CAES
Center for Advanced Energy Studies

A research partnership between Boise State University, Idaho National Laboratory, Idaho State University and University of Idaho.

Center for Advanced Energy Studies

Fission Product Separation by Cold Finger Crystal Growth

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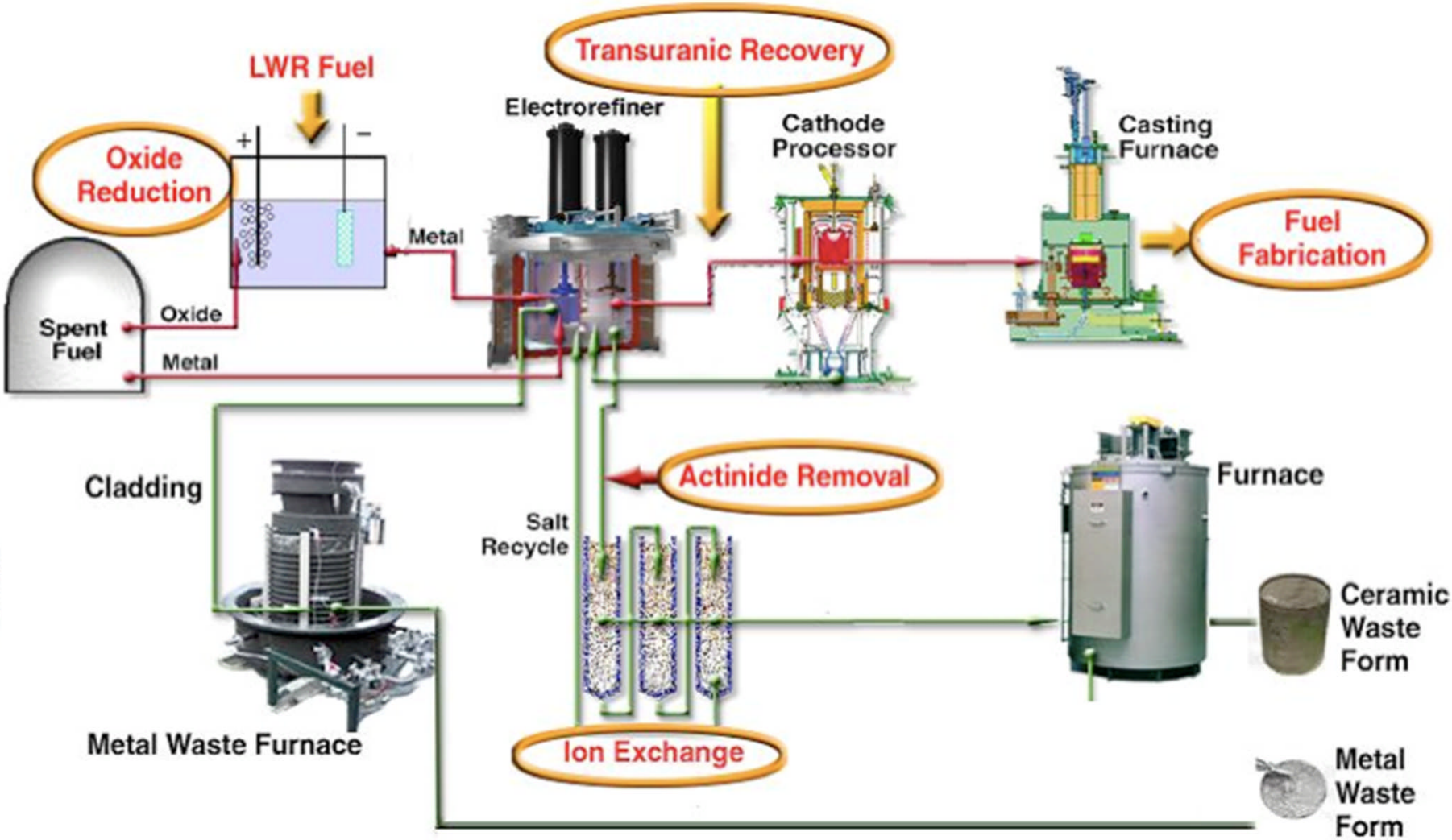
2012 IPRC
Fontana, Wisconsin

Outline

- Background
- Motivation & Goals
- Cold Finger Theory
- Cold Finger Design
- Experimental Program
- Results & Discussion
- Summary & Future Work

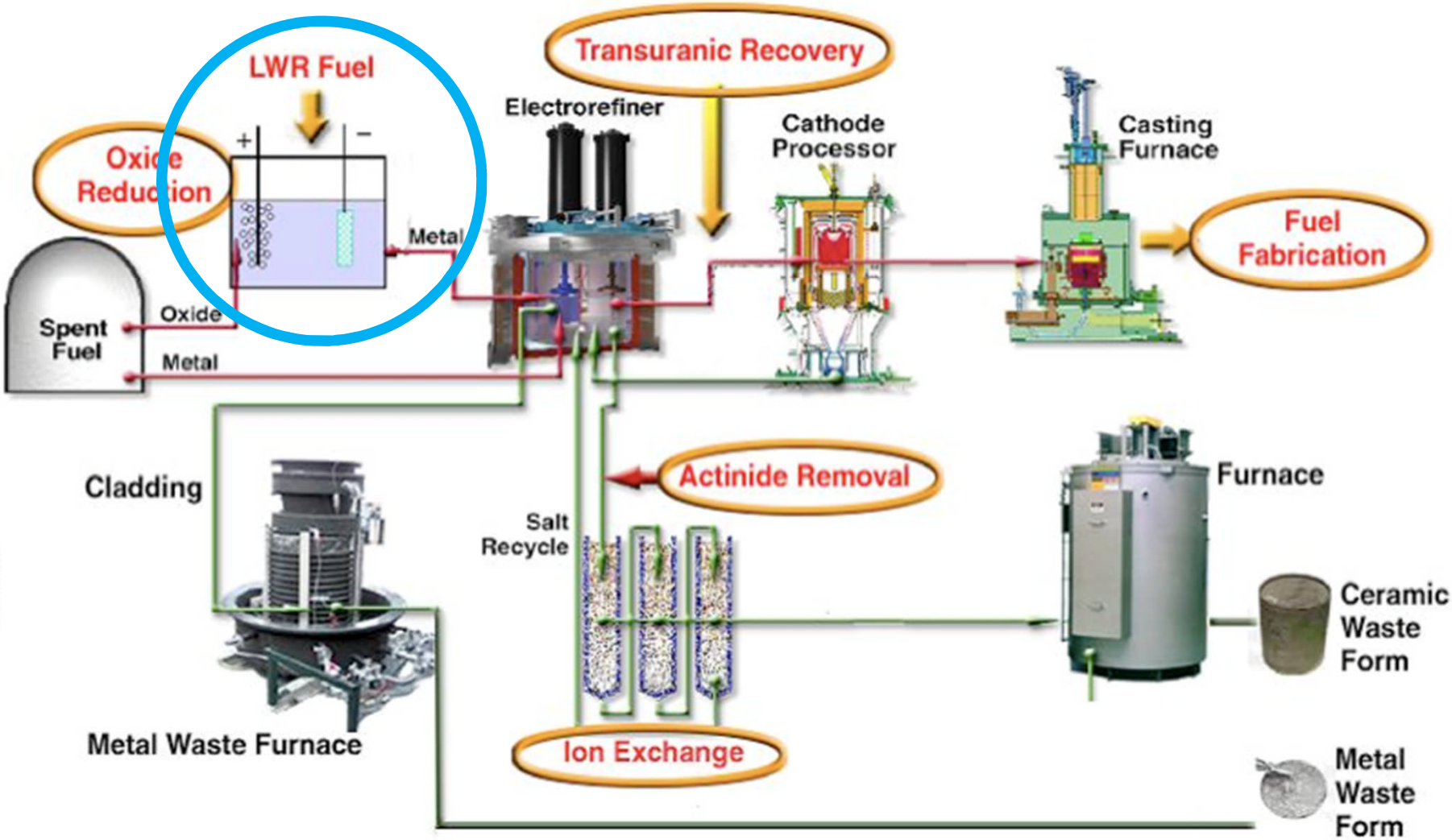
Background

Advanced Pyrochemical Technology Concept



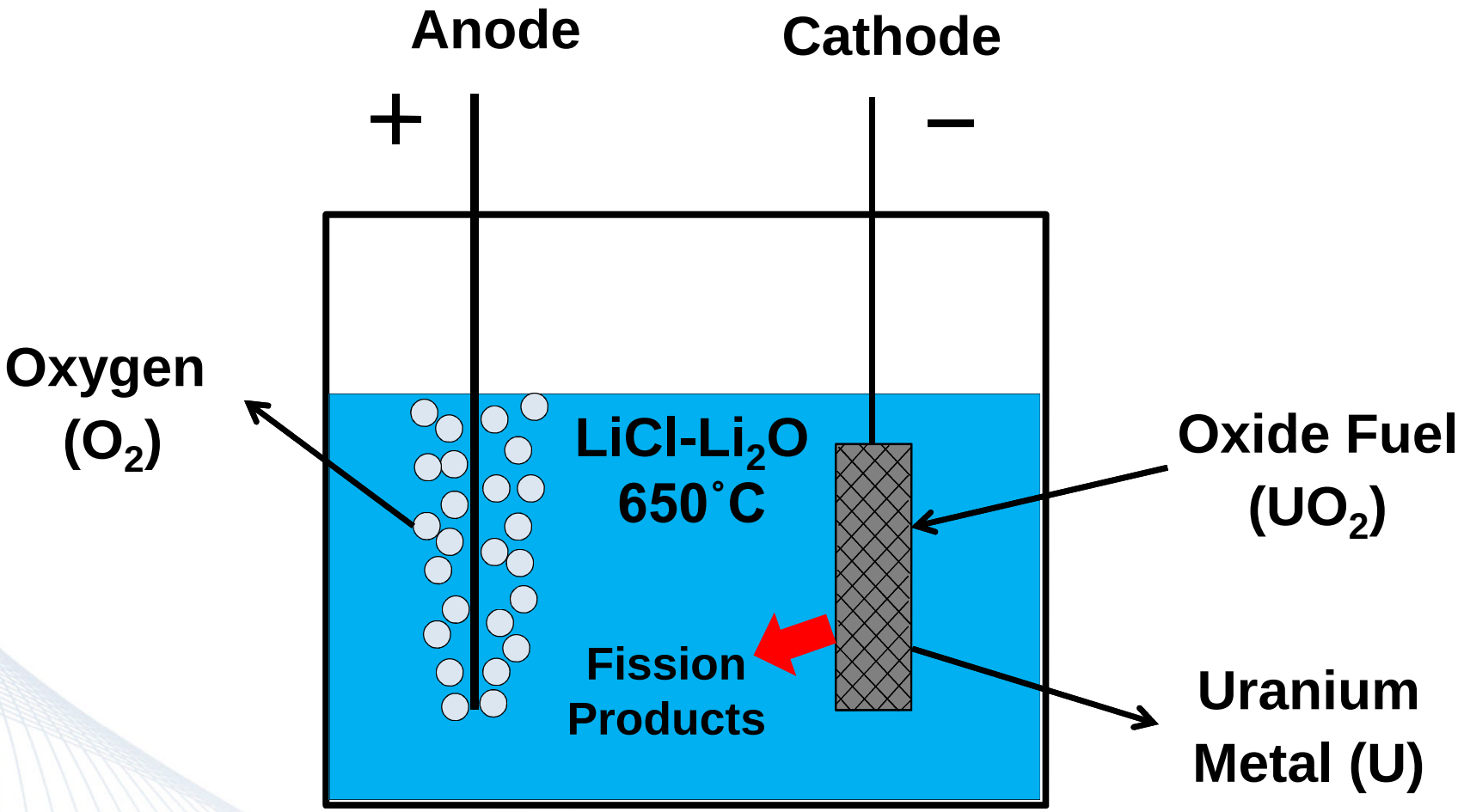
Background

Advanced Pyrochemical Technology Concept



Background

Oxide Reduction Process

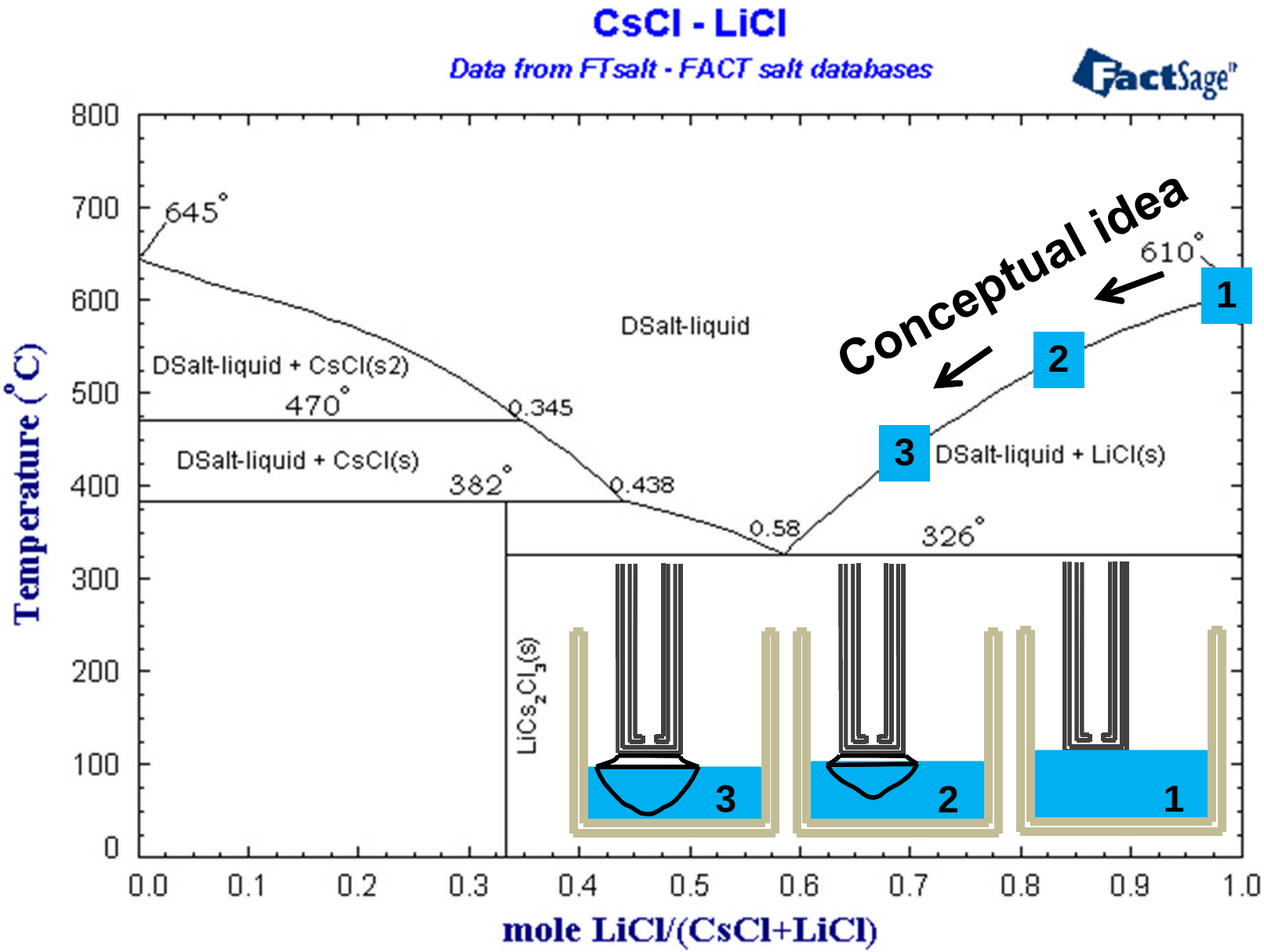


- Fission product build-up effects: electrolyte characteristics and process efficiencies

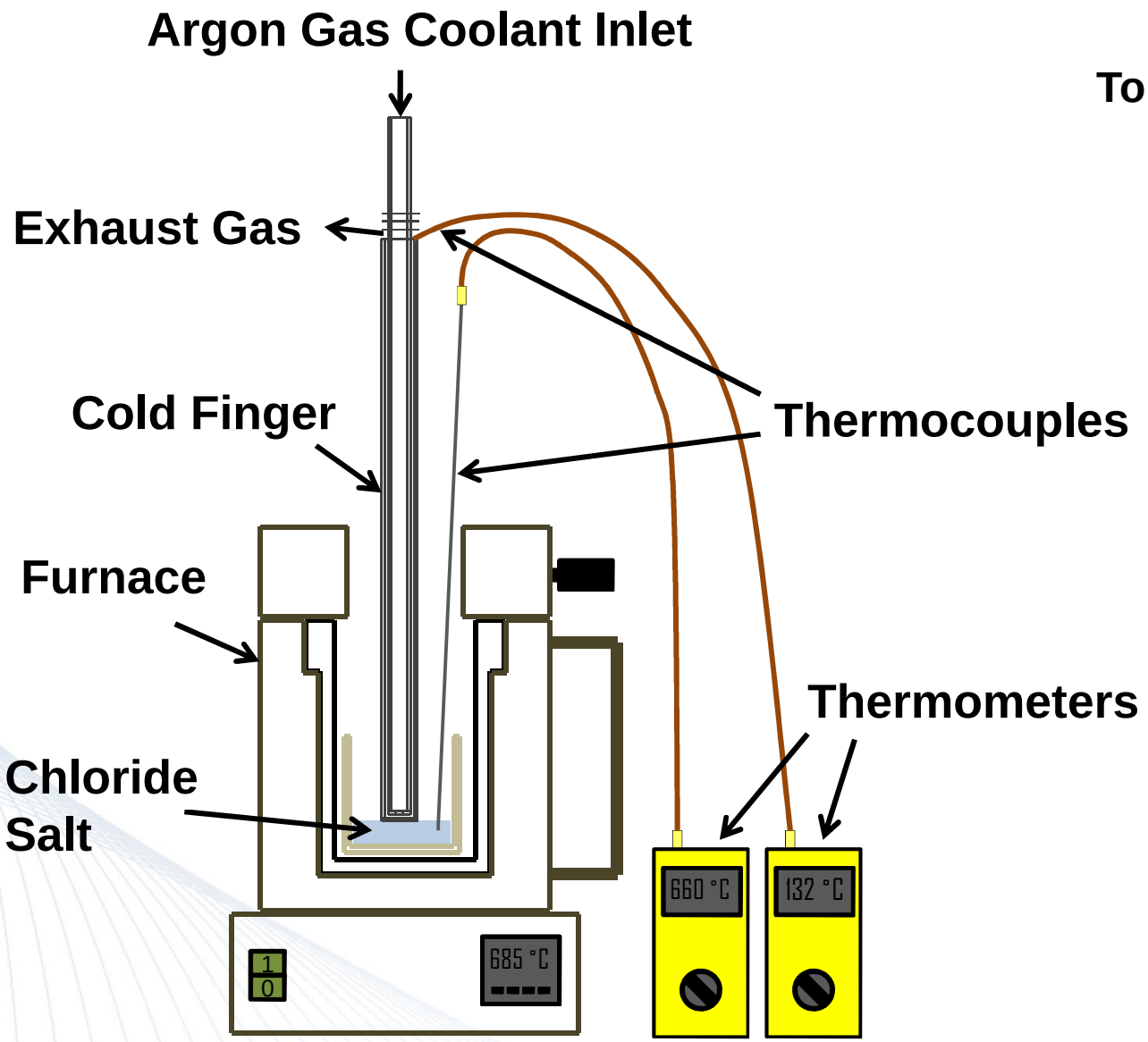
Motivation & Goals

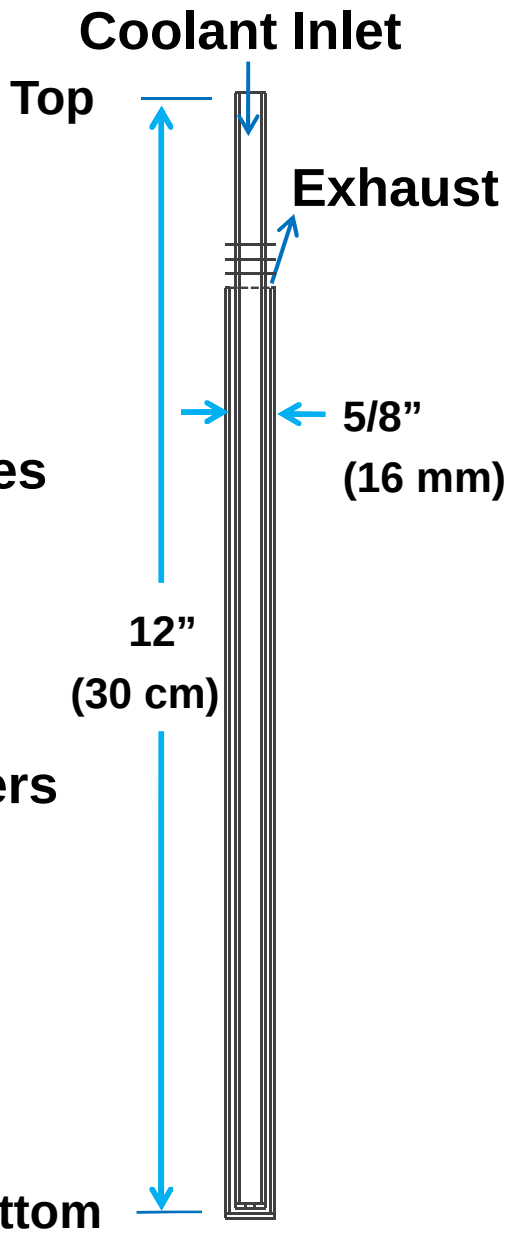
- Methods for cleaning used electrolyte are needed to reduce waste volume and cost
- Melt crystallization techniques have been explored:
  - Zone freezing (KAERI and U of I)
  - Wall freezing (KAERI)
- A cold finger technique has been proposed to study the effect of fundamental parameters on separation
  - Growth time and
  - Cooling rate
- Result will provide information on optimal operating parameters and conditions

Cold Finger Theory

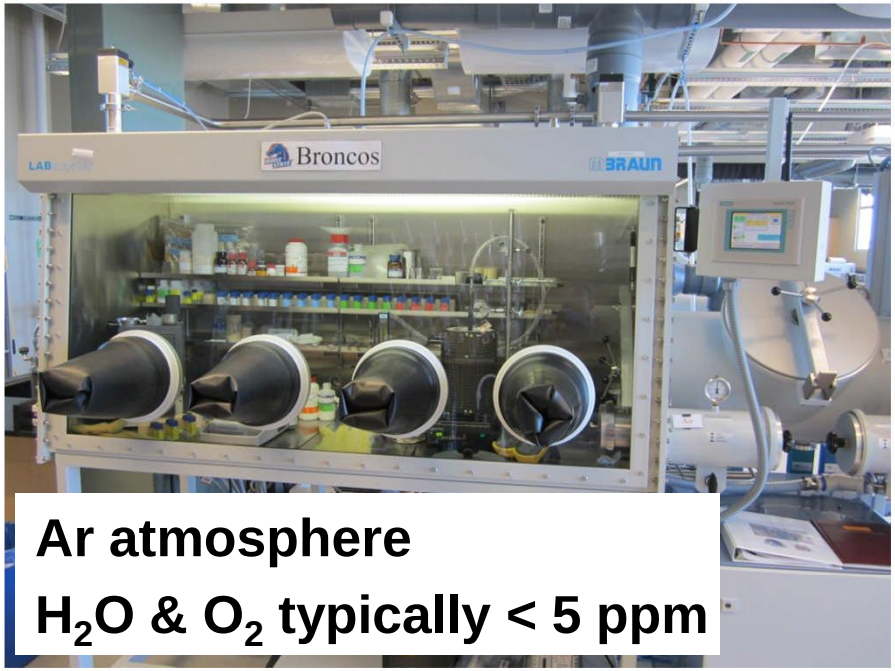


Cold Finger Design





Experimental Setup



Ar atmosphere

H_2O & O_2 typically < 5 ppm

View of salt

Experimental Procedure

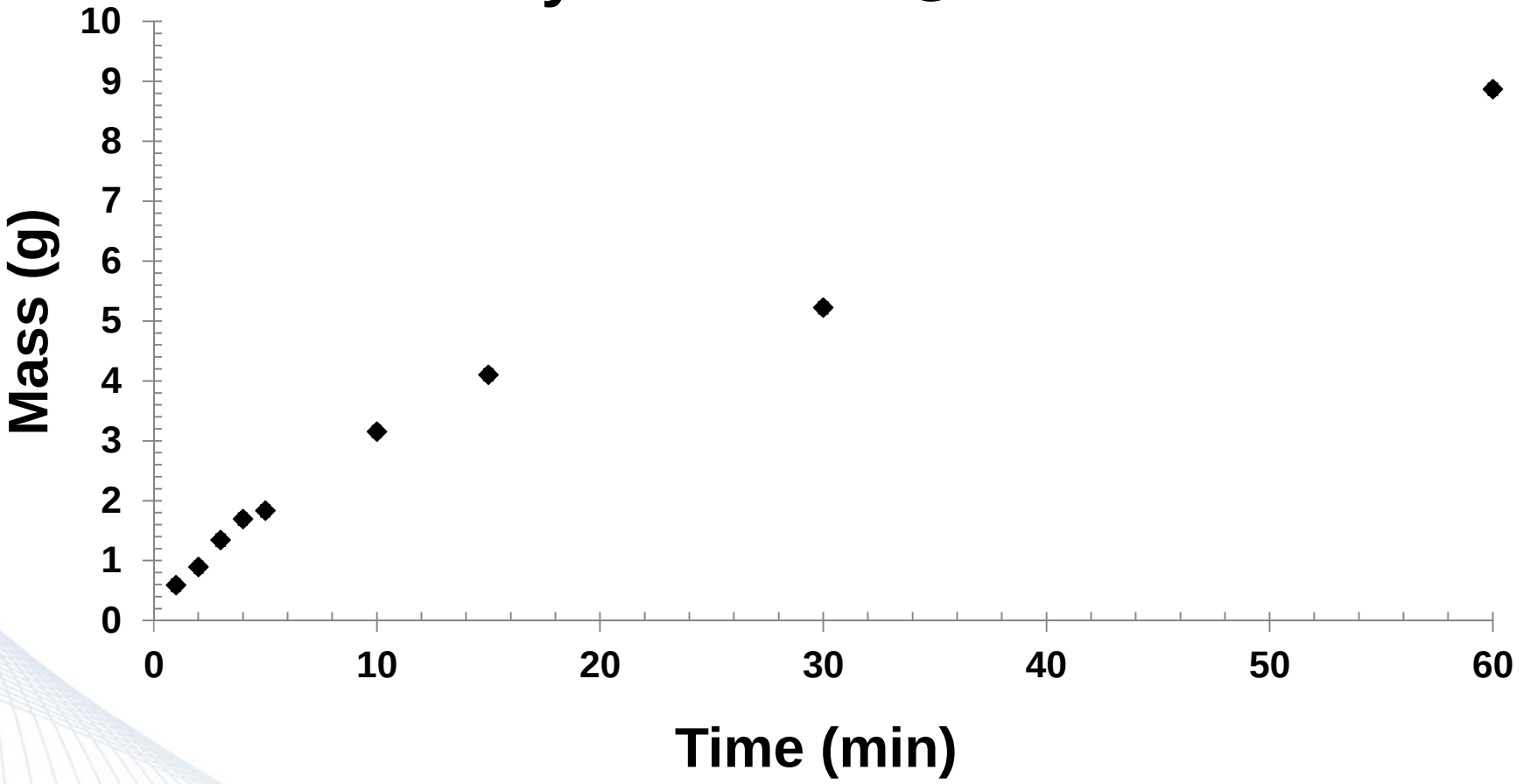
- Loaded 30 grams in MgO crucible
  - LiCl
  - LiCl-CsCl (5 wt% CsCl)
- Melted salt at 685˚C
- Positioned cold finger at the molten salt top surface
- Experiments began once salt temperature reached 660˚C (± 3˚C)
- Varied coolant gas flow rate (Ar) and duration
  - 7.4 to 14.9 L/min
  - 5 to 30 min

Note: 1 L/min = 1.667×10^{-5} m^3/s

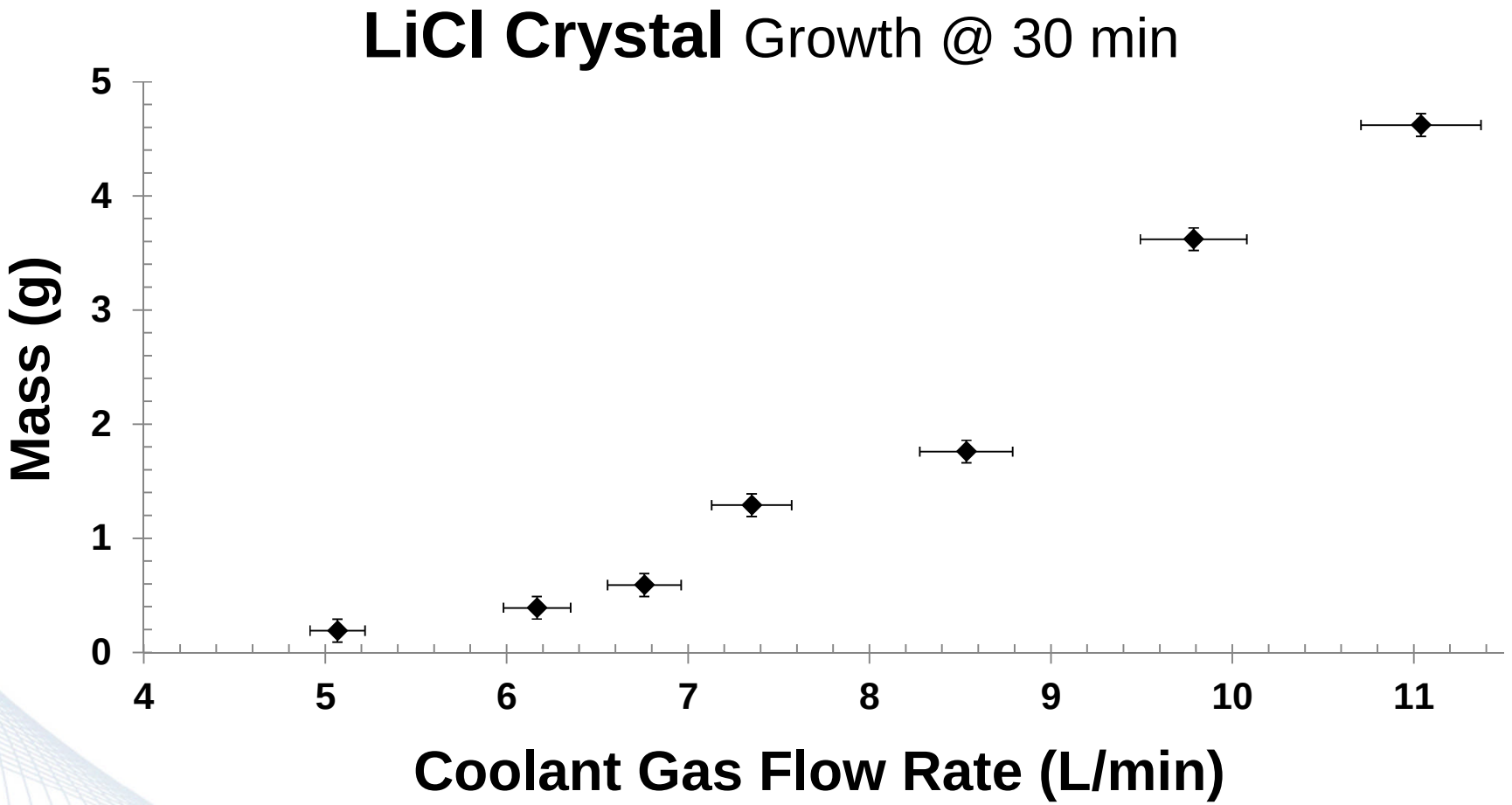
Sample Preparation/Analysis

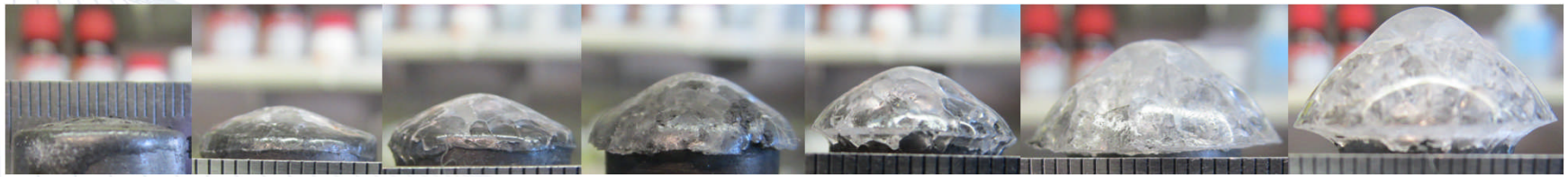
- Crystal
  - Weighed
  - Dimensionally Measured
  - Crushed
  - Homogenized
- Analyzed by ICP-MS
  - Li & Cs concentrations

Duration Effect



Coolant Rate Effect

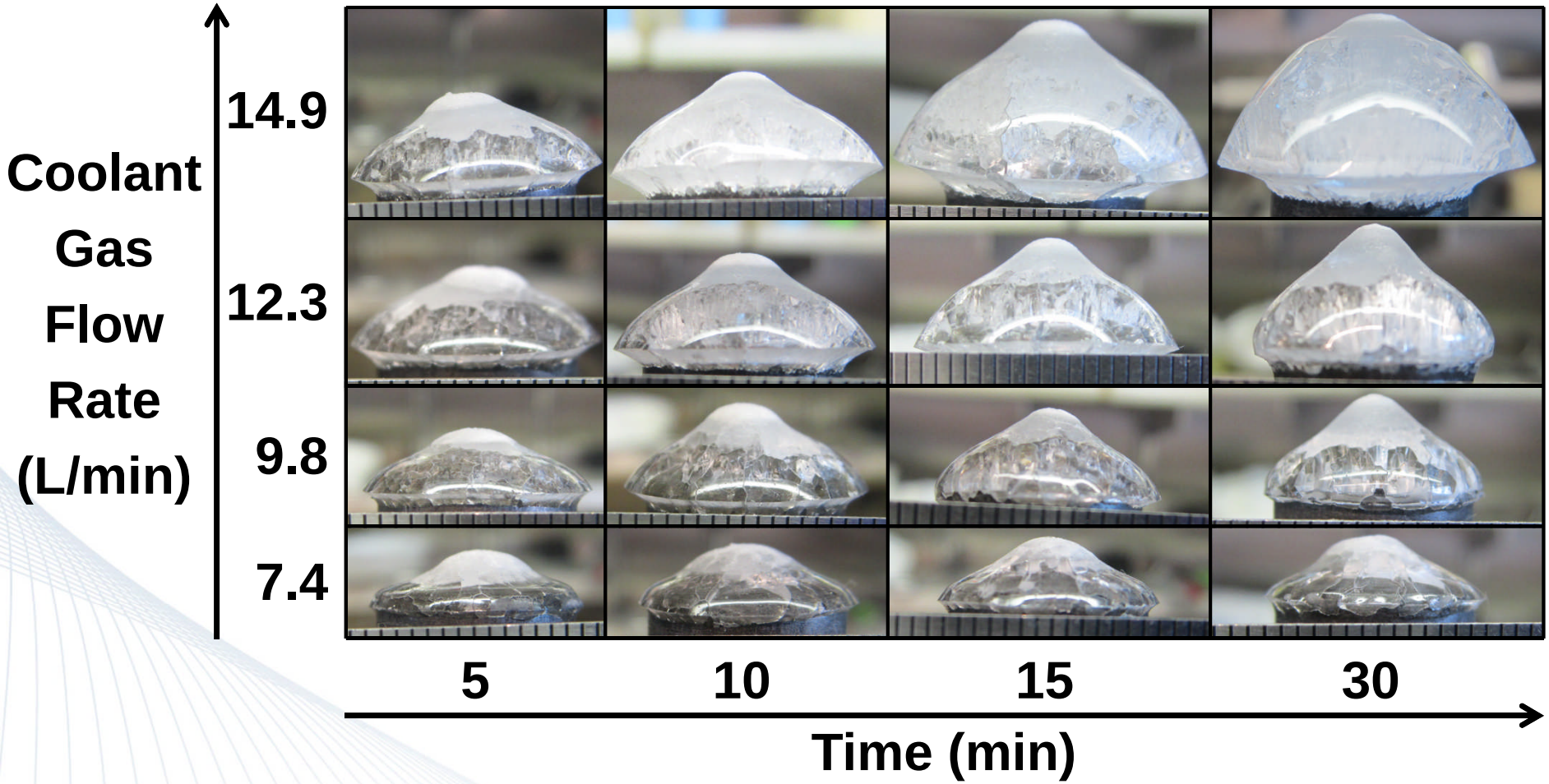




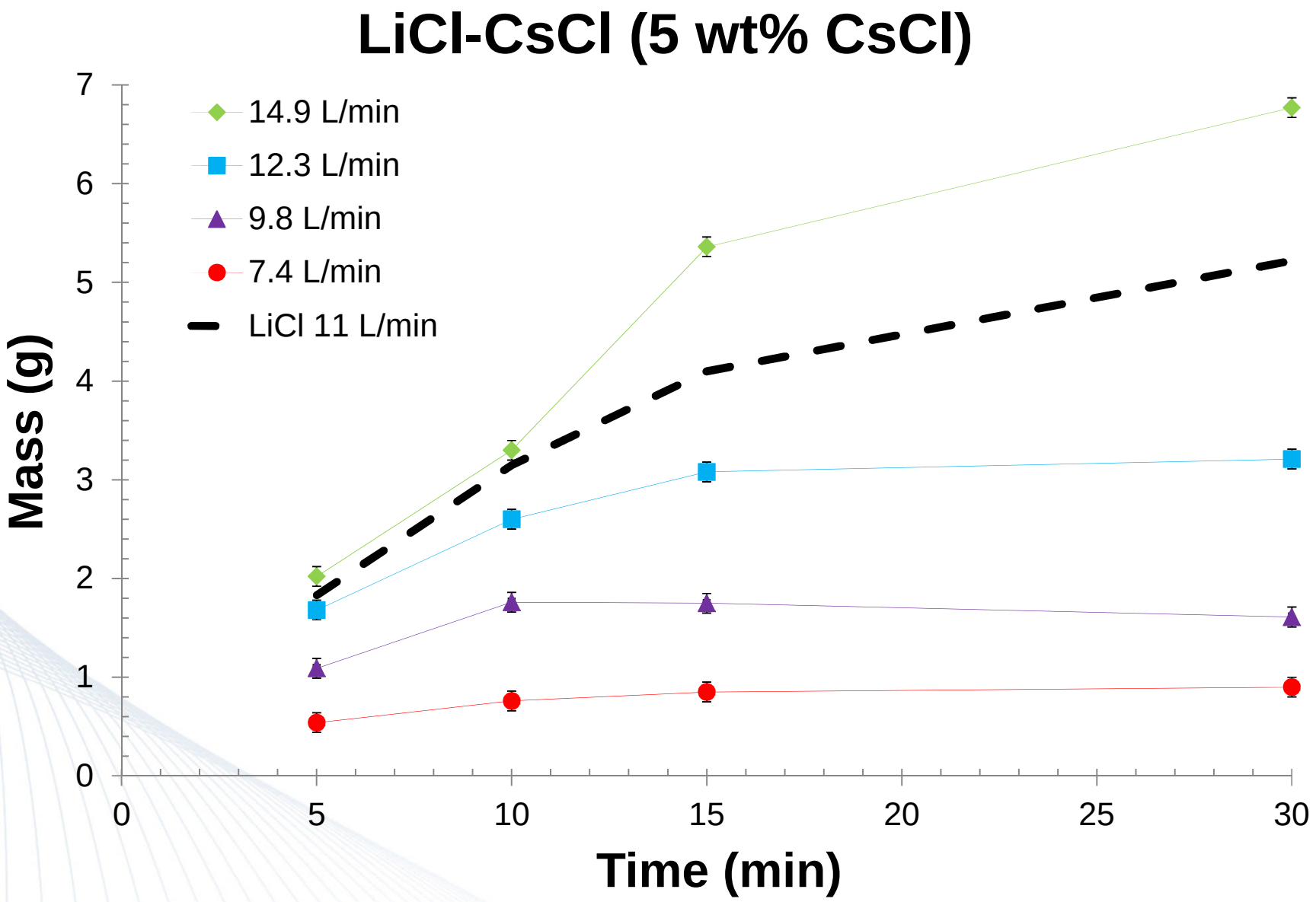
5.1 L/min | 6.2 L/min | 6.8 L/min | 7.4 L/min | 8.5 L/min | 9.8 L/min | 11 L/min

Crystal Formation Matrix

LiCl-CsCl (5 wt% CsCl)



Crystal Growth



wt% CsCl Data Analysis

LiCl-CsCl Crystals

Expected maximum

wt% CsCl

Coolant Gas Flow Rate (L/min)	5	10	15	30
14.9	0.527 ↑	0.364	0.330	0.278
12.3	0.580	0.382	0.277	0.303
9.8	0.434	0.356	0.251	0.303
7.4	1.12	0.578	0.438	0.600 ↓

Expected minimum

Time (min)

Note: LiCl-CsCl initially @ 5 wt% CsCl

LiCl vs. LiCl-CsCl Crystals

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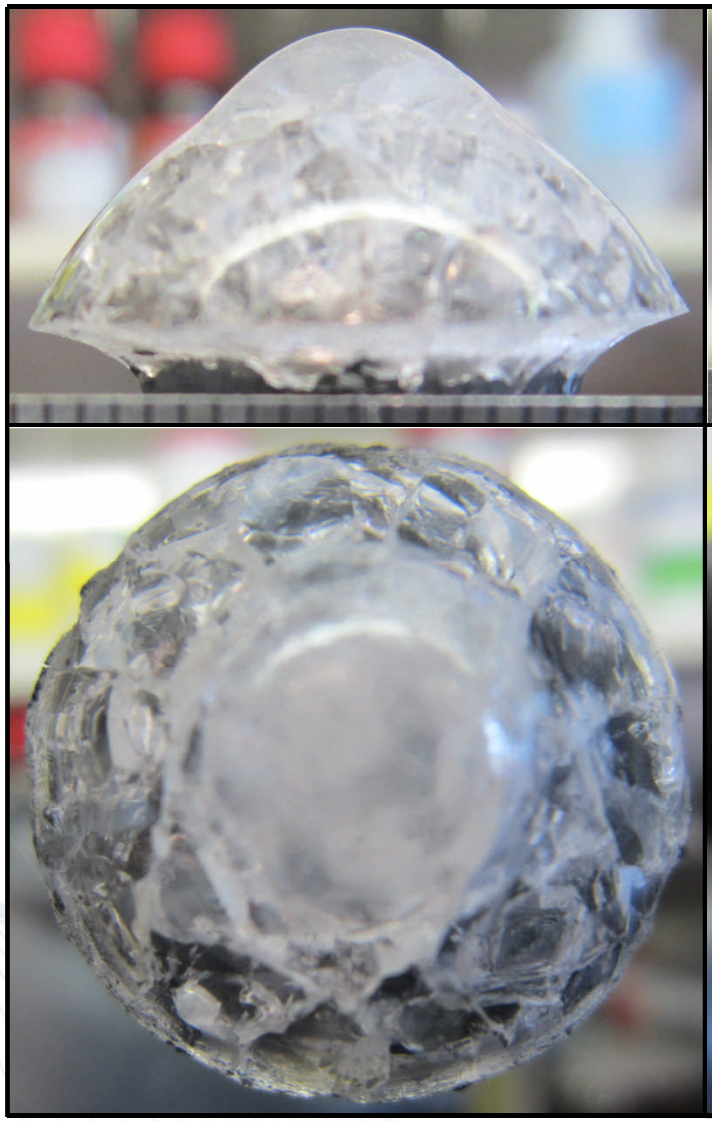
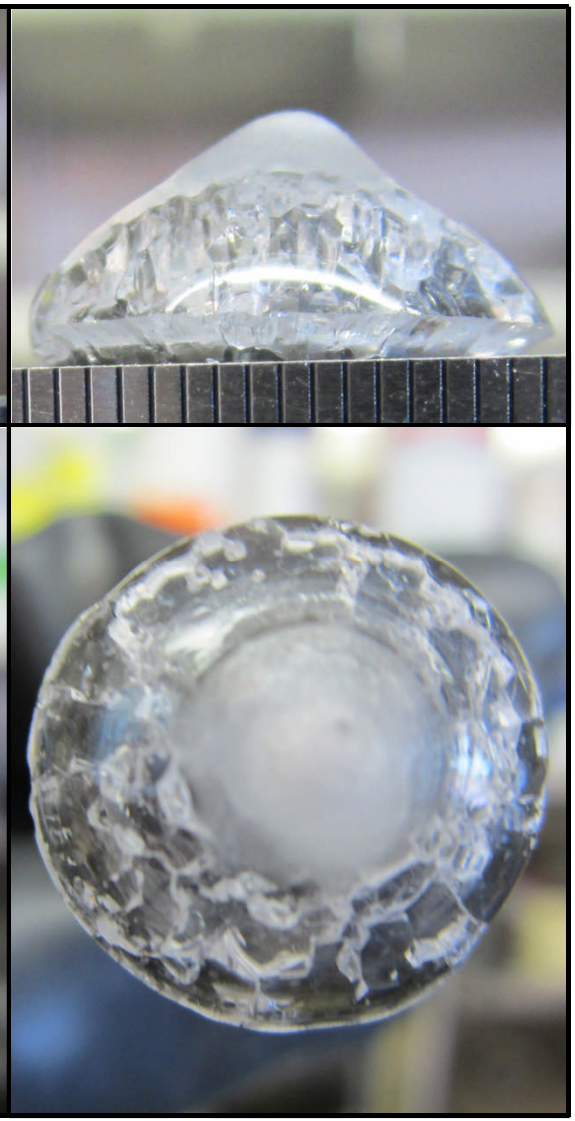
	LiCl	LiCl-CsCl	
11 L/min 30 min 4.6 g			11 L/min 30 min 2.3 g

Note: Crystals scaled relative to each other

LiCl vs. LiCl-CsCl Crystals

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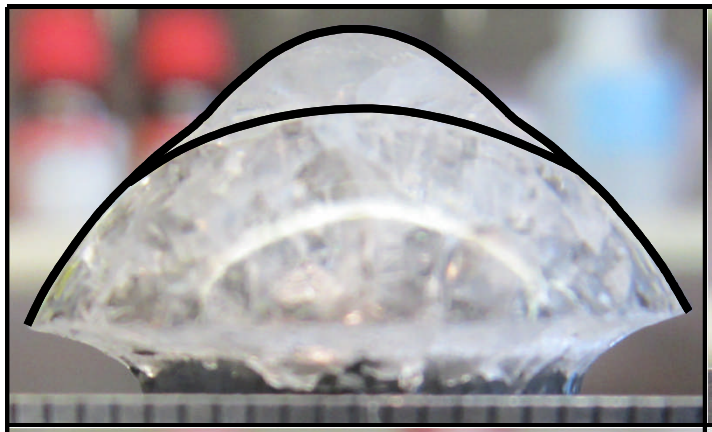
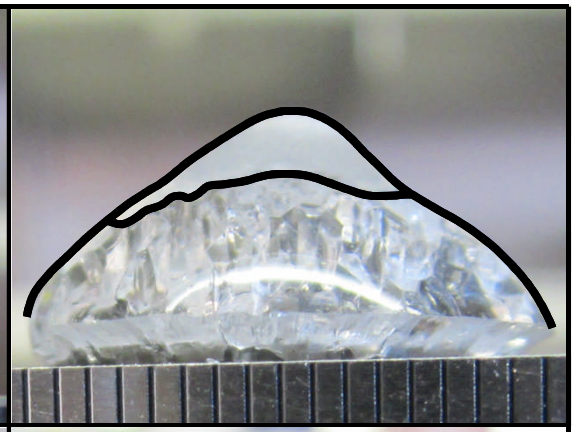
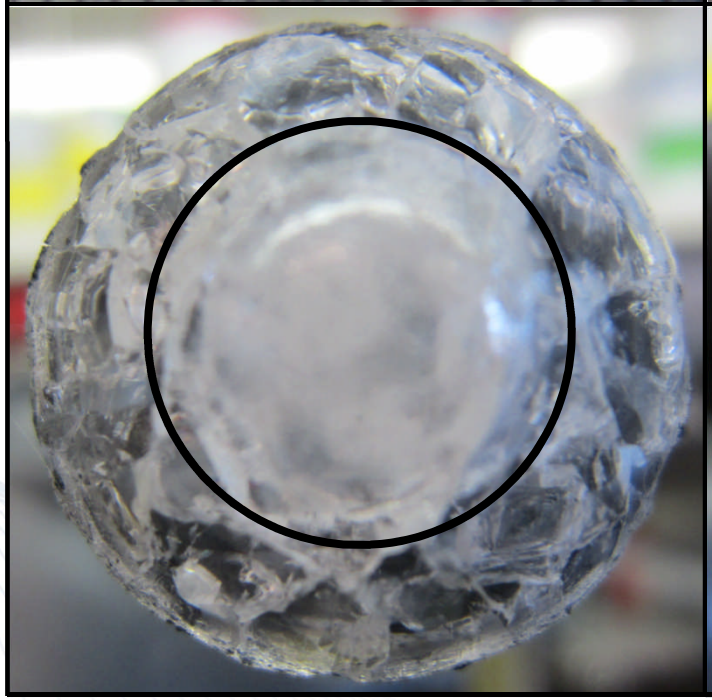
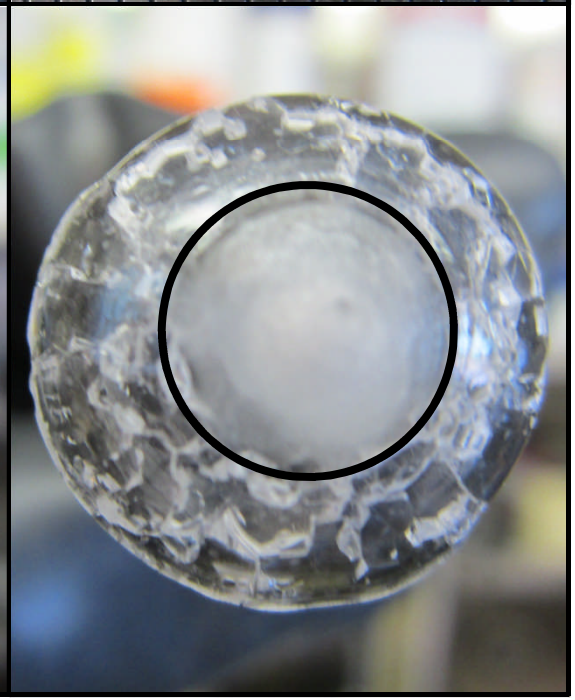
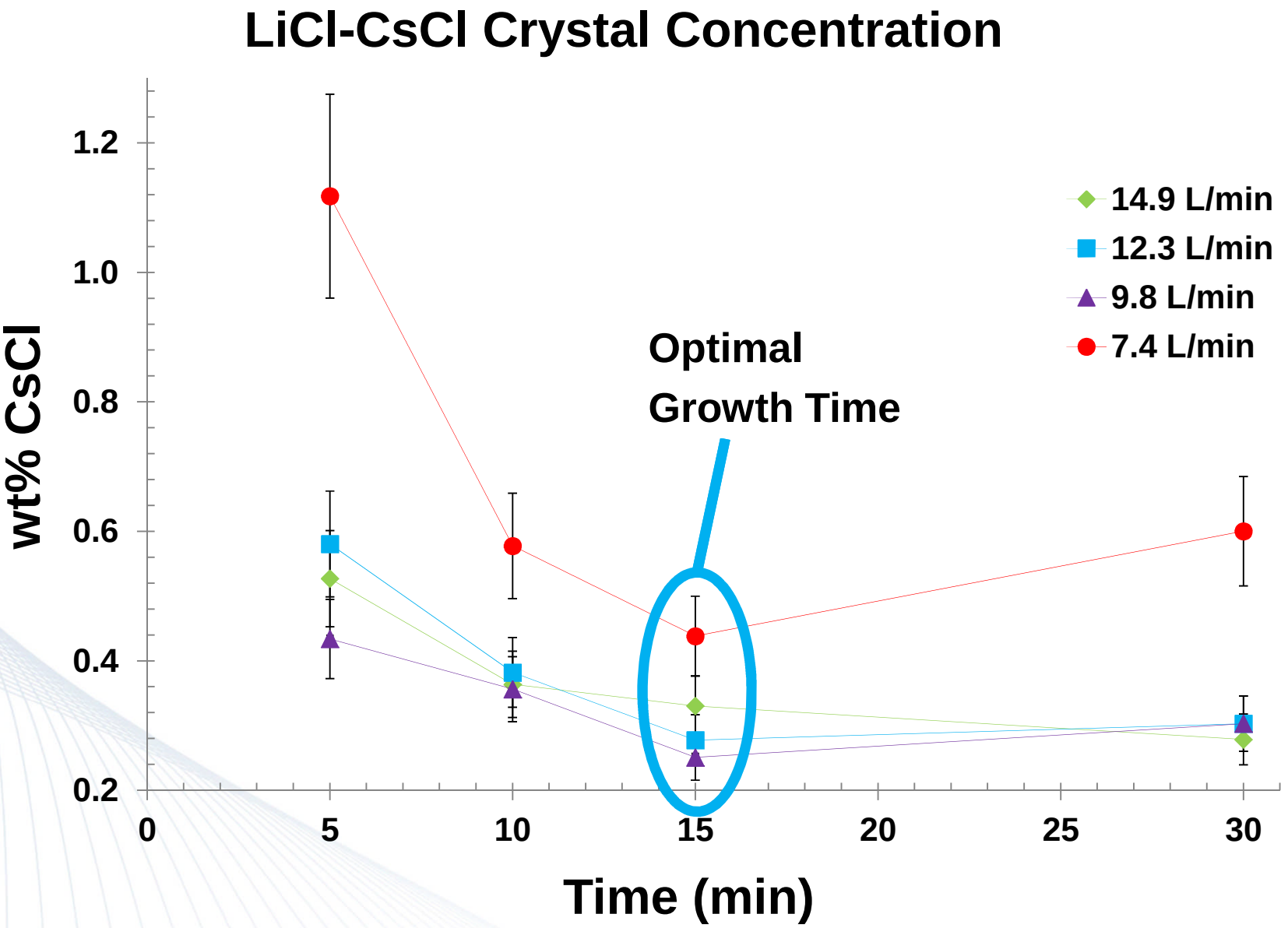
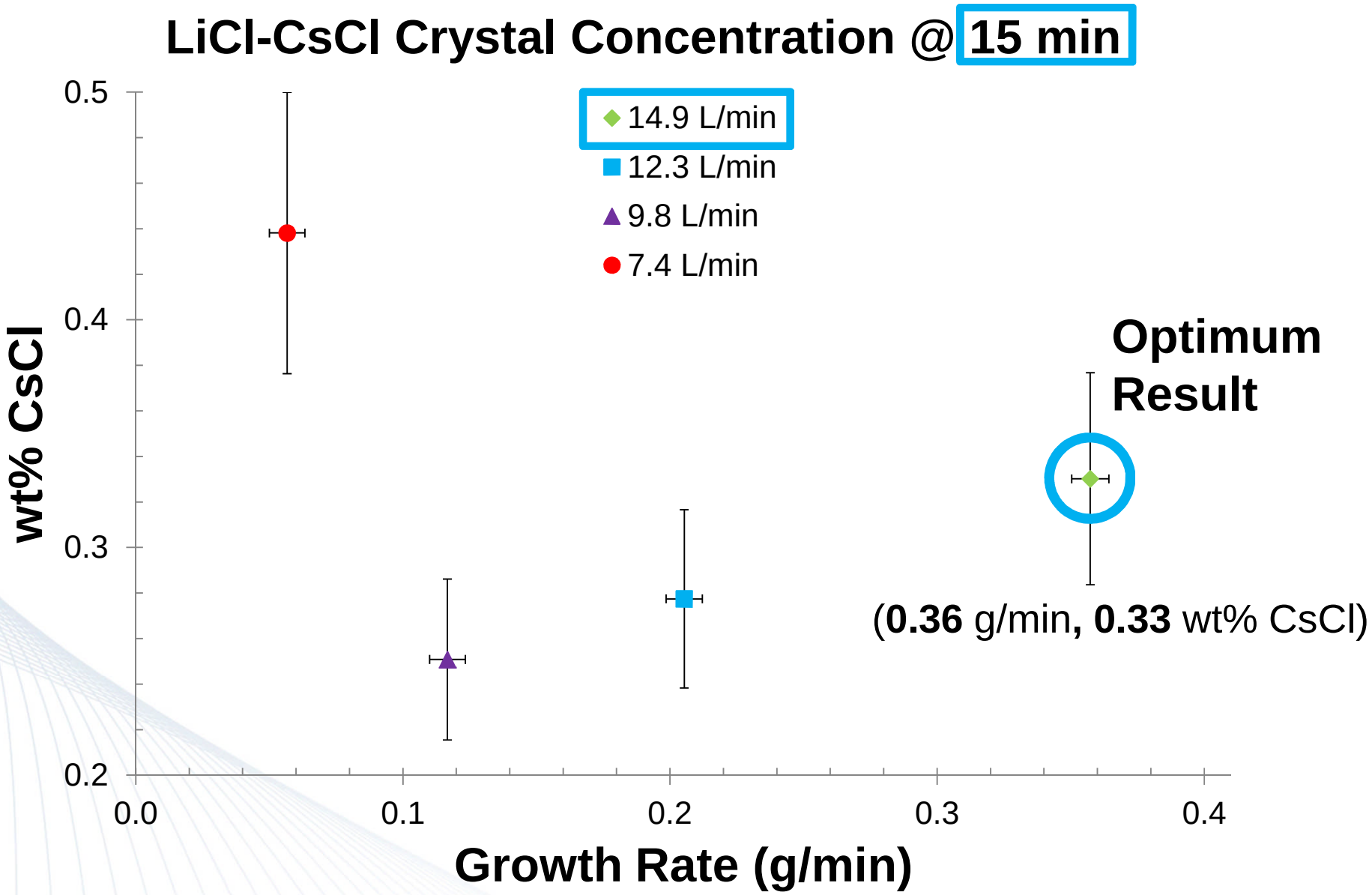
	LiCl	LiCl-CsCl
11 L/min 30 min 4.6 g		
11 L/min 30 min 2.3 g		

Note: Crystals scaled relative to each other

Optimal Duration



wt% CsCl vs. Growth Rate

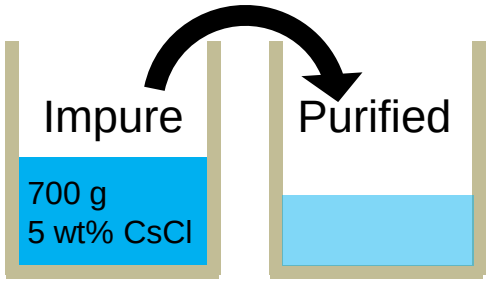


Purposed Separation

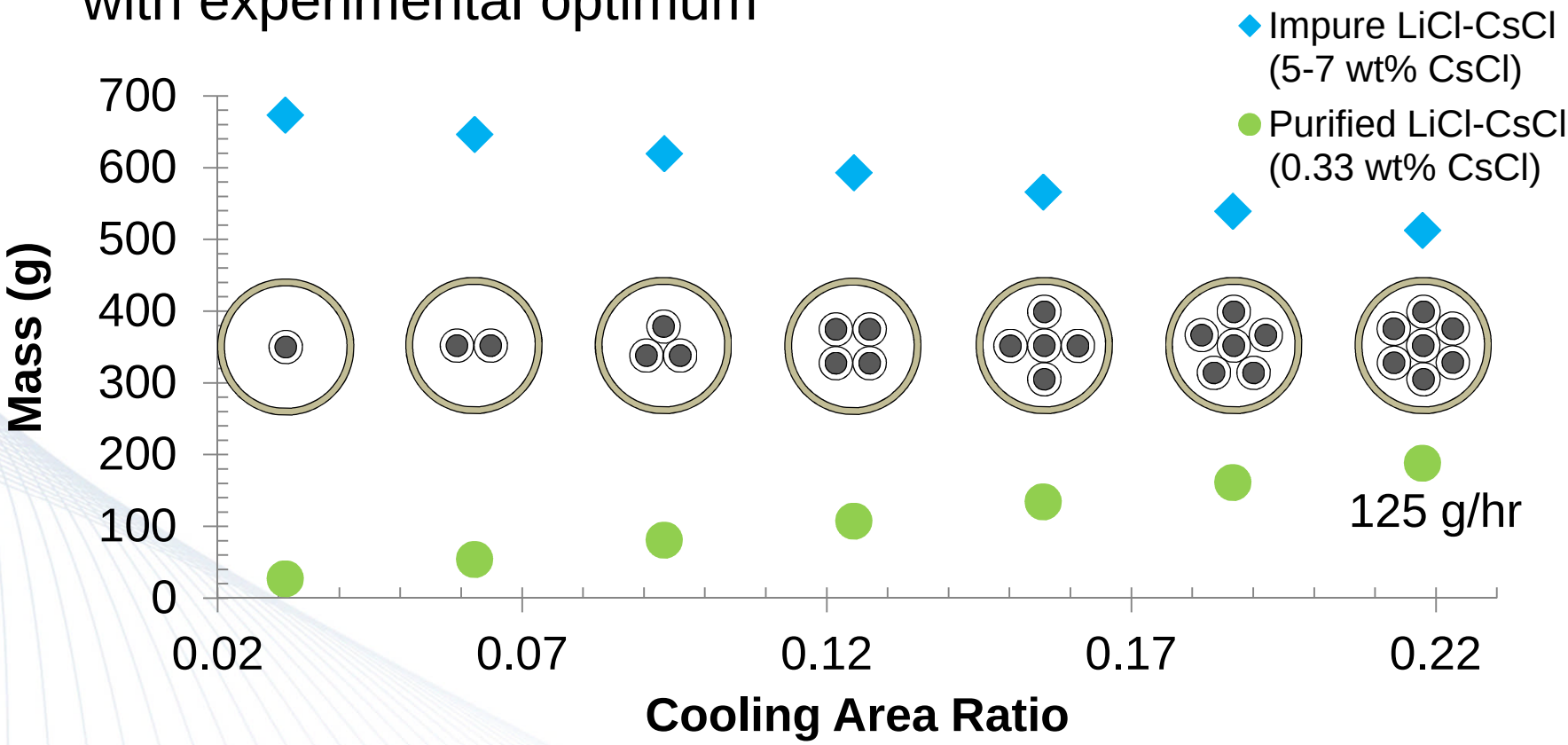
Potential Separation Rates

Given:

5 crystal growth cycles
in 90 minutes
with experimental optimum



MgO Crucible:
10 cm Diameter
11 cm Height



Separation Science and Technology, 41: 1965-1983, 2006

Summary

Optimization Goal

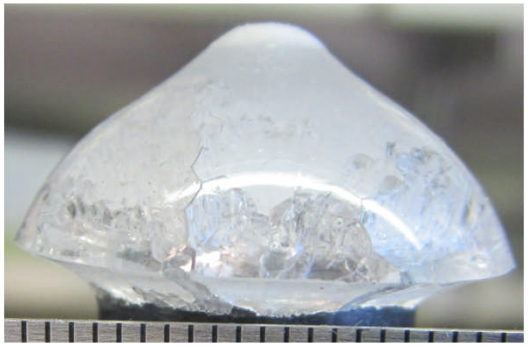
- Minimize CsCl concentration
- Maximize crystal growth rate

Optimal Parameters

- 14.9 L/min (Coolant flow rate)
- 15 min (Growth time)

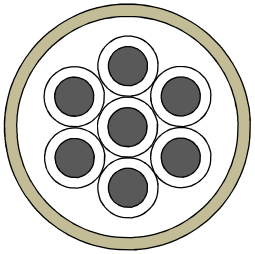
Optimal Results

- 0.36 g/min (Growth rate)
- 0.33 wt% CsCl (Crystal purity)



Potential Separation

- 5 Cycles @ 90 min with 7 fingers
- 125 g/hr (Recycled electrolyte)



Next Step …

Further optimize and fundamentally understand the separation process by:

- Exploring different initial CsCl concentration
- Develop & applying models
  - Heat transfer
  - Crystal growth
- Minimize drip formation
- Experiment with other elements such as strontium (Sr) or neodymium (Nd)

Acknowledgements

- Idaho National Laboratory's (INL) Laboratory Directed Research and Development (LDRD) program for funding
- Center for Advanced Energy Studies (CAES) for support of the LDRD funding and providing laboratory space (Radiochemistry Lab) for this project
- Michael Shaltry, Ammon Williams, Robert Hoover, and Michael Pack
- Debbie Lacroix for ICP-MS analysis