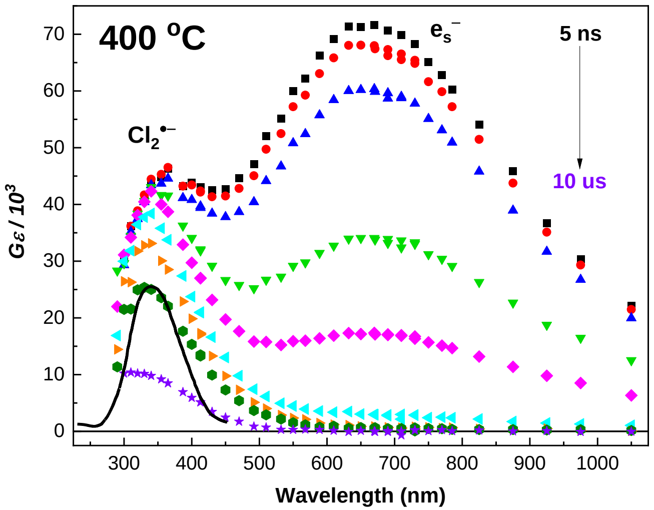
Molten Salt Radiation Chemistry

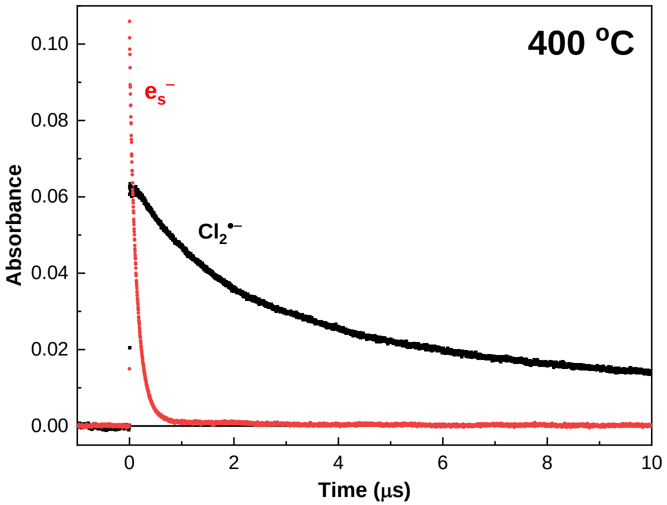
Grimes, Chemical research and development for molten-salt breeder reactors, ORNL-TM-1853, 1967.

"…no major changes in the fuel or the Inconel which could be attributed to the irradiation conditions…"

"…it was found that only when the irradiated fuel was allowed to freeze and cool below 100°C did radiolytic decomposition take place."

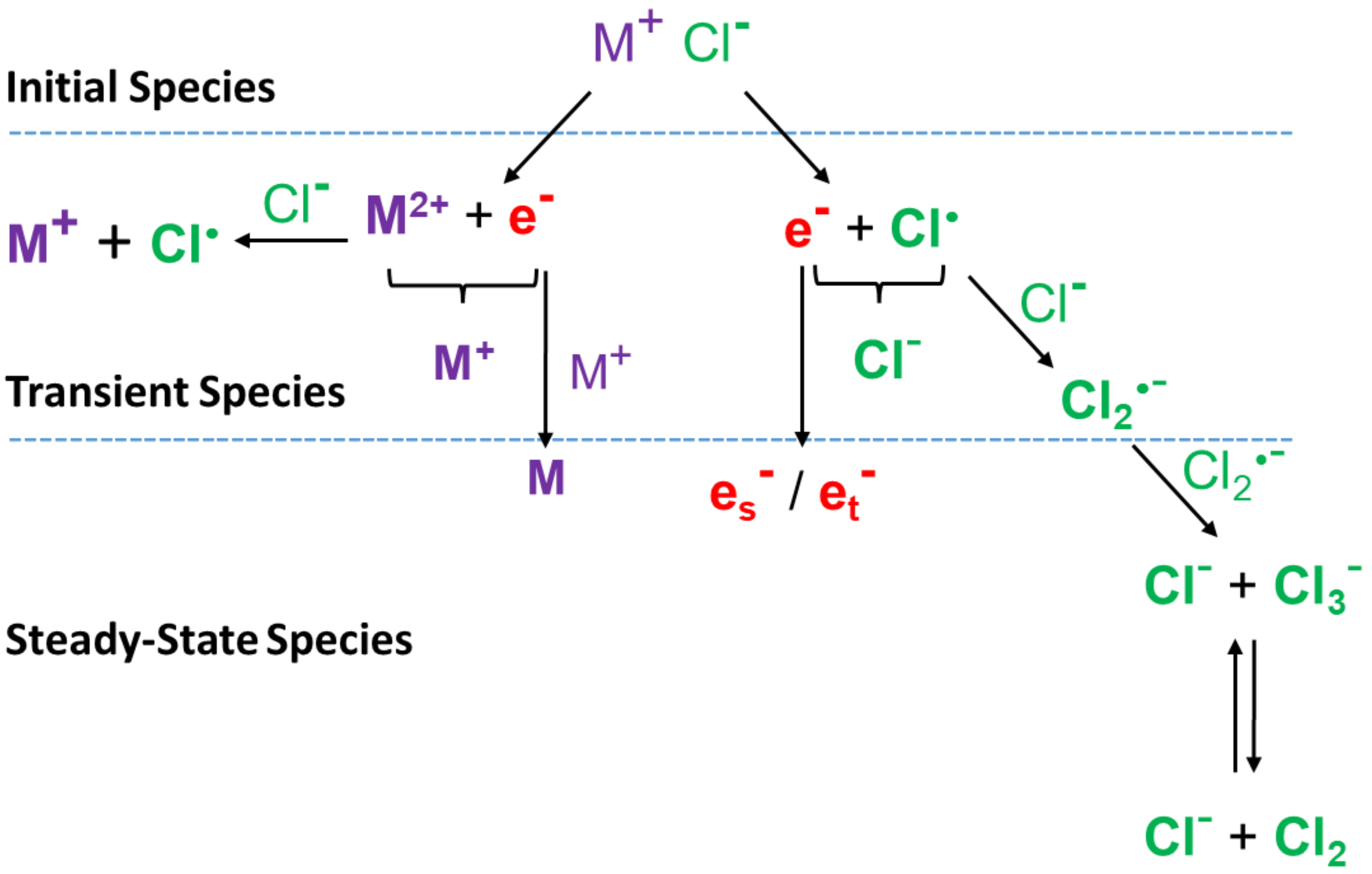






- Pikaev *et al.*, *Rad. Phys. Chem.* **1982**, *19*, 377.
- Iwamatsu et al., *Phys. Chem. Chem. Phys.* **2022**, *24*, 25088.

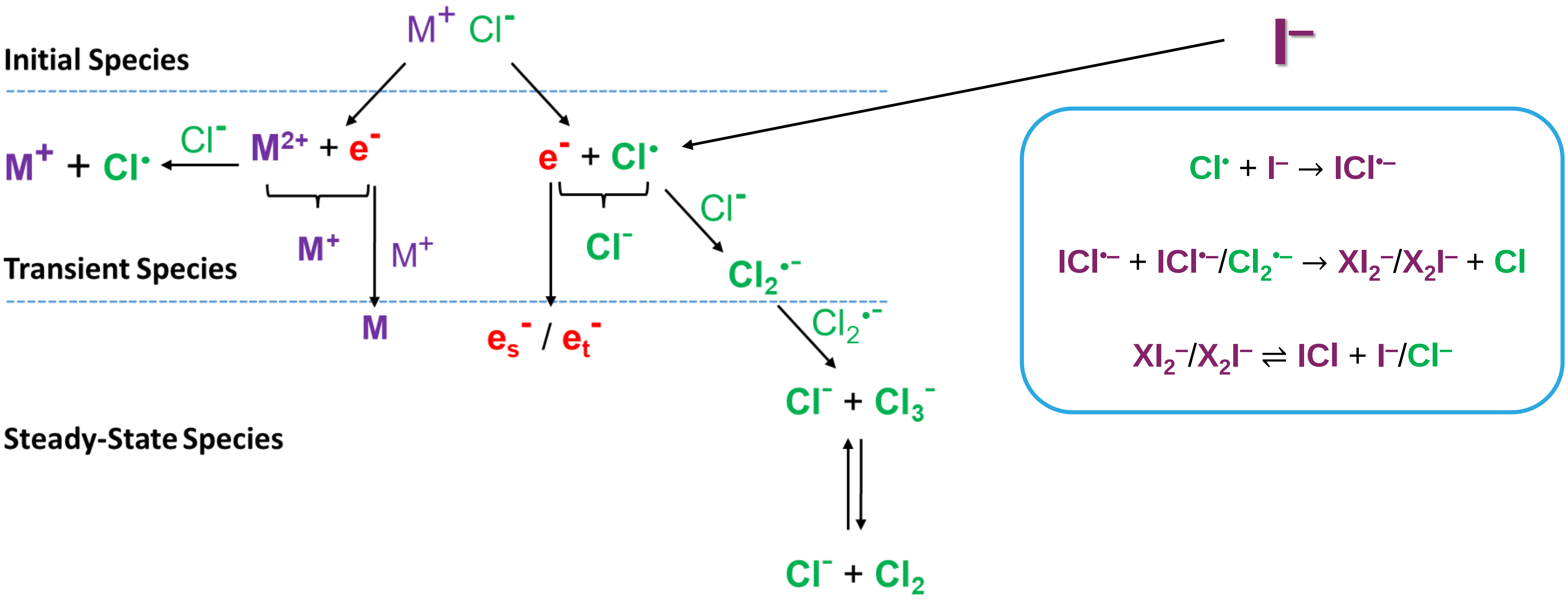
Molten Salt Radiation Chemistry



- Phillips *et al.*, *AIP Rev, Sci. Instr.* **2020**, *91 (8)*, 083105.
- Ramos-Ballesteros *et al.*, *Phys. Chem. Chem. Phys.* **2021**, 23, 10384.
- Dias *et al.*, *J. Phys. Chem. Lett.* **2021**, *12*, 157.
- Ramos-Ballesteros *et al.*, *J. Phys. Chem. C* **2022**, *126 (23)*, 9820.
- Iwamatsu et al., *Phys. Chem. Chem. Phys.* **2022**, *24*, 25088.

- Conrad *et al.*, *Phys. Chem. Chem. Phys.* **2023**, *25*, 16009.

Molten Salt Radiation Chemistry



- Conrad *et al.*, *Phys. Chem. Chem. Phys.* **2023**, *25*, 16009.

Goal

To quantitatively understand and predict radiation-induced iodine speciation, chemistry, and transport in high-temperature molten salt environments.

Question: *How does the absorption of ionizing radiation change the chemical and physical properties of iodine-bearing molten salt systems?*

Central Hypothesis: *The radiation-induced conversion of iodide will yield an extensive suite of transient and steady-state iodine radiolysis products that will alter the bulk chemical and physical properties of the irradiated molten salt system—the speciation, distribution, and chemical transport of which will be dictated by the composition and the availability of multivalent metal cations and metal alloy interfaces.*

Aims

- *Identify and characterize the suite of radiation-induced iodine species formed in high-temperature molten salt mixtures*
- *Determine their impact on the bulk chemical and physical properties of a molten salt mixture*
- *Elucidate their reaction kinetics, mechanisms, and chemical transport as a function of temperature and base salt composition*
- *Evaluate their interplay with multivalent metal ions and metal alloy interfaces*
- *Develop multiscale computer models for the interrogation and prediction of reaction mechanisms, speciation, and chemical transport in irradiated, high-temperature iodine-bearing molten salts.*

The Iodine Team











- **INL**, **BNL**, **Postdoctoral Researchers**, and **Graduate Students**

Ruchi Gakhar

INL Staff Scientist

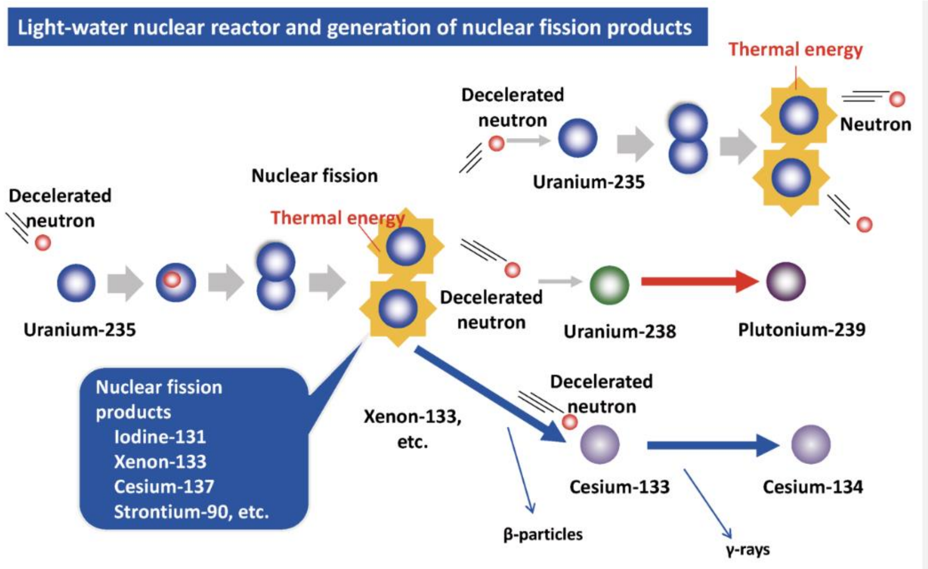
Understand and Predict Radiation-Induced Iodine Speciation, Chemistry, and Transport in High-Temperature Molten Salts

Research Objective #1

Research Objective 1

“What is the chemical speciation of iodine and iodine polyhalides in molten salts, and how do they impact the physical and chemical properties of a molten salt system?”

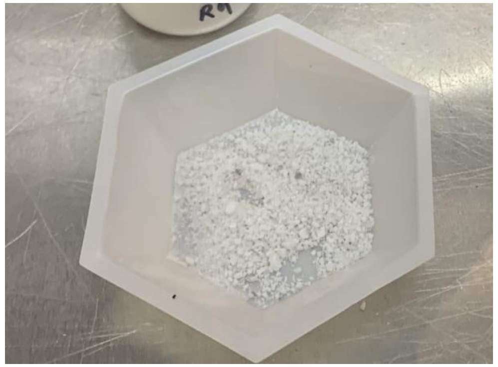
(i) the inclusion of iodine changes the spectroscopic features and physical properties of molten salt mixtures;
(ii) how this speciation changes in the presence of other solutes. e.g., multivalent metal ions; and
(iii) how iodine speciates in the molten salt and the attendant vapor phase in the absence of radiation fields

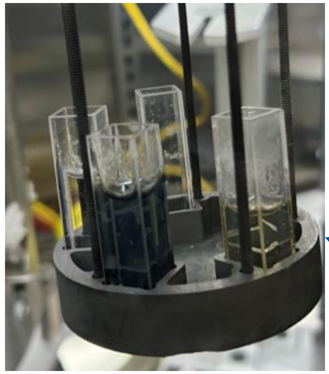


Challenges we faced handling Iodides ..

- Procurement of high-purity, anhydrous reagents
- Preparation of salt mixtures

Without pre-baking step

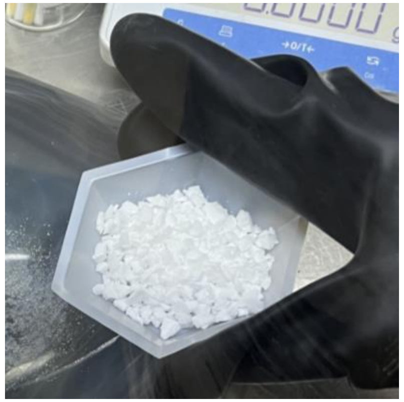
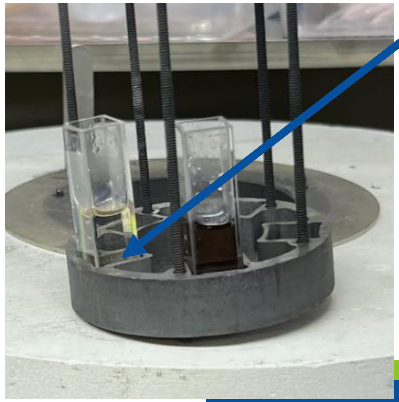




LiI-KI eutectic
in molten state

LiI-KI eutectic batches

With pre-baking step

Challenges we faced handling Iodides ..

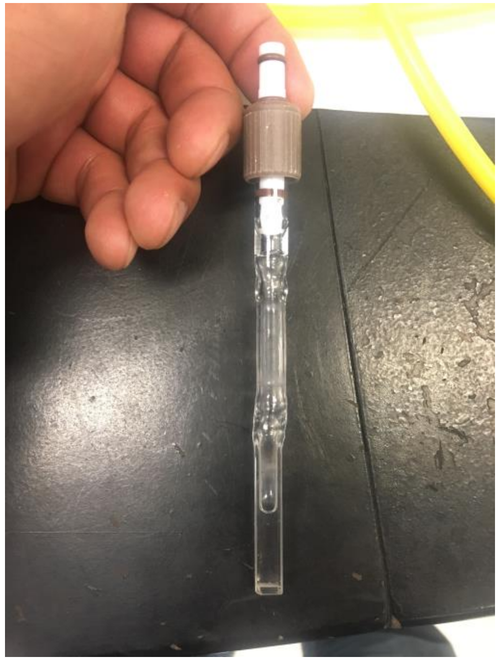
- Flame-sealing of samples

Sample heated at 200ºC

Sample heated at 300ºC

Samples flame-sealed using Swagelok Ultra-torr fittings and sample valves with straight tube

Formation of molecular I_2 apparent



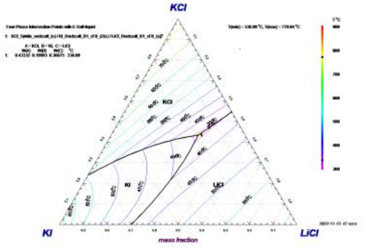
Modified approach – Use of custom Teflon threaded stopcock

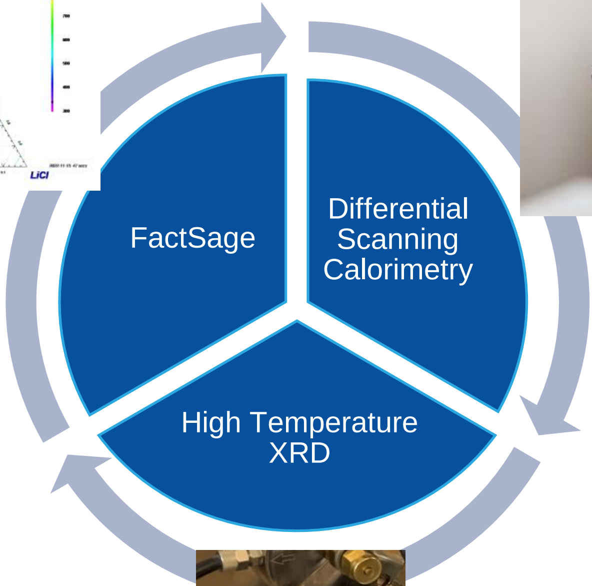
no visual indication of I_2 formation

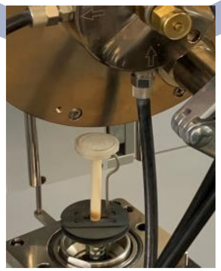
Research Focus 1: Impact of iodide species on phase evolution of chloride salt

Temperature-driven phase evolution of LiCl-KCl in presence of LiI and KI

Goal: To investigate how the thermodynamic properties and crystal structure of the base salt are affected by the addition of different concentrations of iodide.







Research Focus 2: Effect of ligand on Ni^{2+} coordination

Impact of ligand strength on metal ion coordination

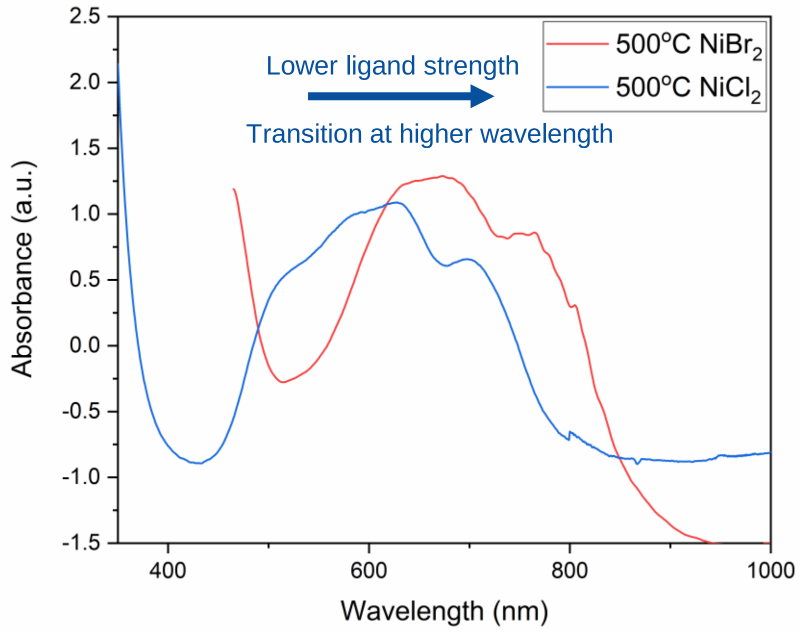
Spectrochemical Series of ligands

I^- < **Br^-** < S^{2-} < NCS^- < **Cl^-** < NO_3^- < F^- < OH^- < H_2O < SCN^- < CH_3CN < NH_3 < NO_2^- < CN^- < CO

Higher ligand strength

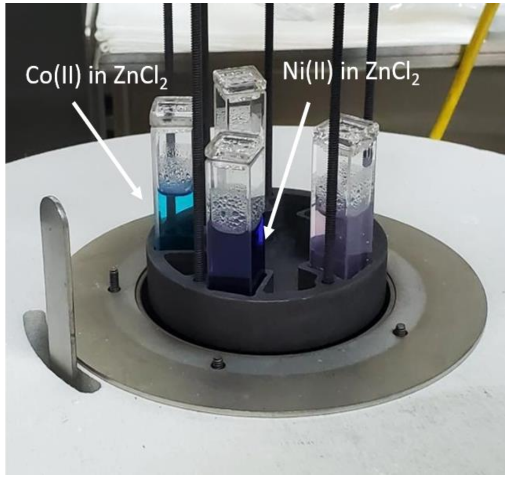
Higher Crystal field splitting energy (CFSE)

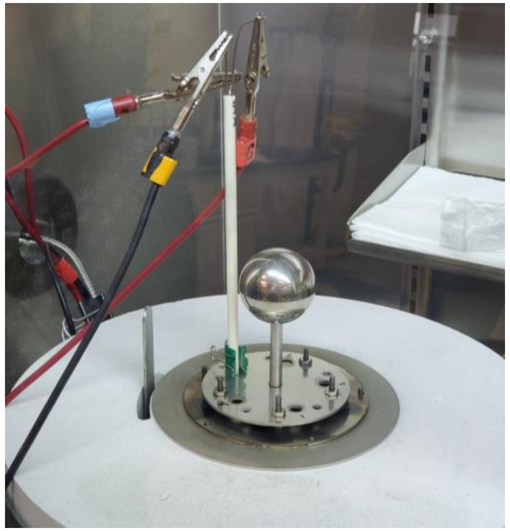
Electronic transition occurs at higher energy/lower wavelength

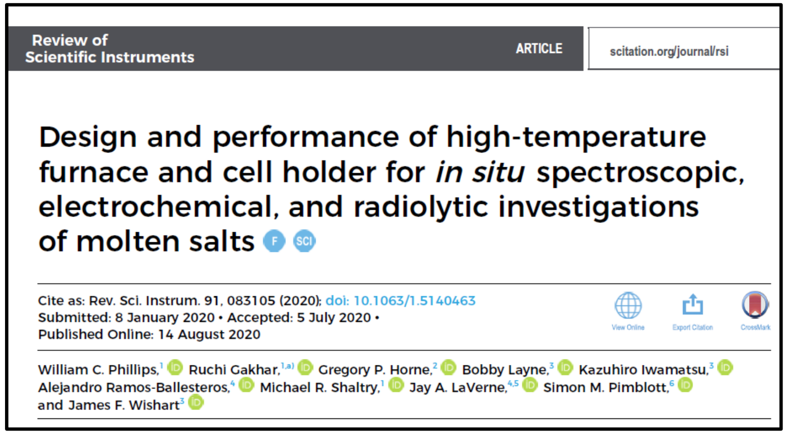


Advanced Spectroscopy Furnace at INL

- **Agilent Cary 5000 UV-Vis-NIR**
  - 175-3,300 nm spectral range
  - Up to 0.01nm resolution
- **Custom high temperature rated fiber optics**
- 200-1400nm (UV-Vis)
  - 300-2300nm (Vis-NIR)
- **Custom-designed furnace for *in-situ* electrochemistry-spectroscopy**
  - Ten optical ports and five sample slots
  - Accommodates standard quartz cuvettes 1cm pathlength







Review of Scientific Instruments | ARTICLE | scitation.org/journal/rsi

Design and performance of high-temperature furnace and cell holder for *in situ* spectroscopic, electrochemical, and radiolytic investigations of molten salts

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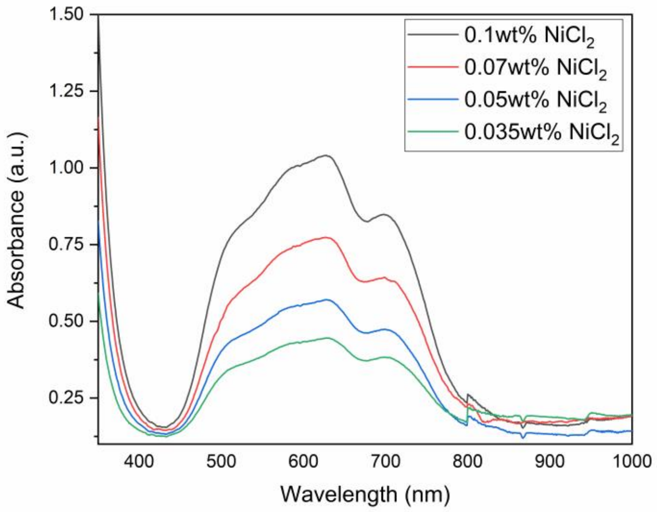
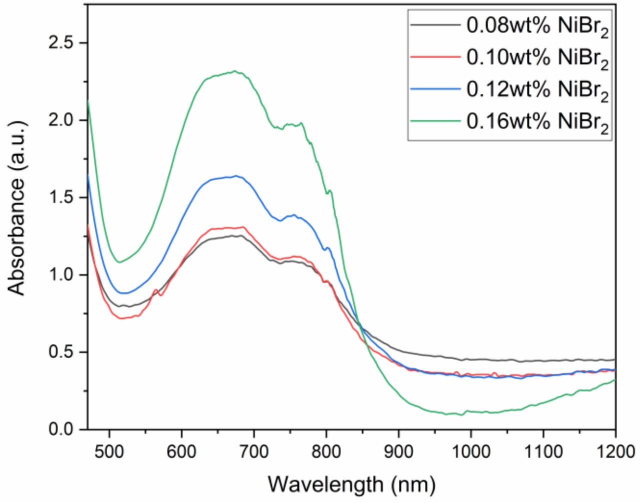
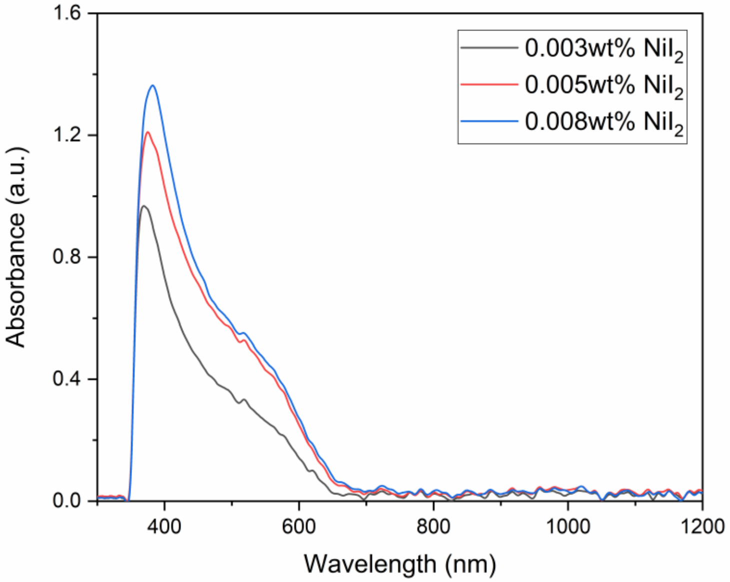
Ni^{2+} in halides: Cl, Br and I







$NiCl_2$ in LiCl-KCl

352ºC

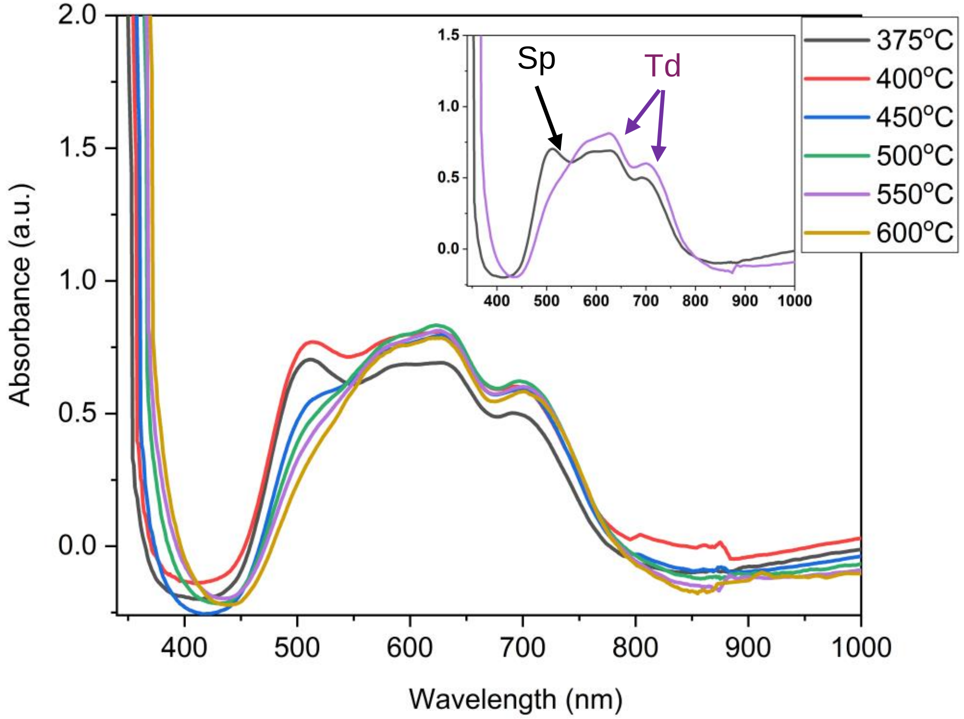
$NiBr_2$ in LiBr-KBr

320ºC

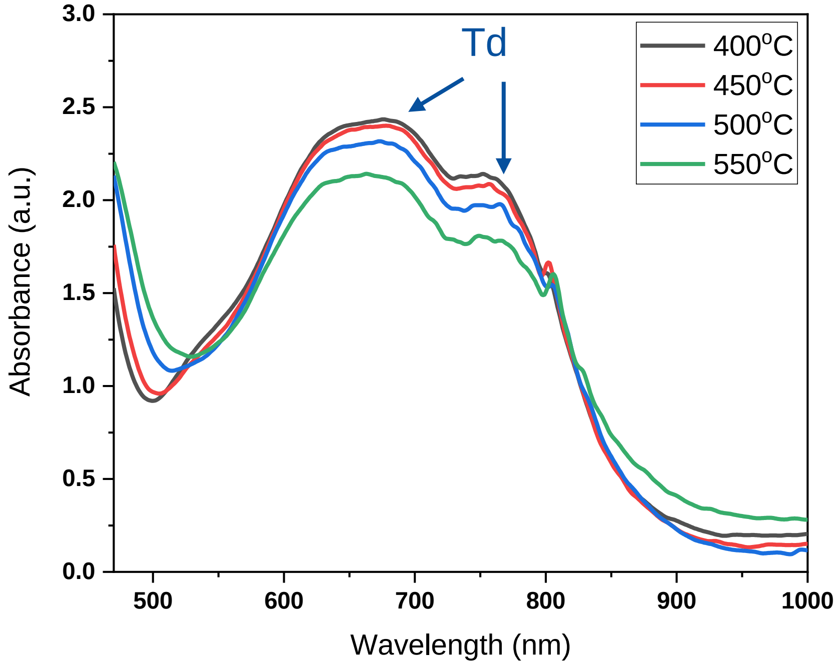
NiI_2 in LiI-KI

285ºC

Temperature-dependence on coordination environment of Ni^{2+}: LiCl-KCl and LiBr-KBr melts



Ni^{2+} in LiCl-KCl eutectic



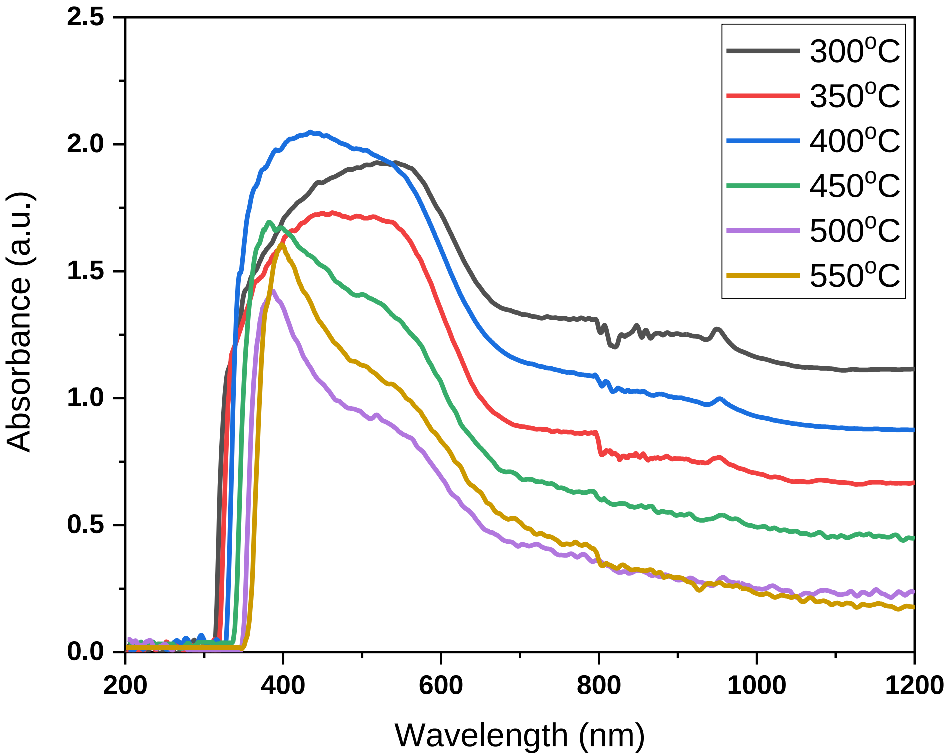
Ni^{2+} in LiBr-KBr eutectic

Structural heterogeneity observed for Ni^{2+} in chloride melt – transition from Sp to Td symmetry with temperature

Parallel XAFS study on these systems was performed by Nirmal Patra and Simerjeet Gill, BNL

In bromide melt, Ni^{2+} coordination is homogenous – symmetrical Td in 400-550°C range

Temperature-dependence on coordination environment of Ni^{2+}: LiI-KI melt



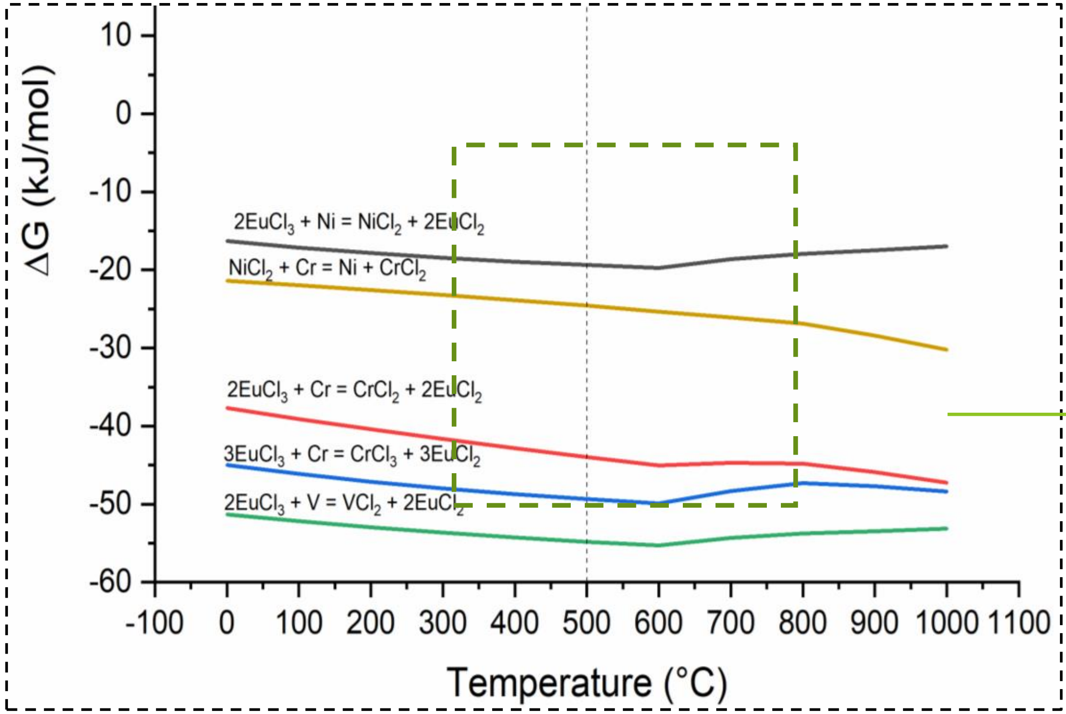
Structural heterogeneity observed for Ni^{2+} in iodide melt

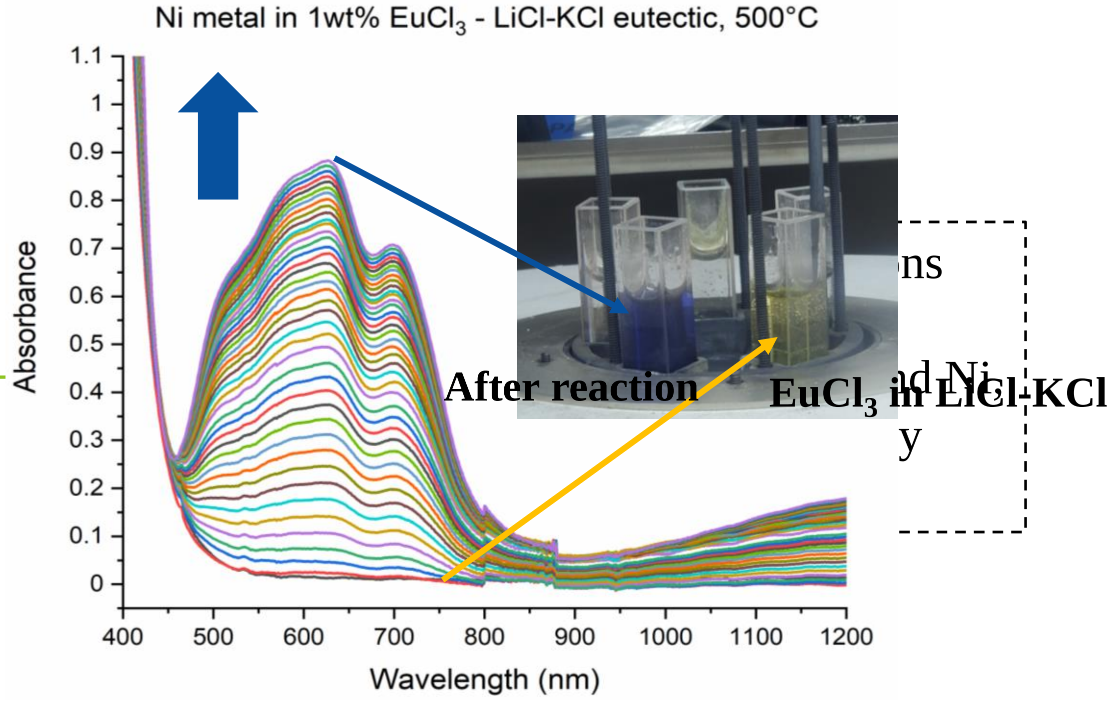
Need to corroborate results with XAFS for peak interpretation and assignment

Research Focus 3: Interaction Between Solute Species and Metallic Alloying Elements in Molten Salts

- **Molten salt chemistry has a complex impact on corrosion.** Influenced by impurities, redox potential, **fission product** buildup, etc.

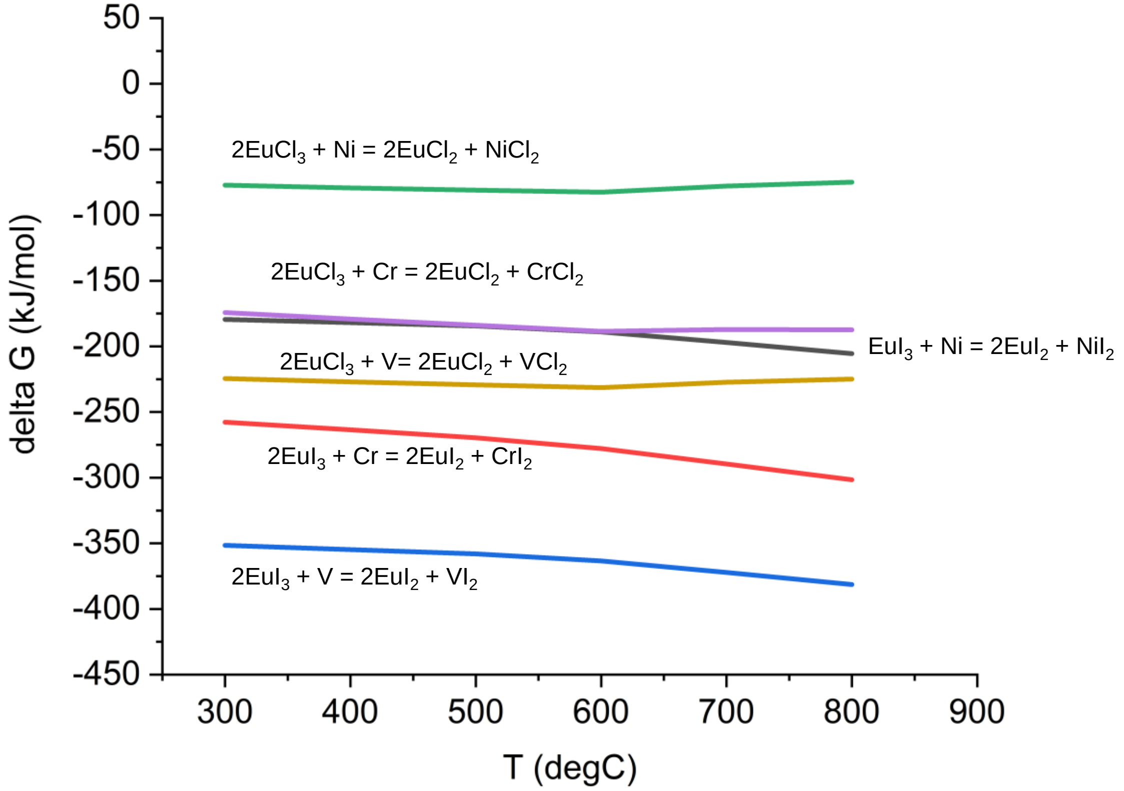
1cm long 1mm dia Ni wire





- Guo *et al.*, *Corrosion Sci.* **2020**, 163.

Impact of iodide species on this chemistry ?



Lower Gibbs free energy of reactions between EuI_3 with metals compared to $EuCl_3$

Impact of Eu^{3+} on Ni metal corrosion in iodide melt

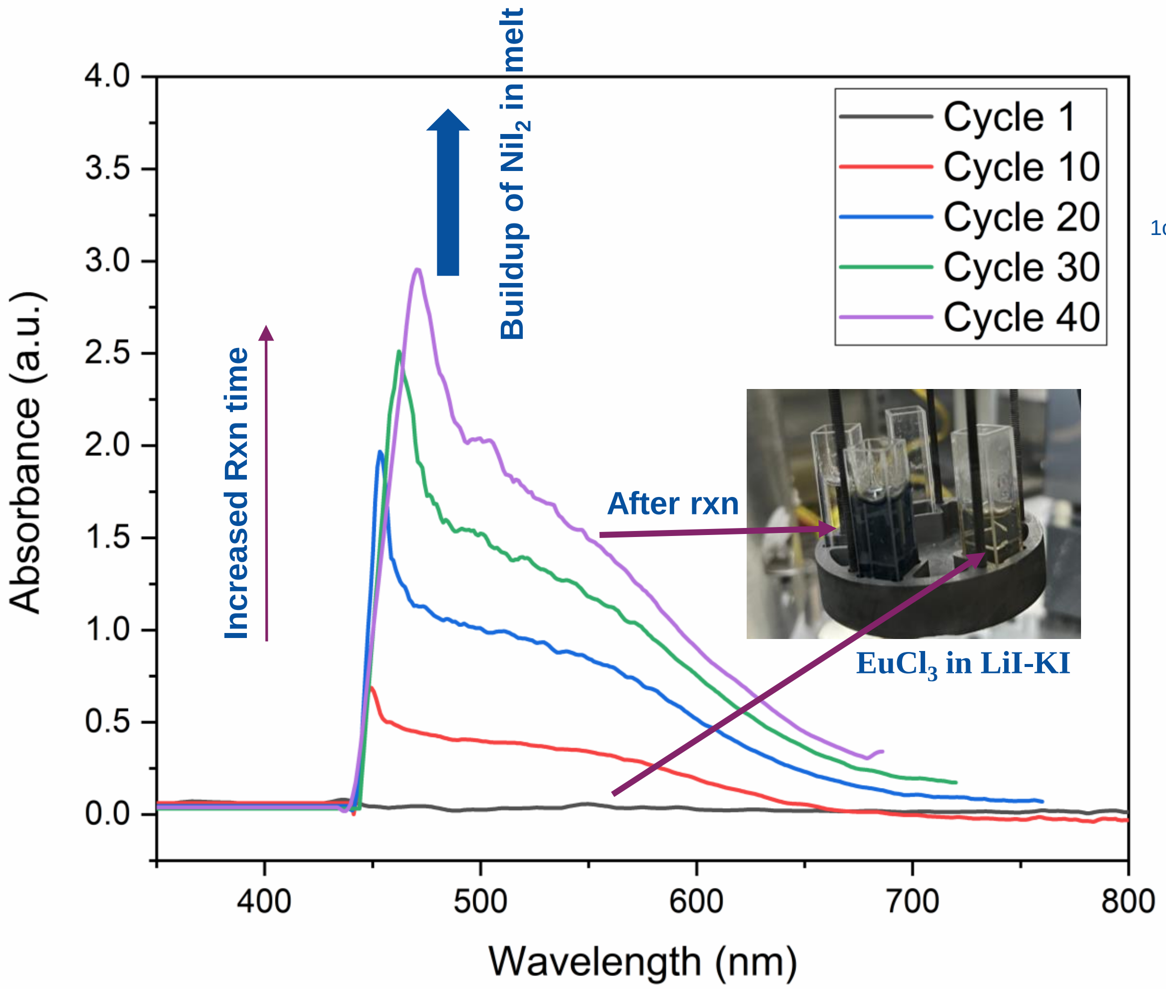
- Ni is quickly oxidized by Eu^{3+} in LiI-KI and brought into solution as NiI_2

$$2EuI_3 + Ni \leftrightarrow NiI_2 + 2EuI_2$$

- 4 min each reaction cycle , Total reaction time = 160 min
- As the reaction proceeds, dissolution rate decreases

Tasks planned for this study:

- ICP-MS analysis of salt before and after to characterize amount of Ni leached out in melt
- Ni wire surface to be characterized using electron microscopy in coordination with **Research Objective 3**



1cm long 1mm dia Ni wire

Ni wire in 1wt% $EuCl_3$ containing LiI-KI eutectic at 500°C

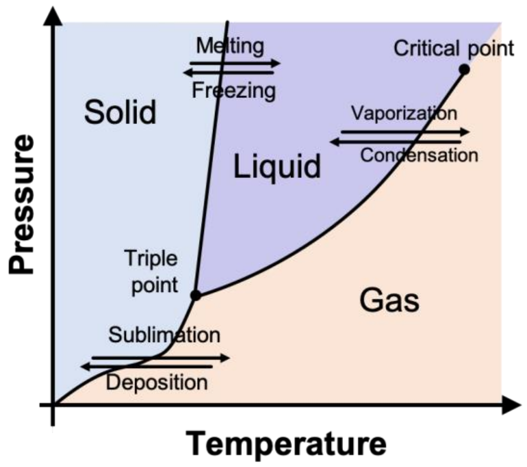
Rocio Rodriguez & **Ruchi Gakhar**

INL Staff Scientists

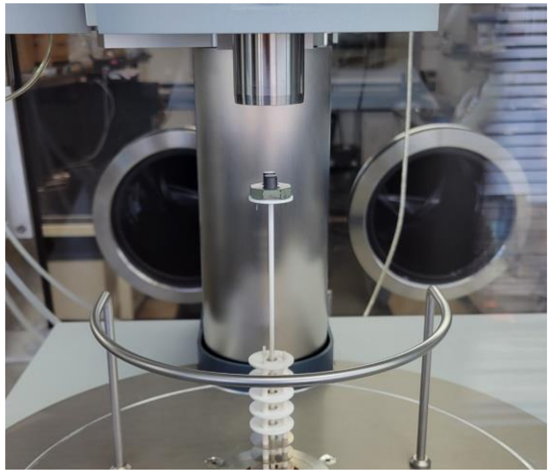
Impact of Alkali Metal Iodides on Crystal Structure and Thermodynamic Properties of LiCl-KCl Eutectic

Goal & methodology

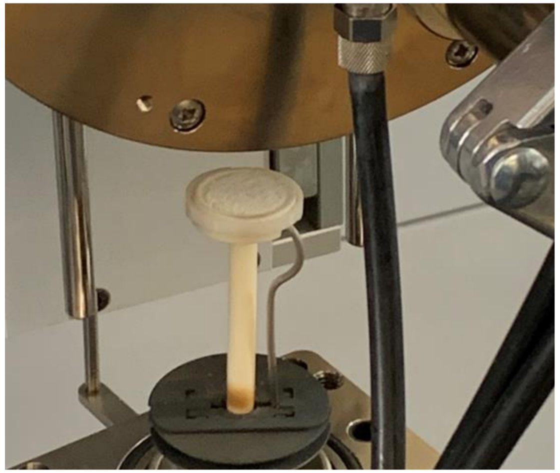
To investigate how the addition of iodide impacts the physicochemical properties of the base system



Thermodynamic calculations



Differential scanning calorimetry (DSC)



High-temperature X-ray diffraction (HT-XRD)

What is the importance of these studies?

The presence of certain concentrations of the solute (iodide) in the base salt may significantly:

Modify the melting temperature of the salt

Composition	Liquidus temperature, °C
LiCl-KCl (eutectic binary)	252.5
1 wt. [illegible]	[illegible]
3 wt. [illegible]	[illegible]
5 wt. [illegible]	[illegible]
10 wt. [illegible]	[illegible]
25 wt. % CsI – KCl – LiCl	337.5

Design and operation of MSRs

Alter the crystal structure of the base salt

Impact salts partition between liquid/solid phases

Safe handling and effective processing of spent nuclear fuel

Systems of interest

Base Salt (eutectic proportion)
LiCl-KCl
LiI-KI
NaCl-CsCl

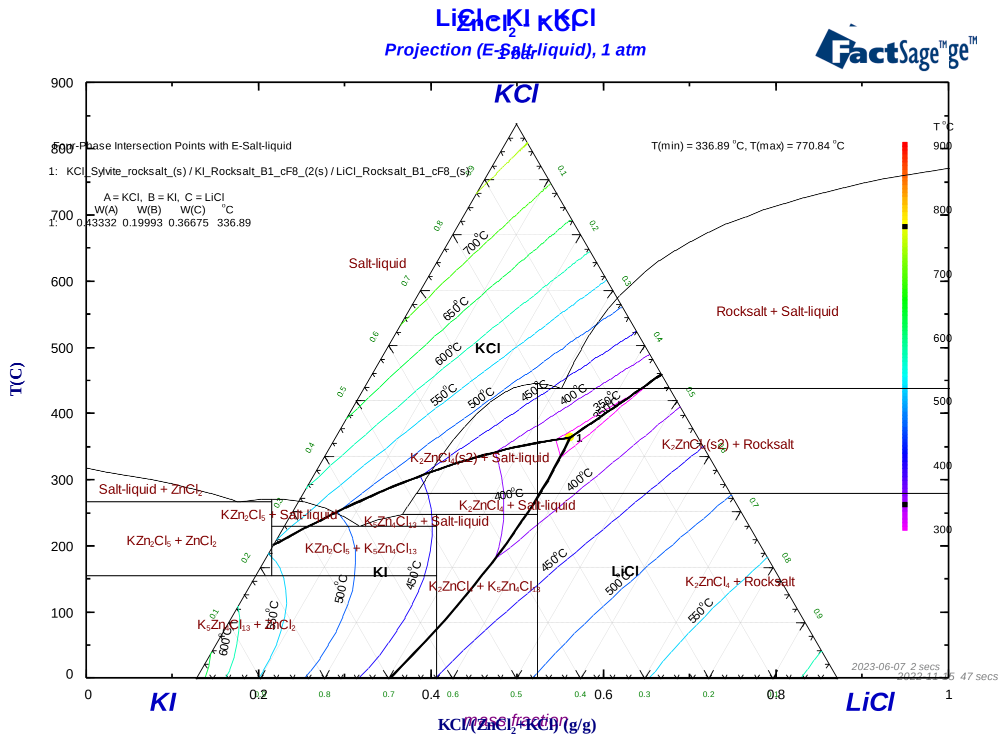
Solute
LiI
KI
CsI

Concentration (wt. %)
3, 5, 10, 20, 25

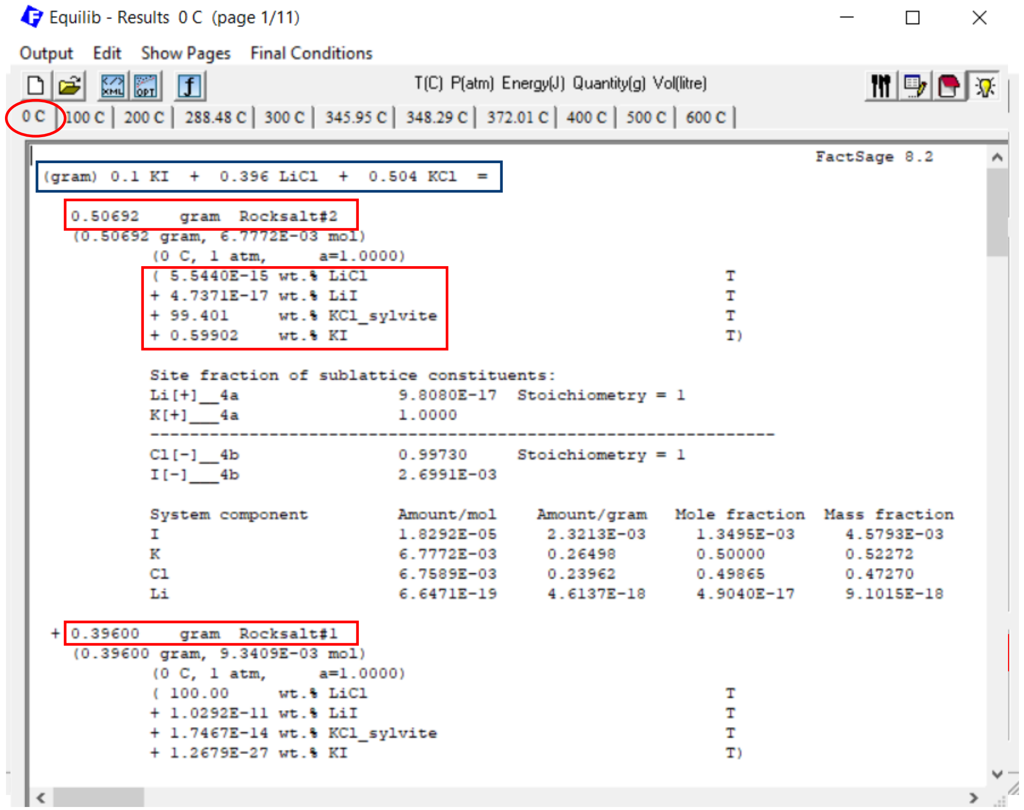
Thermodynamic calculations - FactSage

$$\Delta G^0 = \Delta H - T\Delta S$$

Phase diagram module



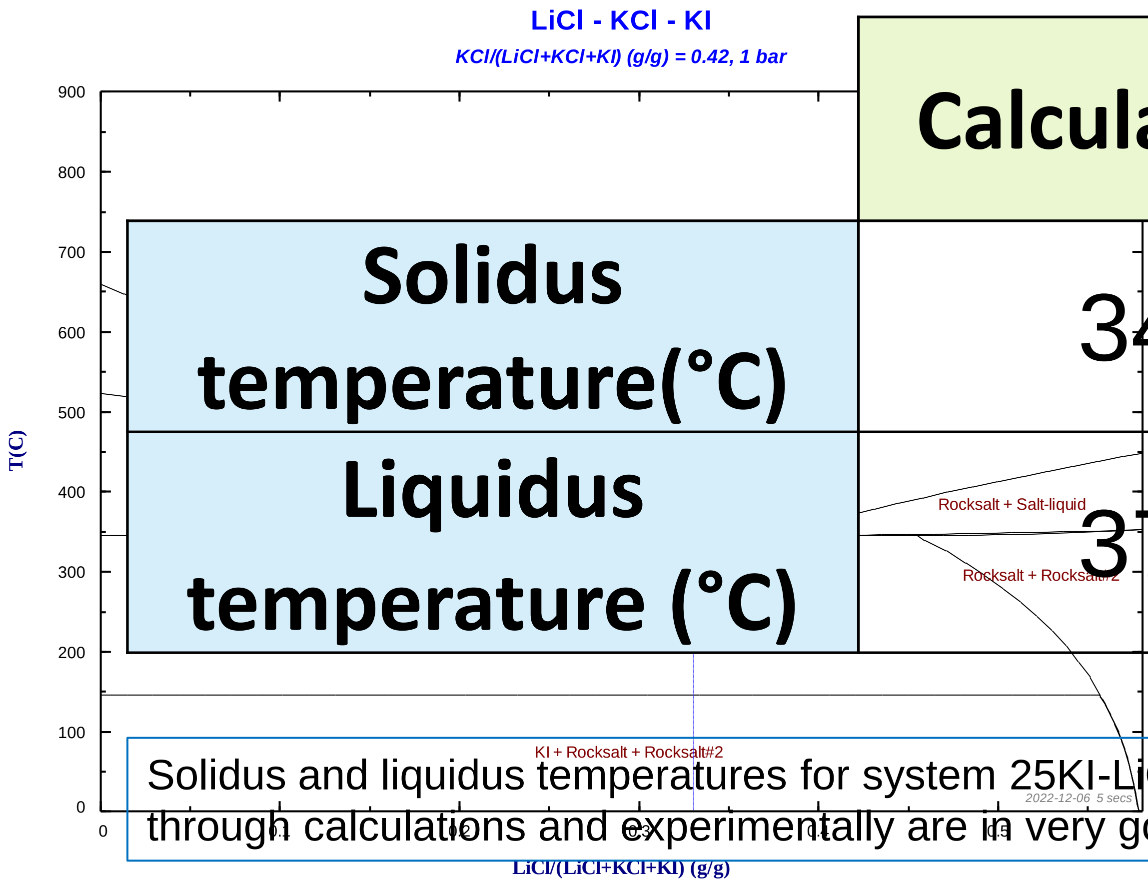
Equilibrium module

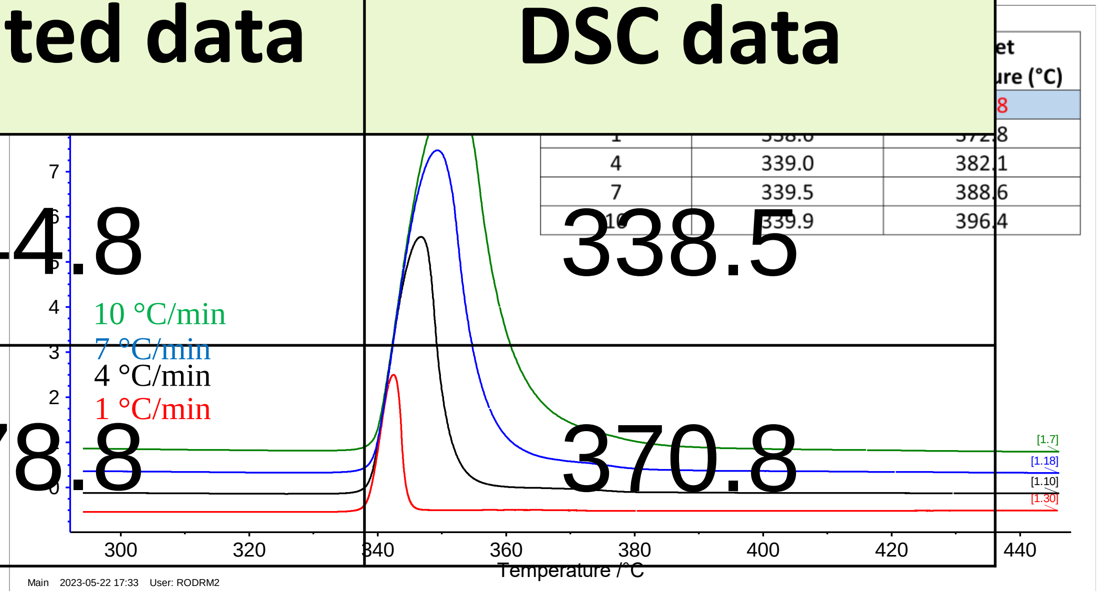


25 wt. % KI in LiCl-KCl

2-axis phase diagram with fixed KCl concentration

Calorimetric curves at 1, 4, 7 and 10 °C/min



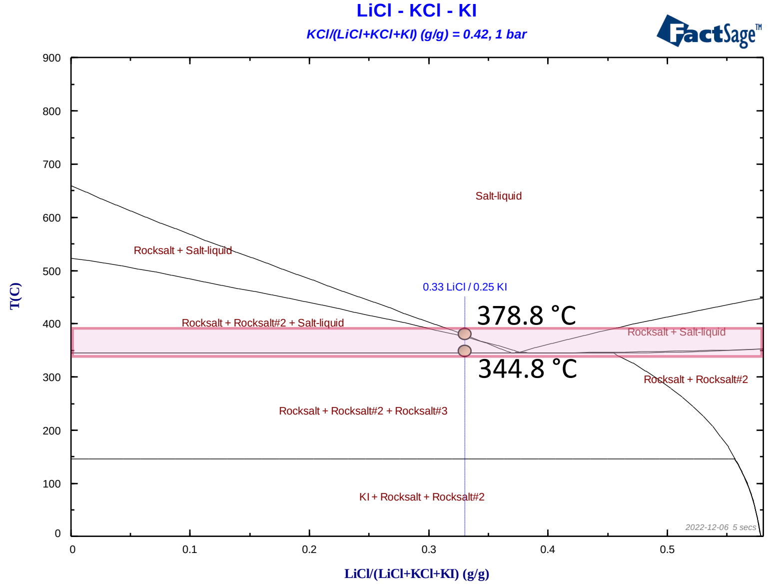


	Calculated data	DSC data
Solidus temperature(°C)	344.8	338.5
Liquidus temperature (°C)	378.8	370.8

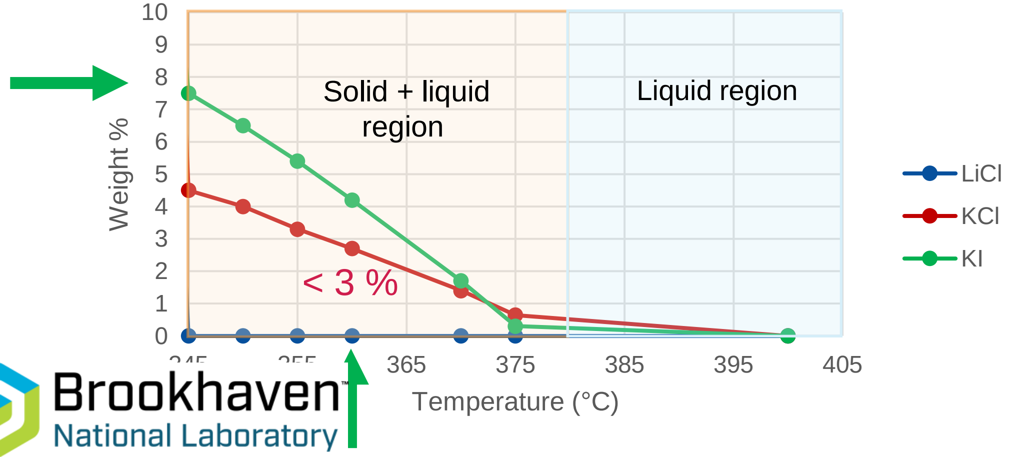
Solidus and liquidus temperatures for system 25KI-LiCl-KCl determined through calculations and experimentally are in very good agreement

Rodriguez Laguna, M. D.-R.; Holmbeck, G. P.; Topsakal, M.; Garcia, R. H.; Anderson, S. T.; Gakhar, R. (**2023**). e-Driven Phase Distribution of LiI and KI in LiCl-KCl Eutectic Salt*. Manuscript in preparation.

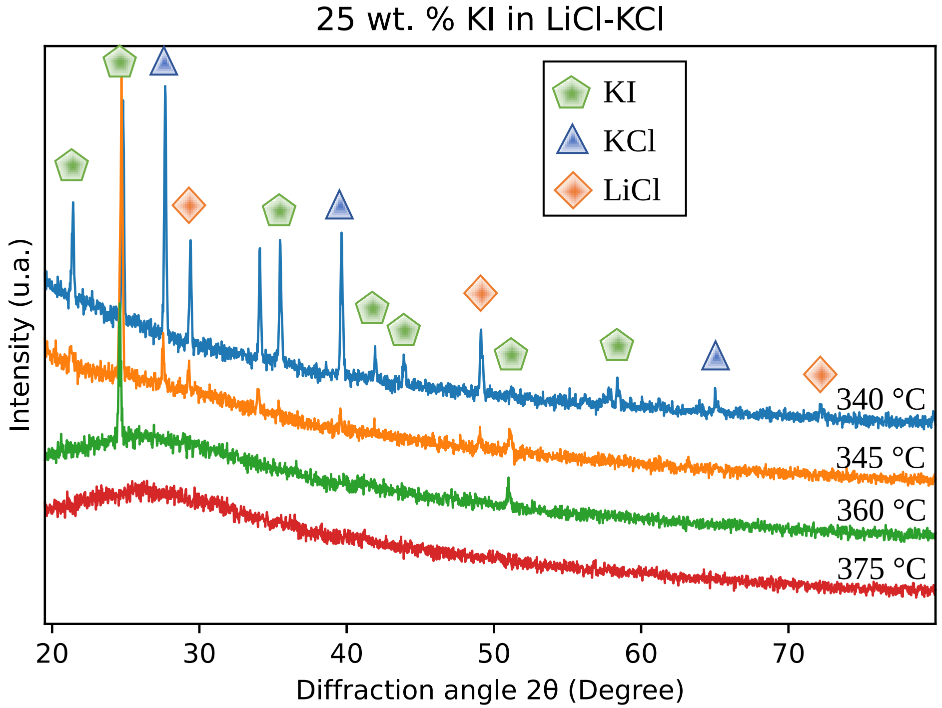
25 wt. % KI in LiCl-KCl: phase evolution as a f(T)



Solid fraction relative to total sample weight
25KI-LiCl-KCl



Phase evolution as a function of temperature



Phases present:

- KI, KCl & LiCl
- KI, KCl & LiCl
- KI
- None

Rodriguez Laguna, M. D.-R.; Holmbeck, G. P.; Topsakal, M.; Garcia, R. H.; Anderson, S. T.; Gakhar, R. (**2023**). *Temperature-Driven Phase Distribution of LiI and KI in LiCl-KCl Eutectic Salt*. Manuscript in preparation.

Conclusions

- The addition of iodide can substantially change the melting/crystallization temperature of the base salt.
- The DSC method, utilizing extrapolation of the onset and endset values (melting peak) to 0 °C/min, effectively determines the solidus and liquidus temperatures.
- The calculated phase diagrams align with the measured solidus and liquidus temperatures, while the thermal equilibrium data agrees with the phases detected using HT-XRD.
- FactSage software can be used as a guide to determine the effect of alkali metal iodides on the thermophysical properties and crystalline structure of the base salt.

Jacy K. Conrad
Staff Scientist

What are the transient and steady-state iodine species formed by the irradiation of high-temperature molten salts, and what are the fundamental radiation-induced mechanisms responsible for their formation and decay?

Research Objective #2

Overview of Research Objective #2

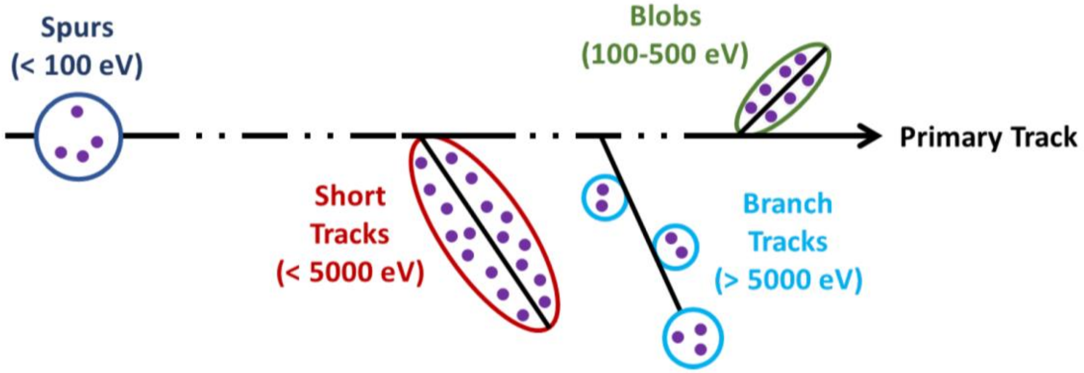
What are the transient and steady-state iodine species formed by the irradiation molten salts, and what are the fundamental radiation-induced mechanisms responsible for their formation and decay?

(i) Elucidation of chemical reaction mechanisms by time-resolved electron pulse radiolysis.

(ii) Steady-state radiation-induced bulk homogeneous phenomena.

(iii) Multiscale studies of high-temperature radiation-induced iodine reaction mechanisms.

Energy Transfer Events Along a Radiation Track



Jacy K. Conrad

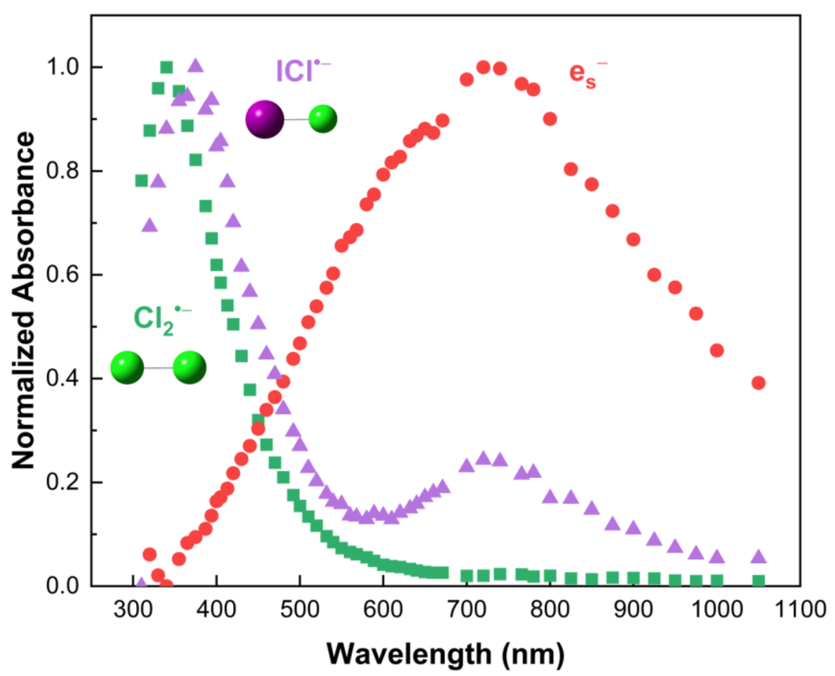
INL Staff Scientist

Radiation-Induced Kinetics and Speciation of Iodine in High Temperature Molten Salts

Brookhaven National Laboratory

INL Idaho National Laboratory

Focus Area #1 : Radiation-Induced Kinetics and Speciation of Iodine in High Temperature Molten Salts



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Cite this: *Phys. Chem. Chem. Phys.*, 2023, **25**, 16009

Impact of iodide ions on the speciation of radiolytic transients in molten LiCl–KCl eutectic salt mixtures†

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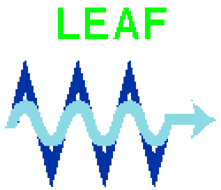
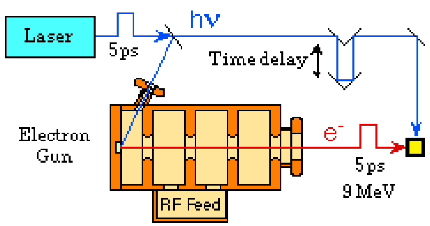
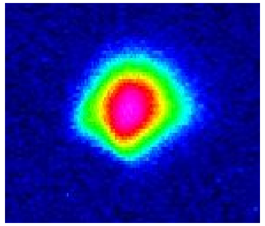
The fate of fission-product iodine is critical for the deployment of next generation molten salt reactor technologies, owing to its volatility and biological impacts if it were to be released into the environment. To date, little is known on how ionizing radiation fields influence the redox chemistry,







Radiation Chemistry of Molten Halide Salts

Direct Radiolysis

$M^{n+}X_n^{-} \rightsquigarrow e^{-}, X^{\bullet}, M^{n+}$

Electron Reactions

$e^{-} \rightarrow e_S^{-}$

$e_S^{-} + M^{n+} \rightarrow M^{(n-1)+}$

Halide Radical Reactions

$X^{\bullet} + X^{-} \rightarrow X_2^{\bullet-}$

$X_2^{\bullet-} + X_2^{\bullet-} \rightarrow X_3^{-} + X^{-}$

$X_3^{-} \rightleftharpoons X_2 + X^{-}$

With Iodine

$X^{\bullet} + I^{-} \rightarrow IX^{\bullet-}$

$IX^{\bullet-} + IX^{\bullet-} / X_2^{\bullet-} \rightarrow I_2X^{-}/IX_2^{-} + X^{-}$

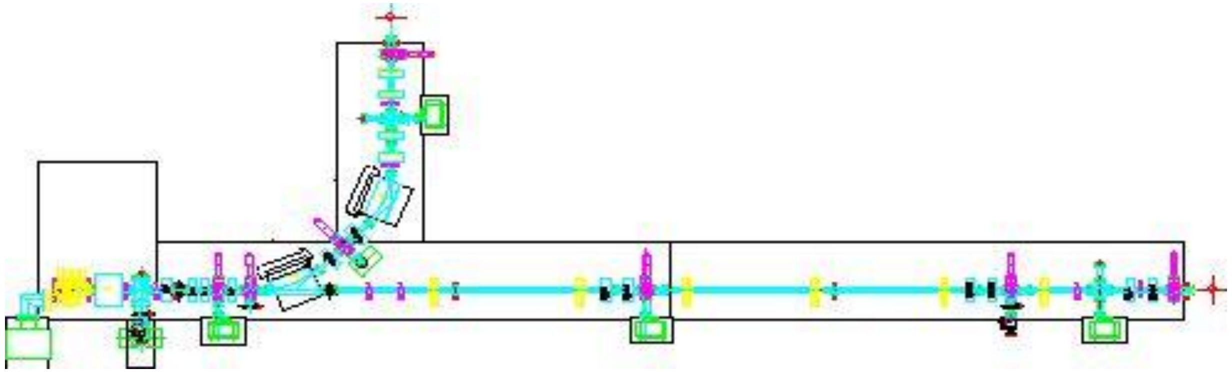
$I_2X^{-}/IX_2^{-} \rightleftharpoons IX + I^{-}/X^{-}$

- Impurities and other intentional additives may act as scavengers for the e_S^{-}, $X^{\bullet}$, and $X_2^{\bullet-}$

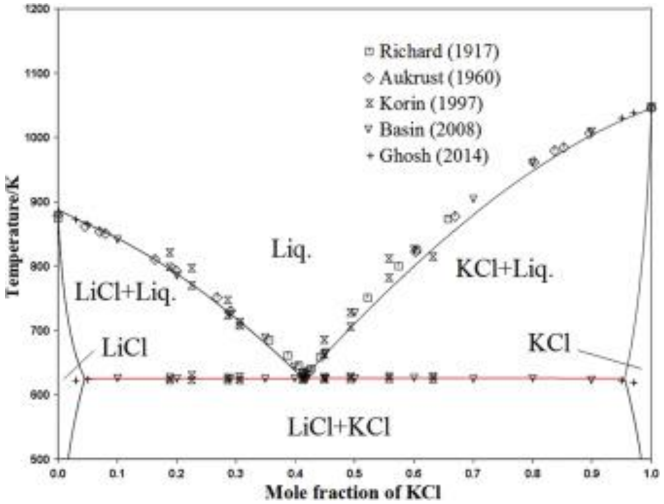
- Iwamatsu *et al.*, *Phys. Chem. Chem. Phys.* **2022**, *24*, 25088.
- Conrad *et al.*, *Phys. Chem. Chem. Phys.* **2023**, *25*, 16009.

Experimental Methods

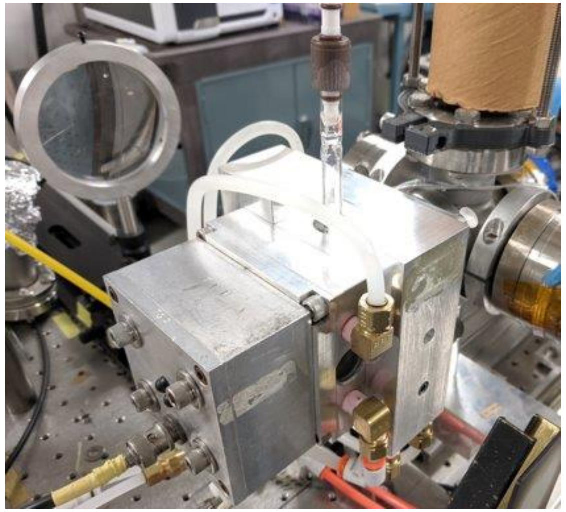
- Prepared salt mixtures of up to 10 wt.% KI in LiCl-KCl eutectic under Ar atmosphere.
- Used a custom-built high temperature cell holder to heat to temperatures from 400 – 700 ºC.
- Used the Brookhaven National Laboratory (BNL) Laser Electron Accelerator Facility (LEAF) to perform picosecond pulse radiolysis with transient absorption spectroscopy techniques.



Phase Diagram for LiCl-KCl Mixtures



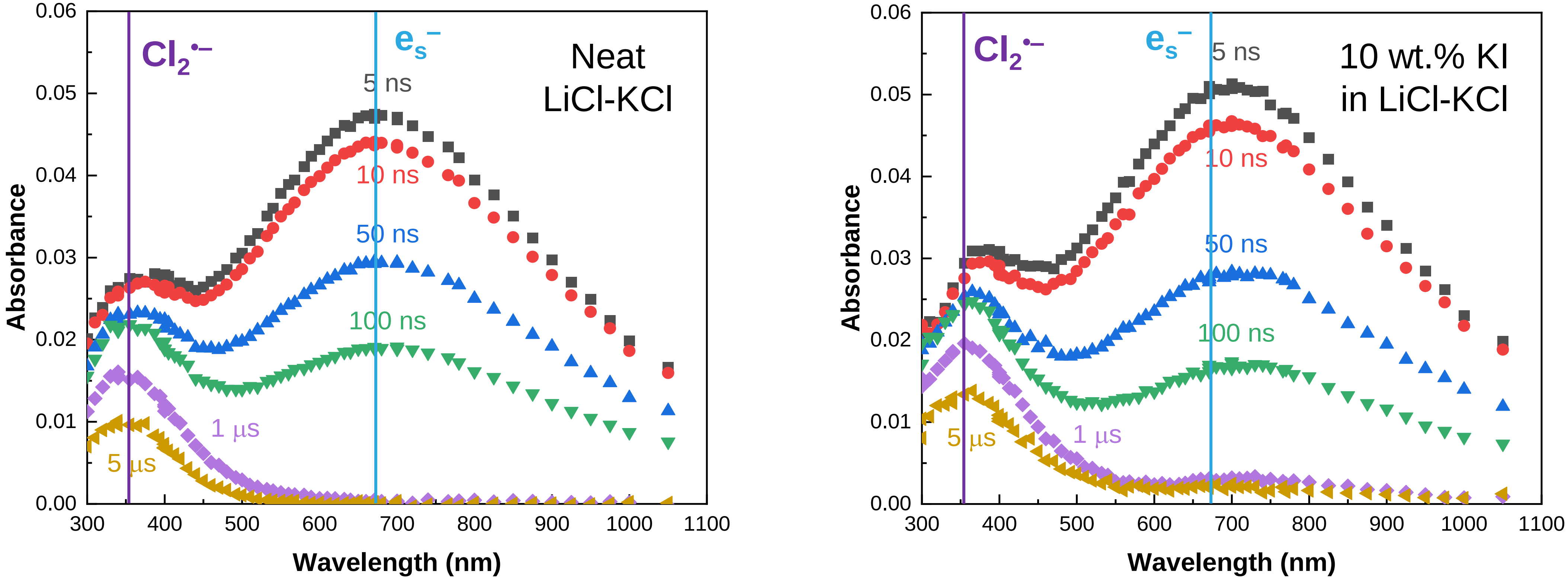
High Temperature Cell Holder



- Phillips *et al.*, *Rev. Sci. Instr*. **2020**, 91 (8), 083105.
- Wishart, Cook, Miller, *Rev. Sci. Instr.* **2004**, 75 (11), 4359–4366.

Results: Transient Absorption Spectra

Spectra at different times after the electron pulse irradiation of salt mixtures at 400 ºC



- Short-lived species in the visible and nIR, attributed to the e_s^-.
- Longer-lived species absorbing in the UV, attributed to $Cl_2^{\bullet-}$.
- At least one additional species in the 10 wt.% KI spectrum.

Brookhaven National Laboratory

- Iwamatsu *et al., Phys. Chem. Chem. Phys.* **2022**, *24*, 25088.
- Conrad *et al.*, *Phys. Chem. Chem. Phys.* **2023**, *25*, 16009.

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Discussion: Transient Absorption Spectra

Potential Halide Radical Anions

$Cl^-/I^- \rightsquigarrow Cl^\bullet/I^\bullet + e^-$

$Cl^\bullet + Cl^- \rightarrow Cl_2^{\bullet-}$

$Cl^\bullet + I^- \rightarrow ICl^{\bullet-}$

$I^\bullet + Cl^- \rightarrow ICl^{\bullet-}$

$I^\bullet + I^- \rightarrow I_2^{\bullet-}$

Electron Fractions of the Salt Mixtures

[KI] (wt.%)	Electron Fraction (%)			
	I^-	Cl^-	Li^+	K^+
0.05	0.04	**67.34**	4.34	28.29
0.11	0.07	**67.31**	4.34	28.28
0.54	0.36	**67.04**	4.32	28.27
1.07	0.73	**66.71**	4.30	28.25
5.11	3.49	**64.23**	4.14	28.13
10.00	6.87	**61.20**	3.94	27.99

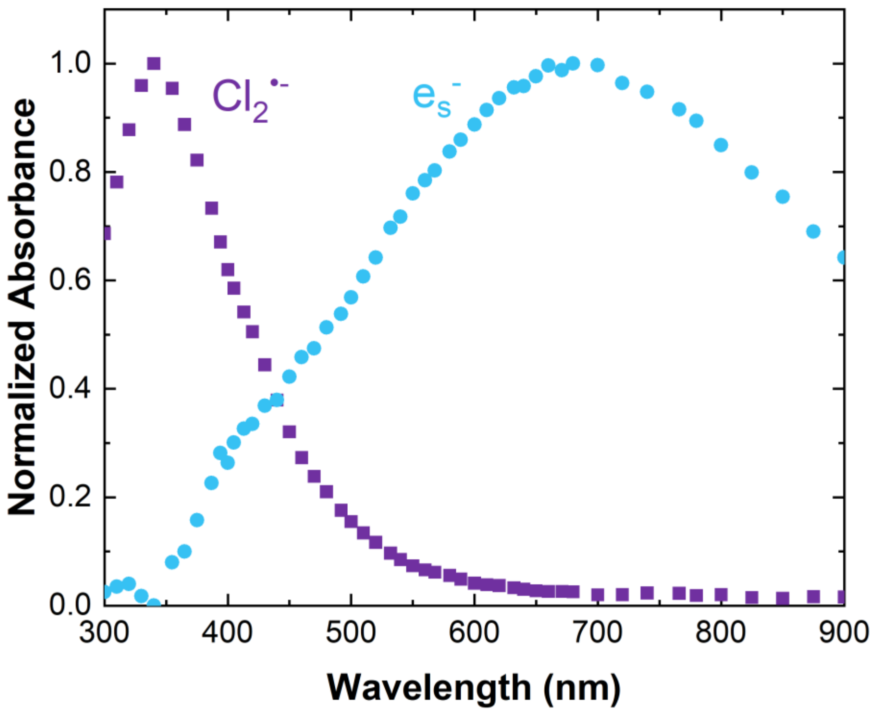
- Probable yields based on relative abundance of species: $Cl_2^{\bullet-} > ICl^{\bullet-} \gg I_2^{\bullet-}$

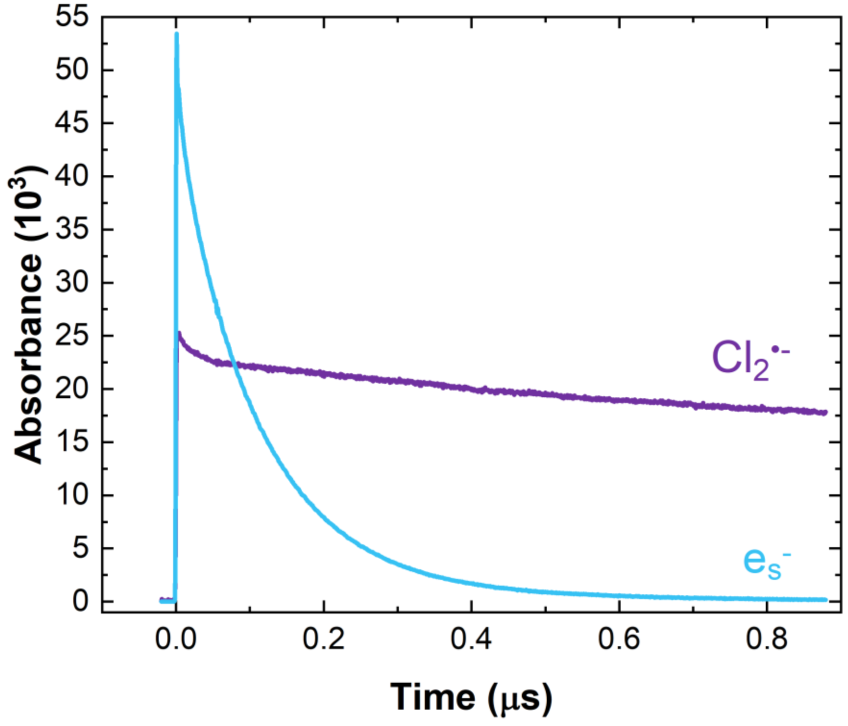
- Conrad *et al.*, *Phys. Chem. Chem. Phys.* **2023**, *25*, 16009.

Results: Deconvoluted Spectra and Kinetics

- *Spectro-Kinetic Analysis* (*SK-Ana*) software was used to deconvolute the overlapping chemical species for the first µs after the electron pulse.

Normalized spectra and kinetics after the electron pulse irradiation of neat LiCl-KCl eutectic at 400 °C



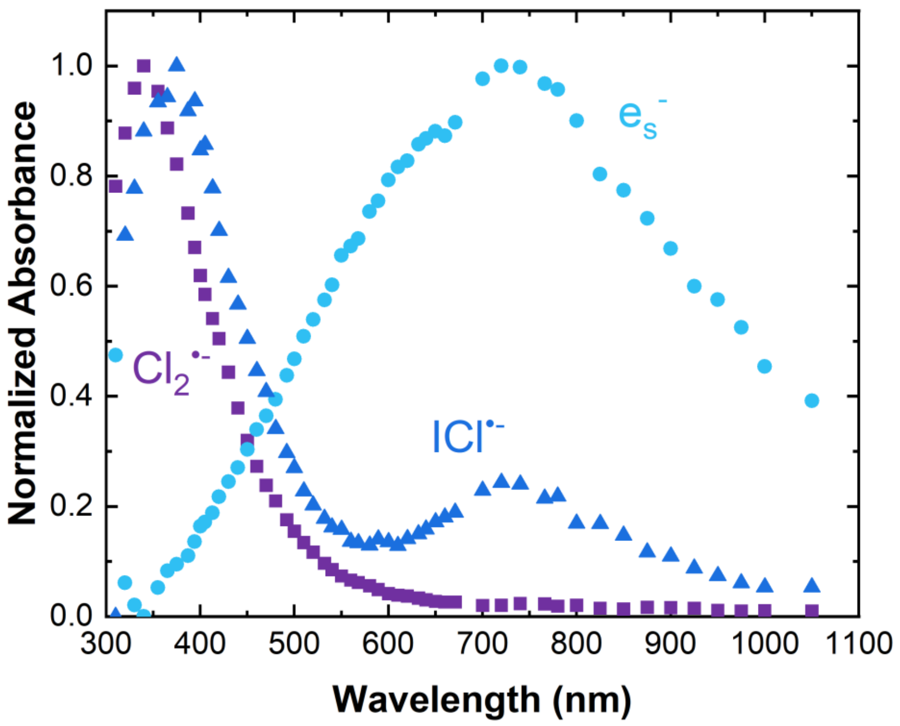


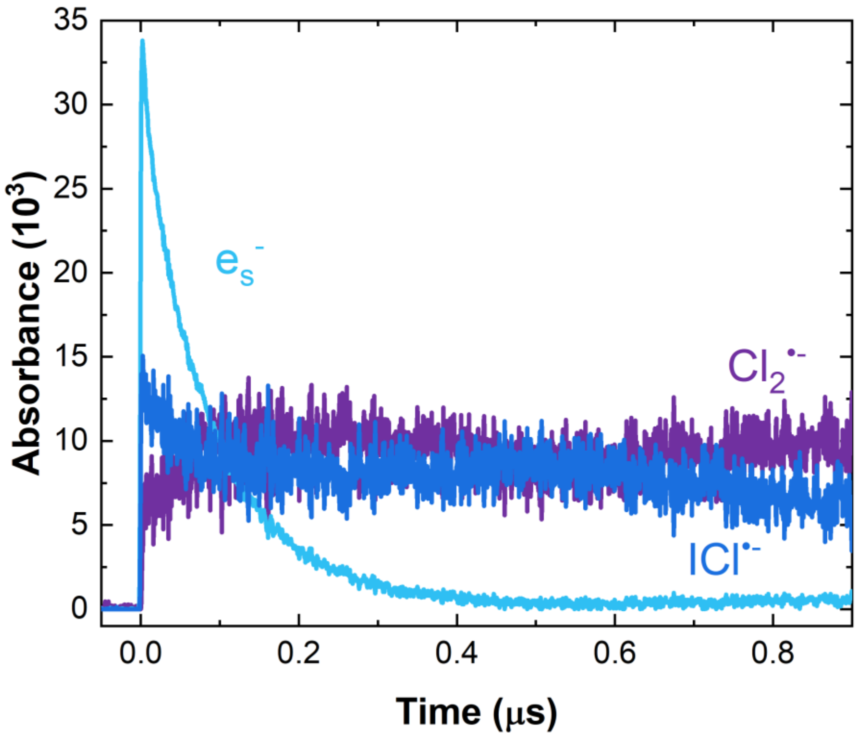
- P. Pernot, SK-Ana: Analysis of Spectro-Kinetic Data, **2018**.
- Iwamatsu *et al.*, *Phys. Chem. Chem. Phys.* **2022**, *24*, 25088

Results: Deconvoluted Spectra and Kinetics

- *Spectro-Kinetic Analysis* (*SK-Ana*) software was used to deconvolute the overlapping chemical species for the first µs after the electron pulse.

Normalized spectra and kinetics after the electron pulse irradiation of 10 wt.% KI in LiCl-KCl eutectic at 400 °C





Brookhaven National Laboratory

- P. Pernot, SK-Ana: Analysis of Spectro-Kinetic Data, **2018**.
- Iwamatsu *et al.*, *Phys. Chem. Chem. Phys.* **2022**, *24*, 25088

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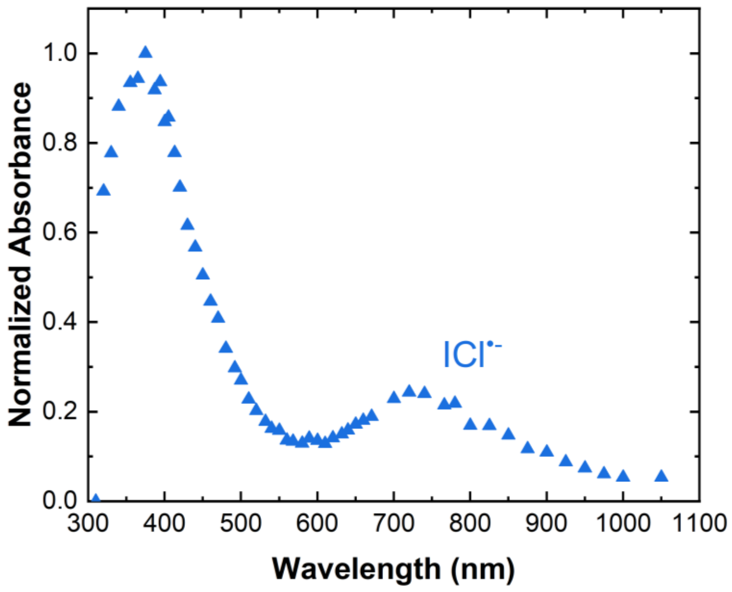
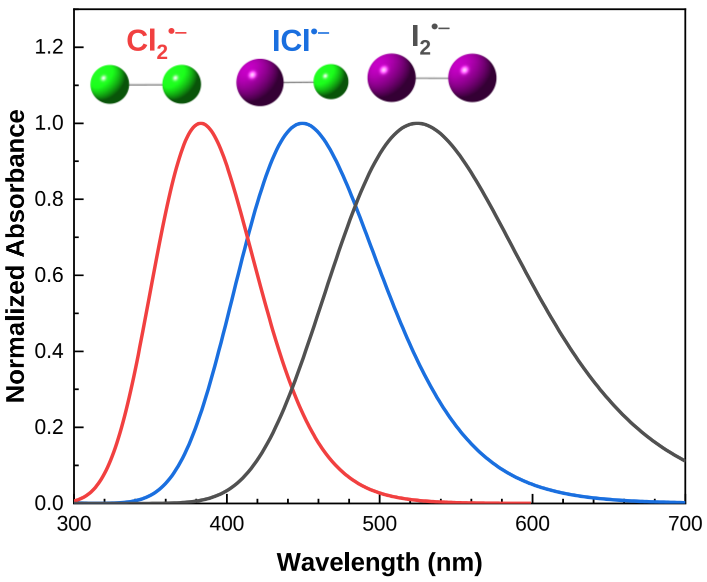
Discussion: $ICl^{\bullet-}$ Spectrum

- Comparison with spectrum from gas phase TD-DFT.
- Not consistent with $I_2^{\bullet-}$ spectrum measured at 800 ºC.

Spectrum Assigned to $ICl^{\bullet-}$ at 400 °C

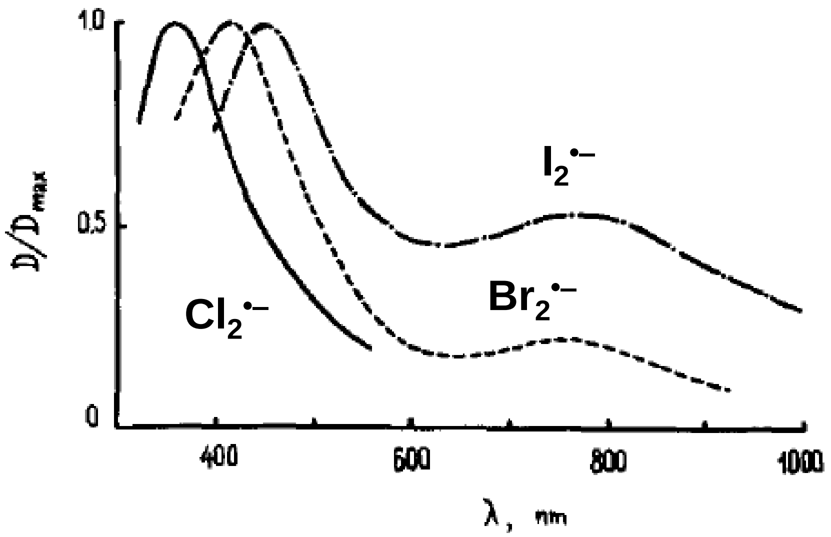


TD-DFT Gas-Phase Absorption Spectra



Spectra of $X_2^{\bullet-}$ at 800 °C, X = Cl, Br, I

(Pikaev, Makarov, Zhukova (1982) Radiat. Phys. Chem.)

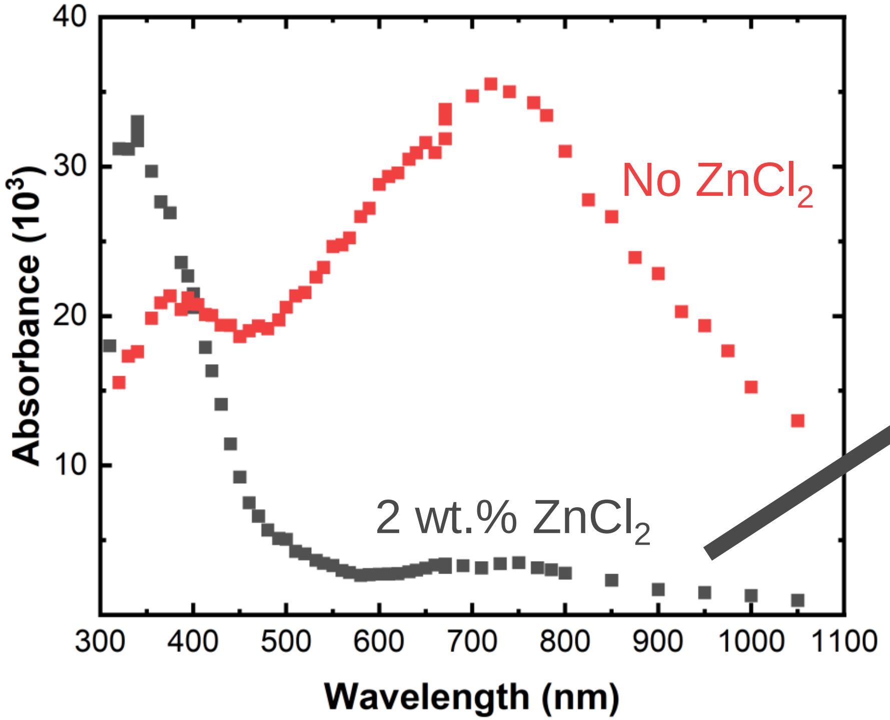


- Conrad *et al.*, *Phys. Chem. Chem. Phys.* **2023**, *25*, 16009.

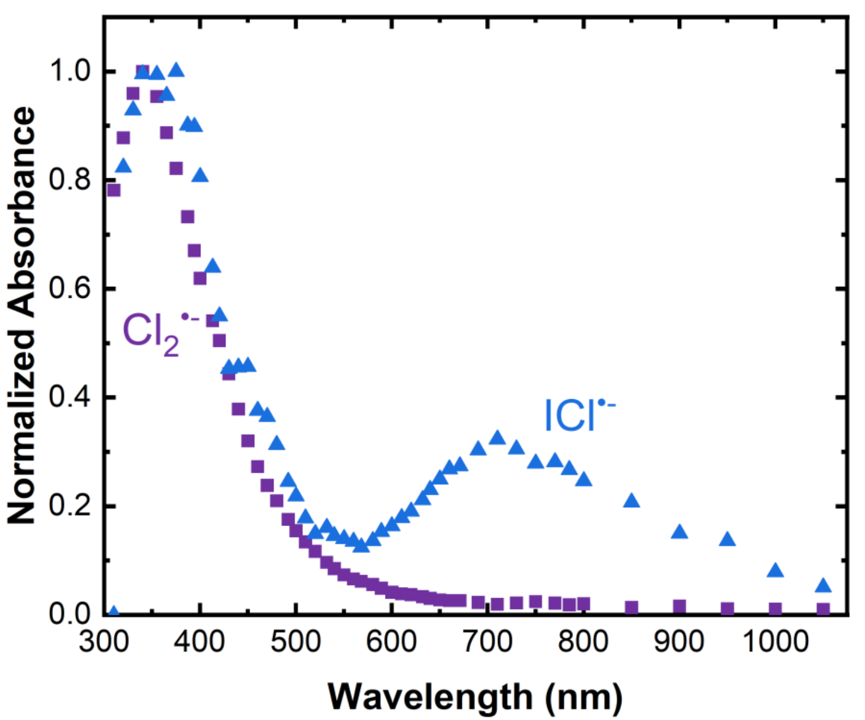
Result: $ICl^{\bullet-}$ Spectrum with 2 wt.% $ZnCl_2$

- When $ZnCl_2$ is added to system: **e^-/e_S^- + Zn^{2+} → Zn^+**

Transient absorption spectra 5 ns after electron pulse irradiation of 10 wt.% KI in LiCl-KCl



Normalized spectra from *SK-Ana* deconvolution of the $ZnCl_2$ containing salt mixture at 400 ° C

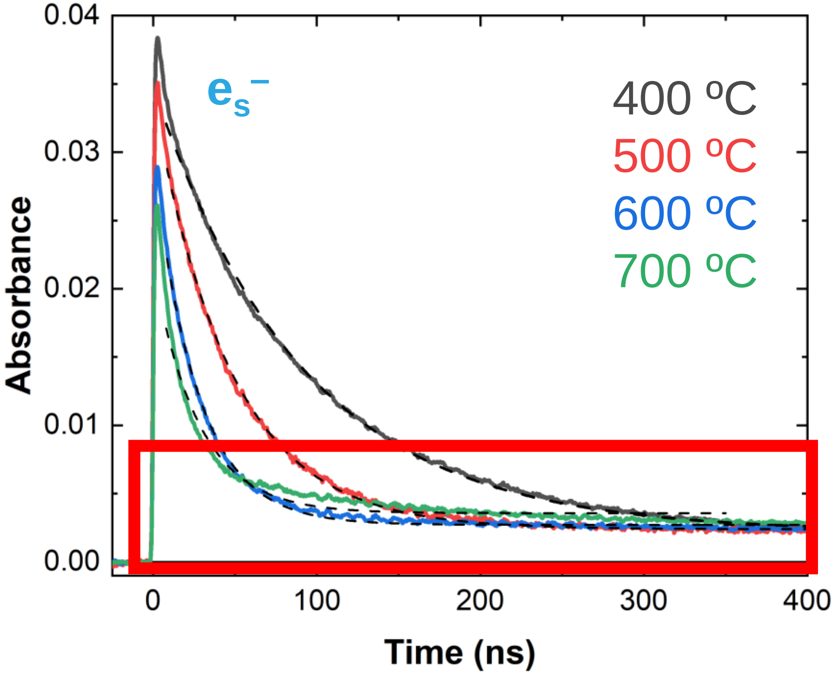


- Conrad *et al.*, *Phys. Chem. Chem. Phys.* **2023**, *25*, 16009.

Results: Kinetics of Electron Decay

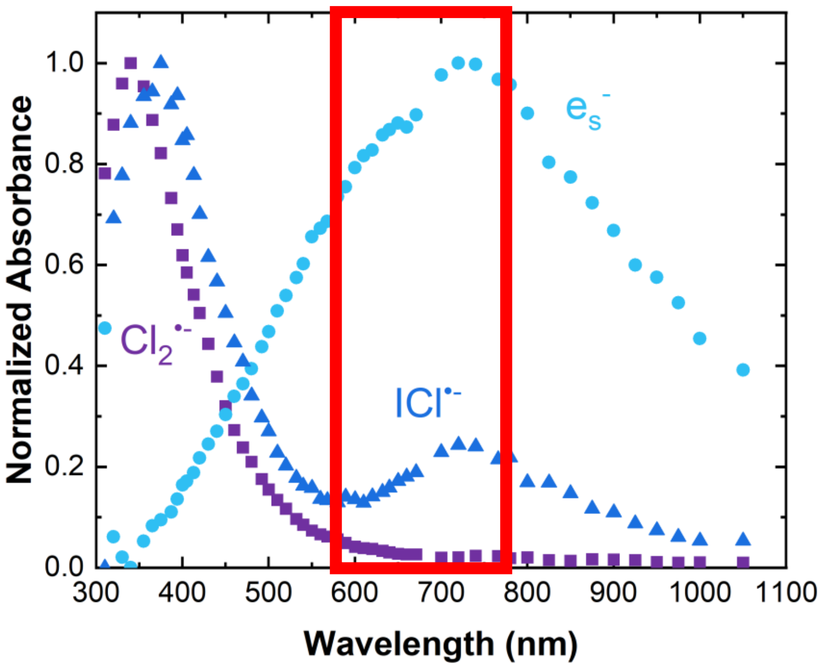
- Decay of the e_s^- is not affected by the addition of KI.
- Showed linear Arrhenius behavior for T = 400 – 700 ºC.

Transient absorbance at 671 nm after electron pulse irradiation of 10 wt.% KI in LiCl-KCl



Not returning to baseline

Normalized transient spectra of 10 wt.% KI in LiCl-KCl eutectic at 400 ºC

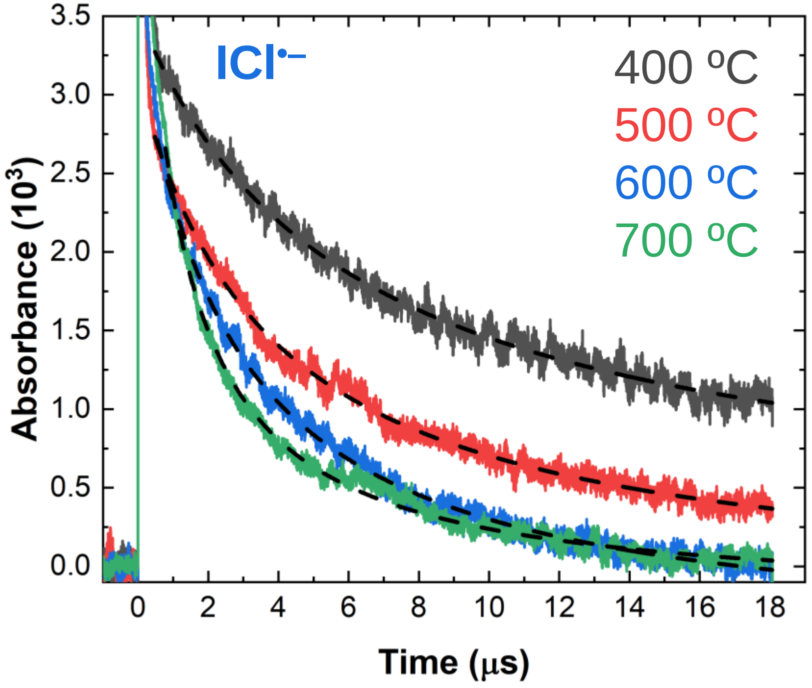


- Conrad *et al.*, *Phys. Chem. Chem. Phys.* **2023**, *25*, 16009.

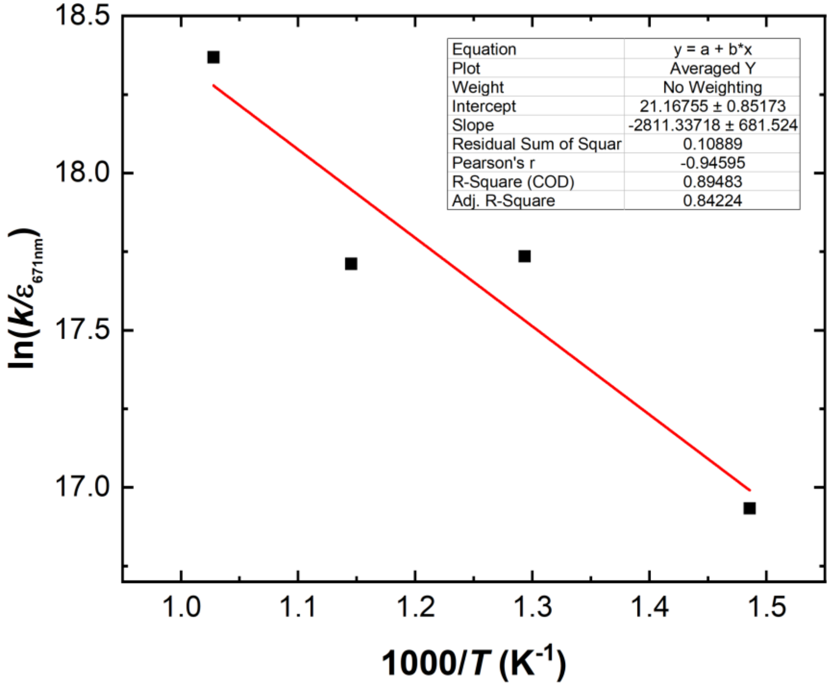
Results: Kinetics of ICl•– Decay

- Can be fitted with a single second-order decay function.
- Reaction rate increases with temperature.

Transient absorbance at 671 nm after e_s^- decay in 10 wt.% KI in LiCl-KCl



ln(k/ε_{671nm}) vs 1/T for 10 wt.% KI in LiCl-KCl



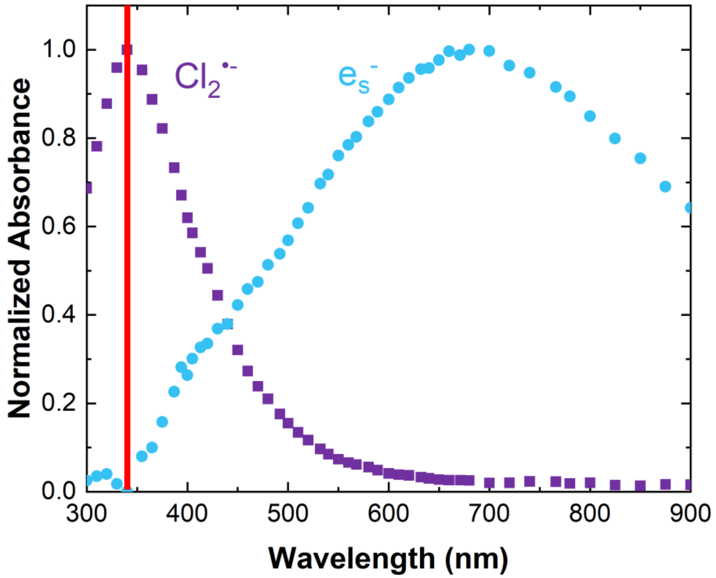
- Conrad *et al.*, *Phys. Chem. Chem. Phys.* **2023**, *25*, 16009.

Discussion: Mechanism of $Cl_2^{\bullet-}$ Decay

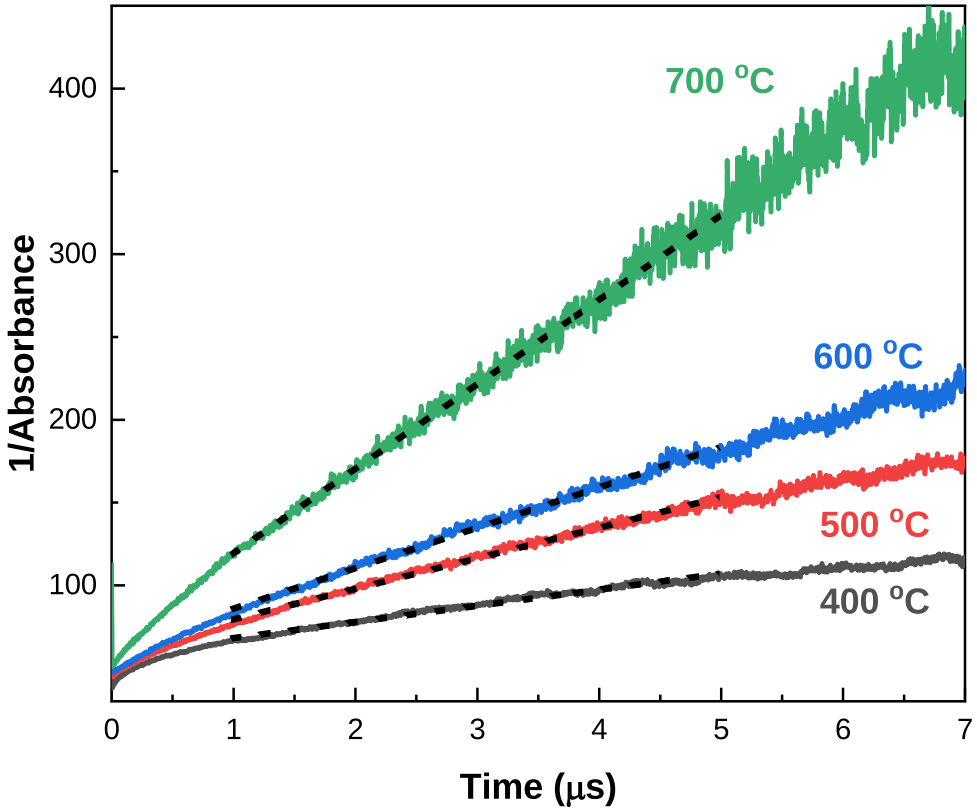
- In neat LiCl-KCl eutectic, $Cl_2^{\bullet-}$ is primarily consumed via disproportionation:

$$Cl_2^{\bullet-} + Cl_2^{\bullet-} \rightarrow Cl_3^{-} + Cl^{-}$$

Transient spectra after the electron pulse irradiation of neat LiCl-KCl eutectic at 400 °C



Fitted 1/Abs after the electron pulse irradiation of neat LiCl-KCl eutectic at 340 nm



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- Conrad *et al.*, *Phys. Chem. Chem. Phys.* **2023**, *25*, 16009.

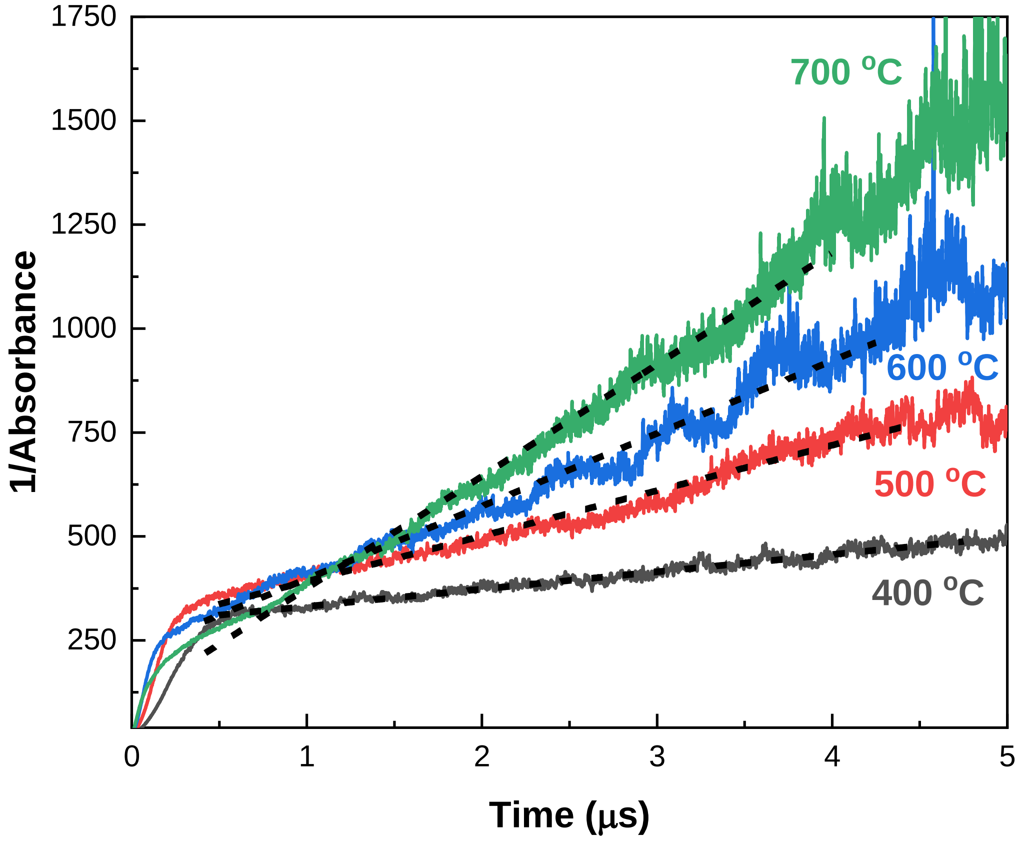
Discussion: Mechanism of $ICl^{\bullet-}$ Decay

- Similarly, for 10 wt.% KI in LiCl-KCl at 671 nm, where only $ICl^{\bullet-}$ is absorbing, the 1/Abs plot is linear, likely indicating the following mechanism:

$$ICl^{\bullet-} + Cl_2^{\bullet-} \rightarrow ICl_2^{-} + Cl^{-}$$

- In addition, E_a **= 23.4 ± 5.7 kJ mol**$^{-1}$ is similar to the values reported for $Cl_2^{\bullet-}$ disproportionation (E_a **= 24–26 kJ mol**$^{-1}$).

Fitted 1/Abs after the electron pulse irradiation of 10 wt.% KI in LiCl-KCl eutectic at 671 nm



- Iwamatsu *et al.*, *Phys. Chem. Chem. Phys.* **2022**, *24*, 25088
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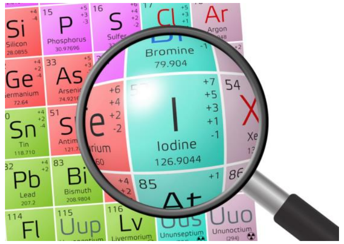
Summary

Objectives:

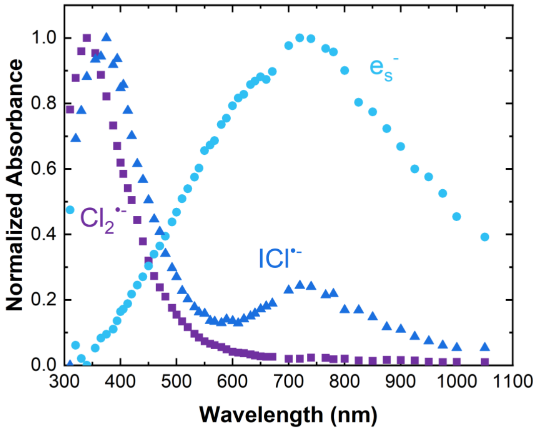
- To understand the fundamental radiation-induced behavior of iodine in molten salt environments.



Approach:

- Time resolved picosecond pulsed electron measurements of kinetics and transient absorption spectra for e_s^-, $Cl_2^{\bullet -}$, and $ICl^{\bullet -}$

Transient spectra of 10 wt.% KI in LiCl-KCl eutectic at 400 °C

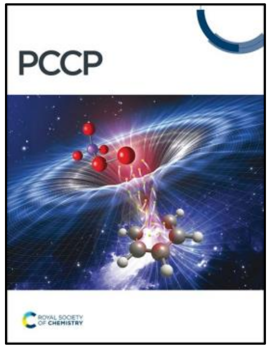


Outcome:

- First measurements of iodide ions in molten chloride salts.
- First spectrum and lifetime of $ICl^{\bullet -}$ reported.
- Manuscript submitted to *Physical Chemistry Chemical Physics*:

Conrad *et al.* (2023) Impact of Iodide Ions on the Speciation of Radiolytic Transients in Molten LiCl-KCl Eutectic Salt Mixtures. *Phys. Chem. Chem. Phys.* 25, 16009 – 16017.

DOI: 10.1039/d3cp01477k



Alejandro Ramos-Ballesteros
INL Postdoctoral Researcher

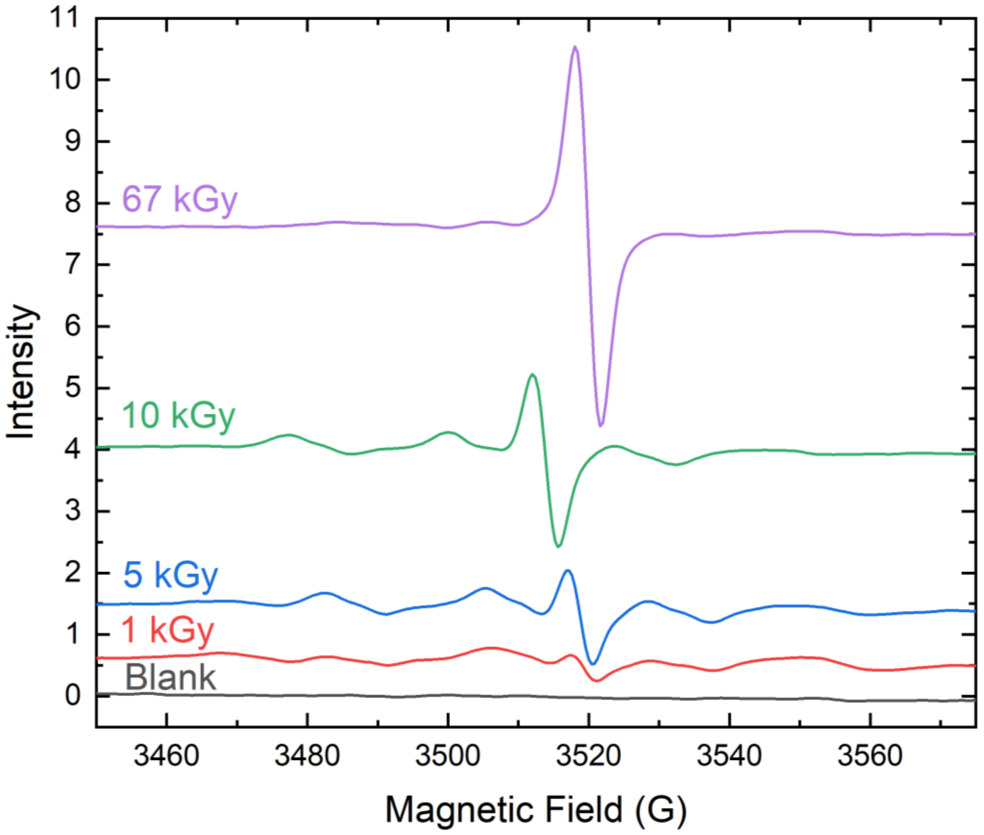
Solid-State Radiolysis of Iodide Salts

Brookhaven National Laboratory

INL Idaho National Laboratory

Focus Area #2 : Solid-State Radiolysis of Iodide Salts

Electron Paramagnetic Resonance (EPR) Spectrum of solid KI with Increasing Gamma Dose



Benchtop EPR Instrument

New Postdoctoral Researcher:
Alejandro Ramos-Ballesteros

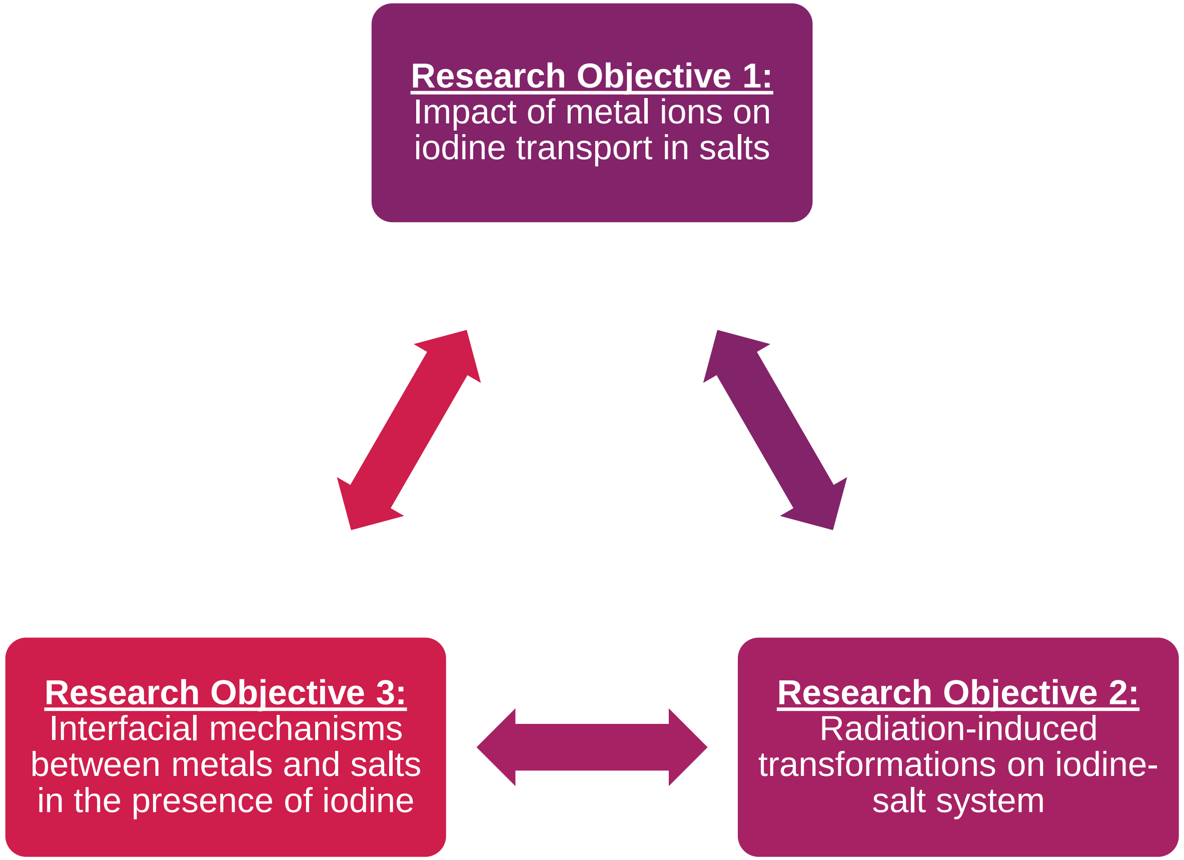
Trishelle Copeland-Johnson & **Simerjeet Gill**
INL and BNL Staff Scientists

Understand and Predict Radiation-Induced Iodine Speciation, Chemistry, and Transport in High-Temperature Molten Salts

Research Objective #3

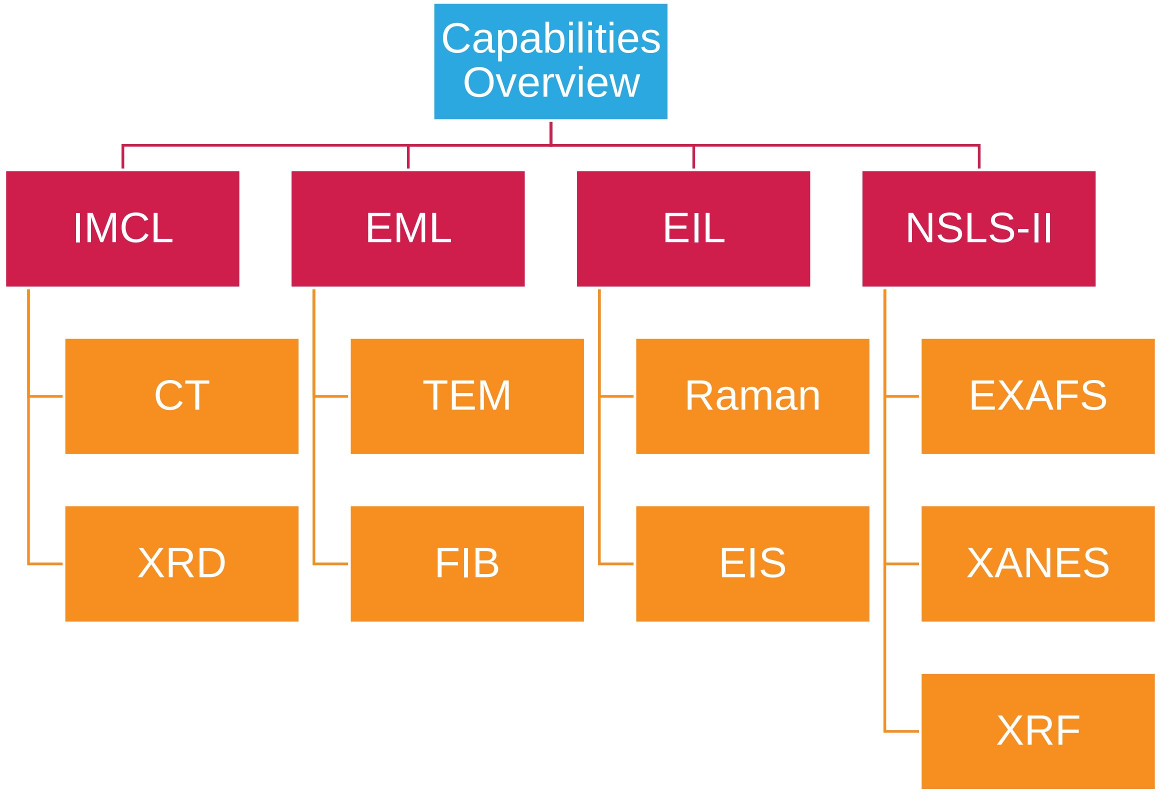
Overview of Research Objective 3

- ***How do interfacial processes affect the formation, speciation, and transport of iodine in molten salts?***
  - Understanding the interfacial reactivity of iodine under extreme conditions is critical for the development of MSR technologies.
  - Alloying elements in metal alloys play a critical role in controlling the dissolution and corrosion kinetics of different materials in molten salts.
- These activities are in conjunction with other Research Objectives…

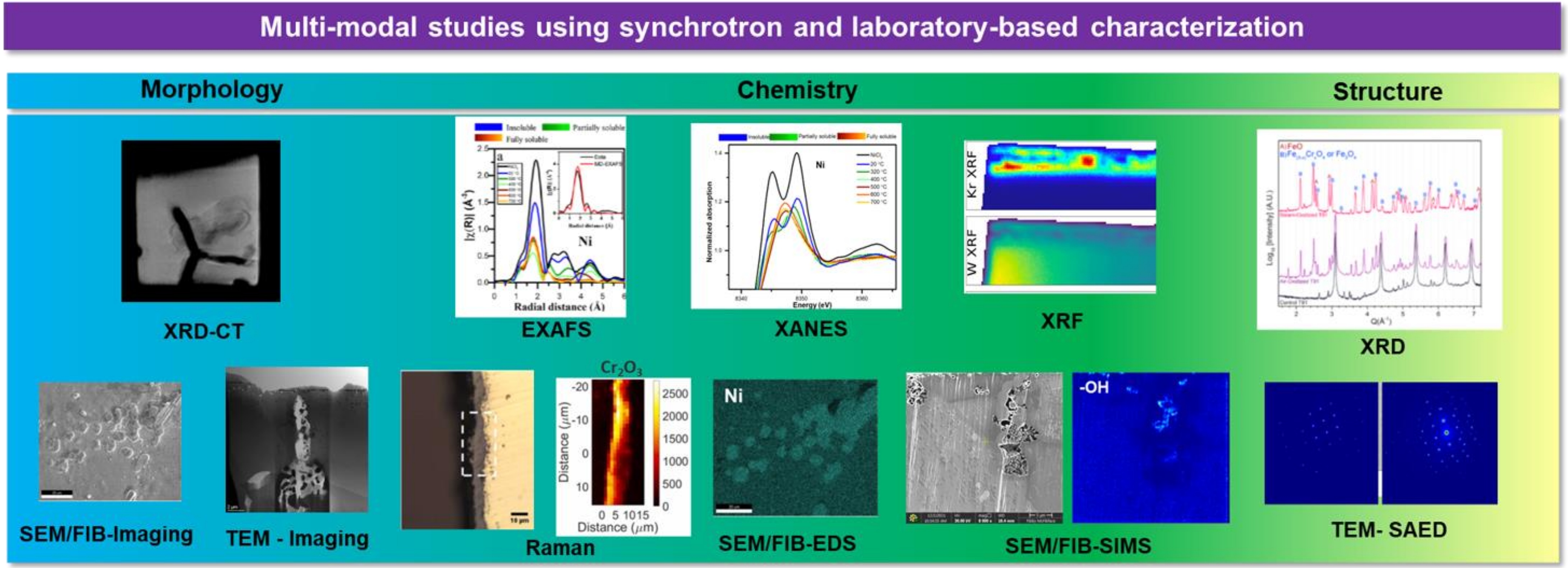


Overview of Research Objective 3 (cont.)

- **The goal of Research Objective 3 is to understand the influence of interfacial processes on determining the final disposition of iodine in high temperature molten salts, notably the structure and chemical speciation of iodine at metal-salt interfaces.**
- This goal will be accomplished through use of microscopy, spectroscopy techniques such as:
  - Scanning electron microscopy/focused-ion beam (SEM/FIB)
  - Transmission electron microscopy (TEM)
  - X-ray computed tomography microscopy (CT)
  - Raman spectroscopy
  - Electrochemical Impedance Spectroscopy (EIS)
- These techniques are available at INL through:
  - Irradiated Materials Characterization Laboratory (IMCL)
  - Electron Microscopy Laboratory (EML)
  - Energy Innovation Laboratory (EIL)
- In-situ studies at National Synchrotron Light Source II (NSLS-II) at Brookhaven National Laboratory
  - Full X-ray Imaging (FXI)
  - X-ray Fluorescence Microprobe (XFM)



How will the evolution of metal-salt corrosion interfaces be characterized?



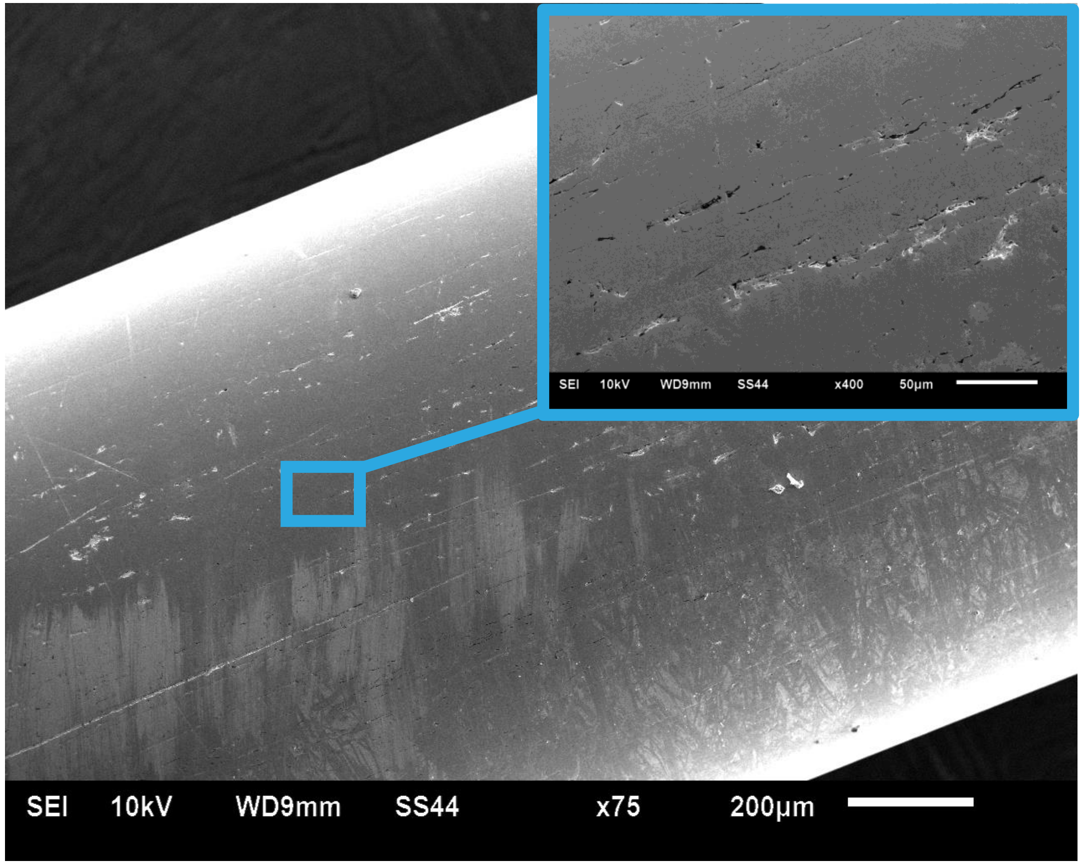
How will the interfacial corrosion mechanism between the metal and salt be evaluated?

- Static corrosion studies of Ni-20Cr (model system) and Hastelloy N (commercial alloy) in:
  - **LiCl-KCl**
  - LiCl-KCl-NiI_2
  - LiCl-KCl-LiI
  - LiCl-KCl-KI

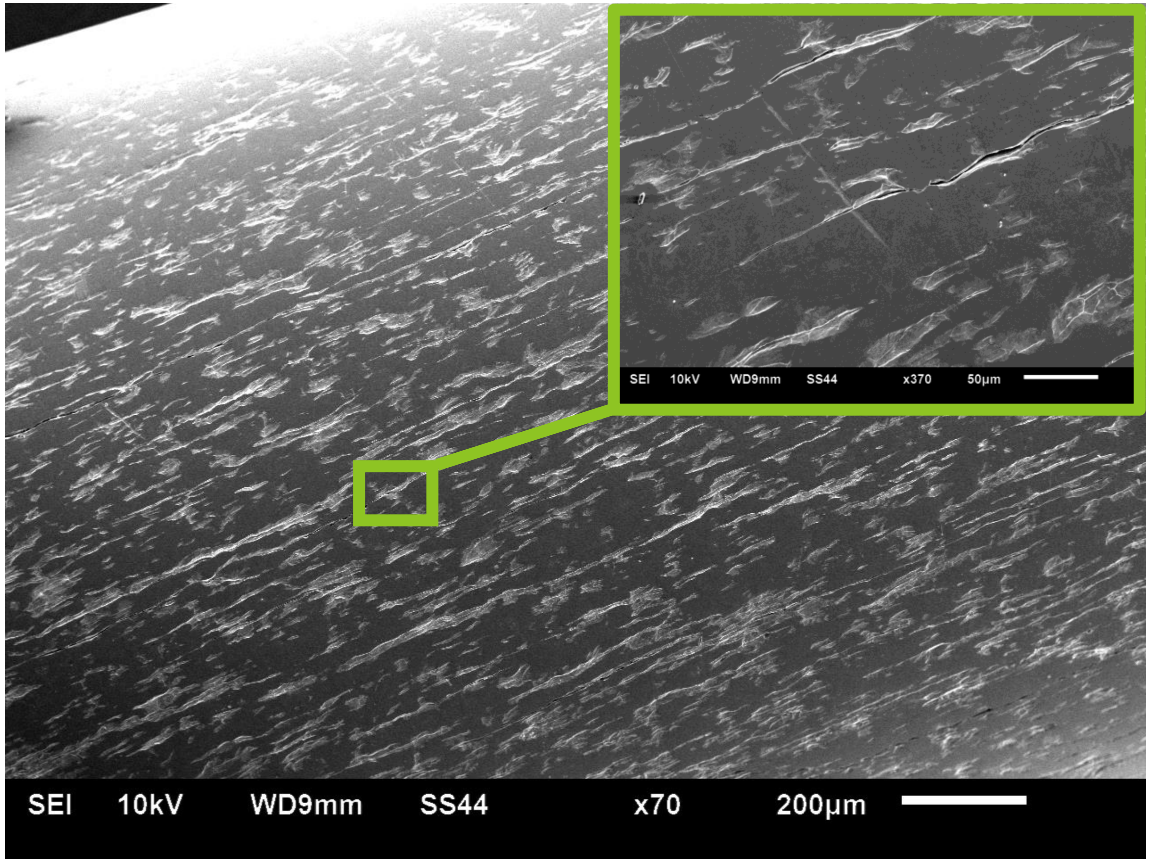
- Salts systems chosen were identical to Research Objective 1

What is the state of as-received specimens before a static corrosion experiment?

As-Received Ni-20Cr wire
(After hand polished to < 1 μm finish)

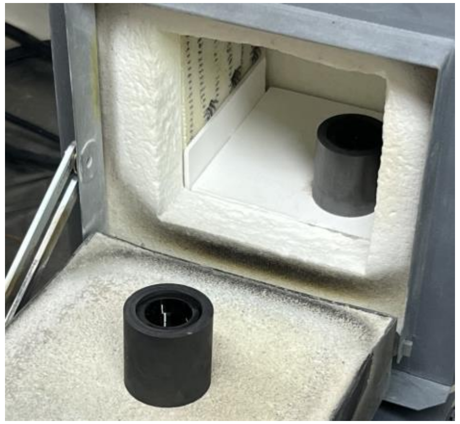


As-Received Hastelloy N wire
(After hand polished to < 1 μm finish)

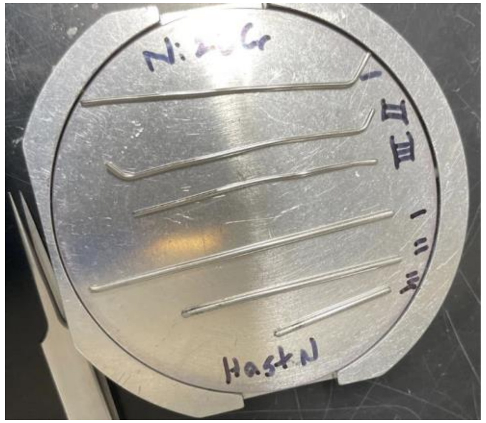


How are specimens prepared for static corrosion studies?

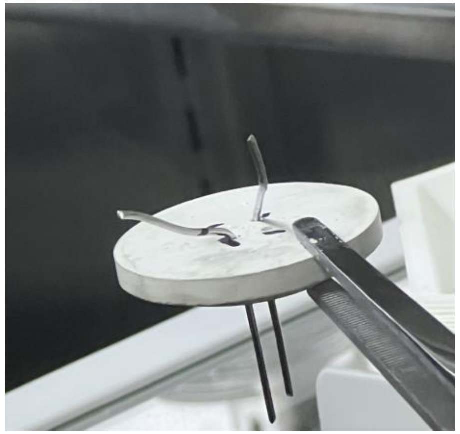
LiCl-KCl eutectic salt loaded



Melted at 500ºC



Polished wire specimens



Wires secured into Boron Nitride lid

Specimens are setup in crucibles with molten salt, ready for study

Idaho National Laboratory

Battelle Energy Alliance manages INL for the U.S. Department of Energy's Office of Nuclear Energy. INL is the nation's center for nuclear energy research and development, and also performs research in each of DOE's strategic goal areas: energy, national security, science and the environment.