INL/CON-24-76277-Revision-0

# A Molten Salt Aerosol Spectroscopy Approach to Online Process Monitoring in Molten Salt Reactors

July 2024

Changing the World's Energy Future

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**July 2024**

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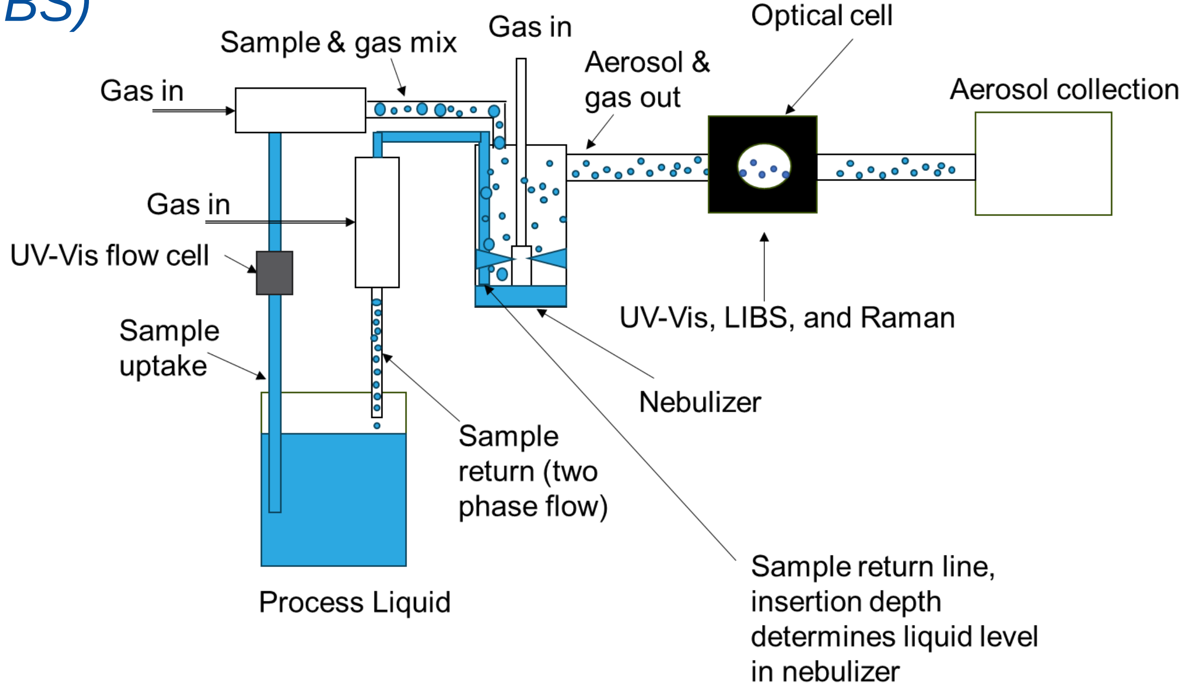
# Introduction and Approach

- *Motivation is to provide a robust in-situ process monitoring technology to monitor actinides in MSRs for safeguards*
- *Goal is to develop a sampling loop combining spectroscopy techniques in-situ and integrate signals using **chemometrics** for MSR salt quantification and tracking*
  - *Laser-Induced Breakdown Spectroscopy (LIBS)*
  - *UV-VIS*
  - *Laser Induced Fluorescence (LIF)*
  - *Raman*
- *Objective for this phase of the work:*
  - *Demonstrate sampling loop and nebulizer*
  - *Perform LIBS, UV-VIS in situ at INL*
  - *Setup LIF at UM*

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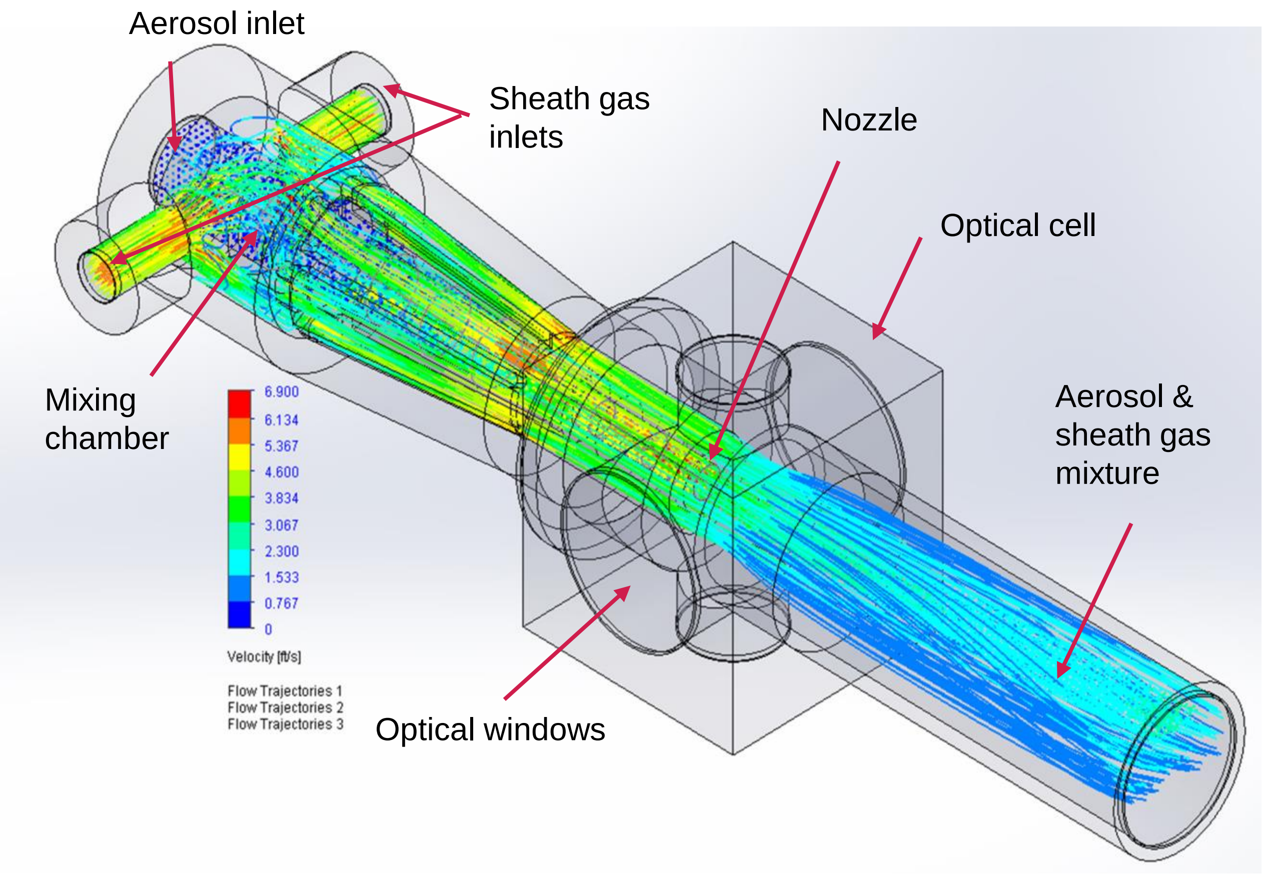
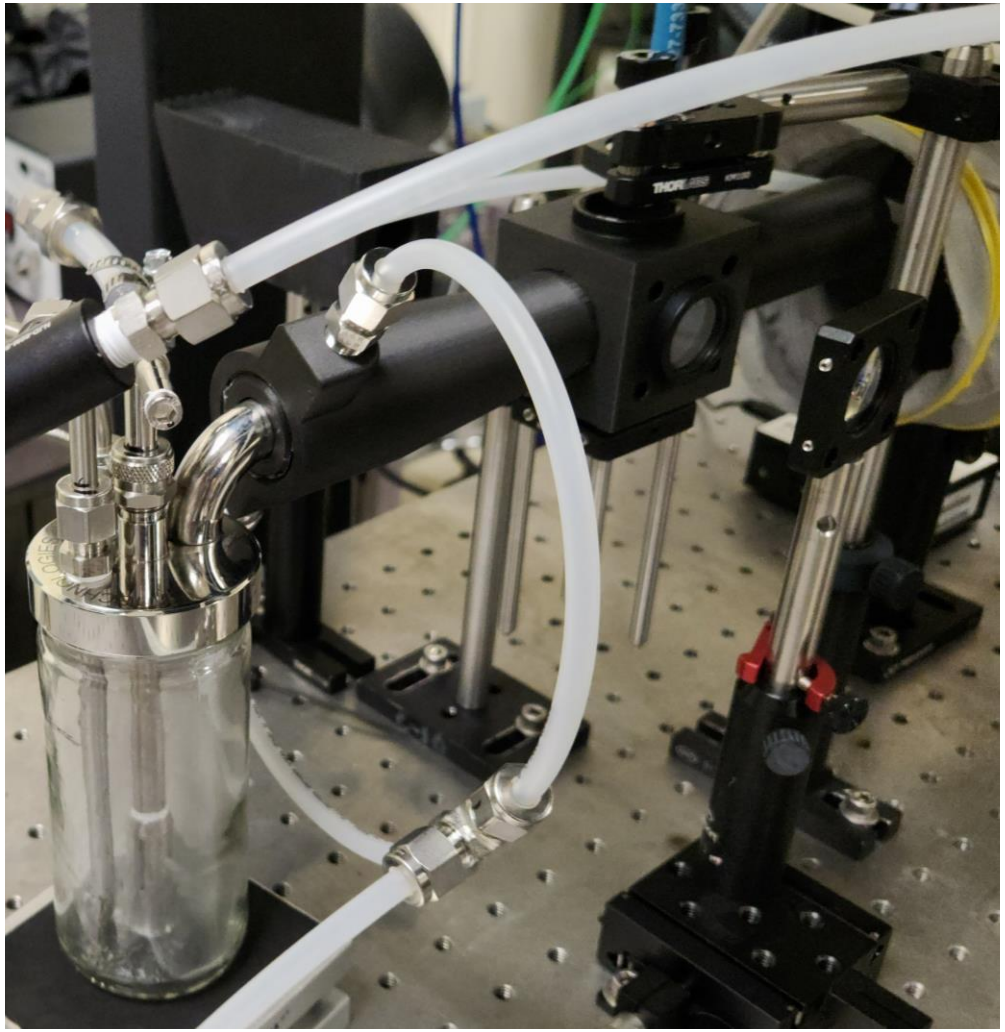
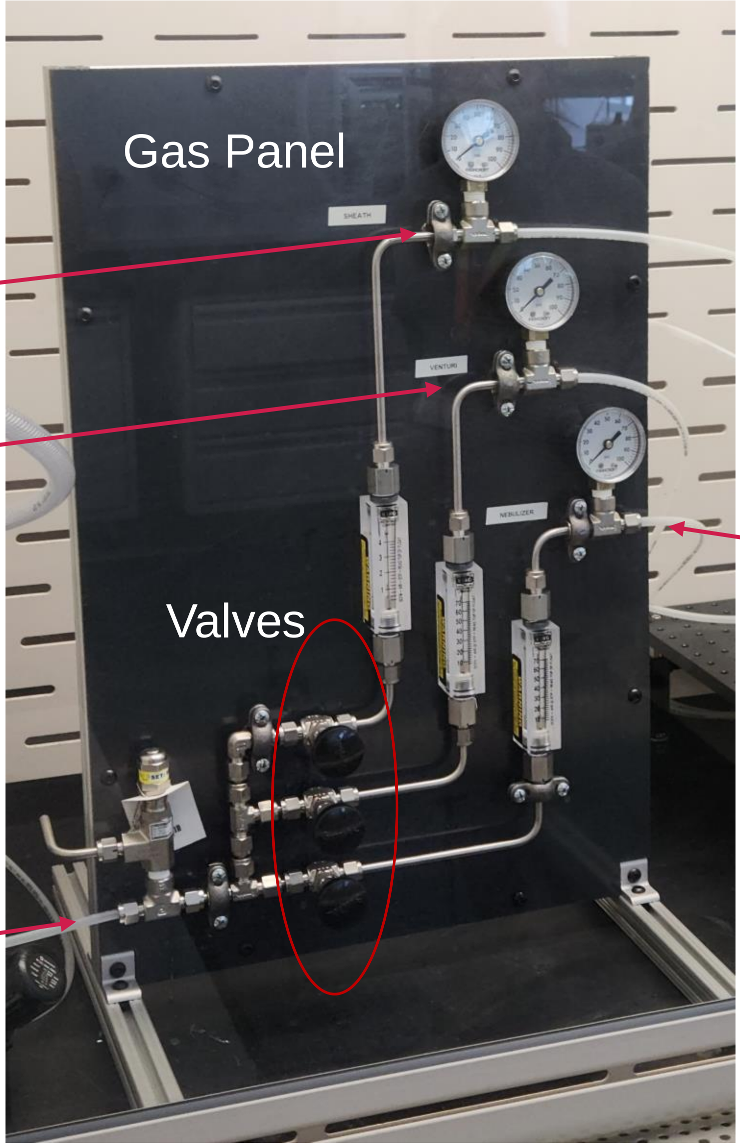
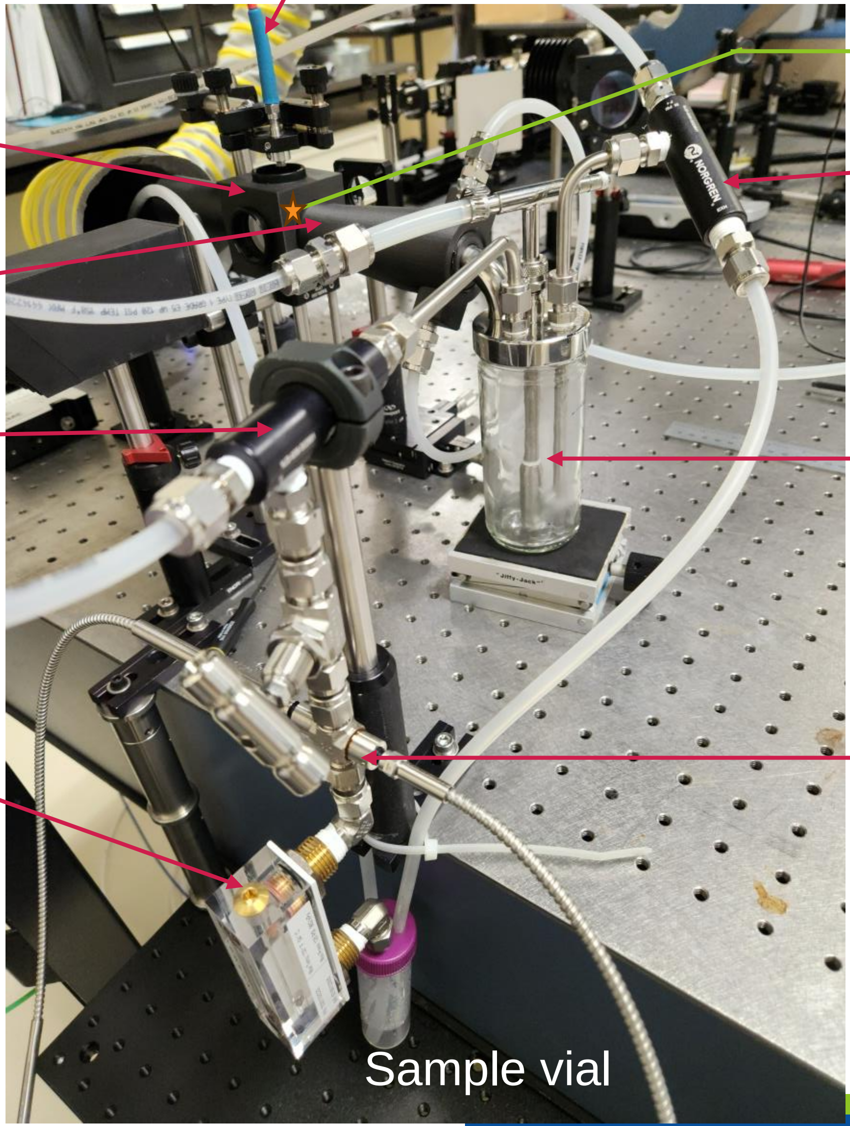
# Aerosol Nozzle and Optical Cell Design



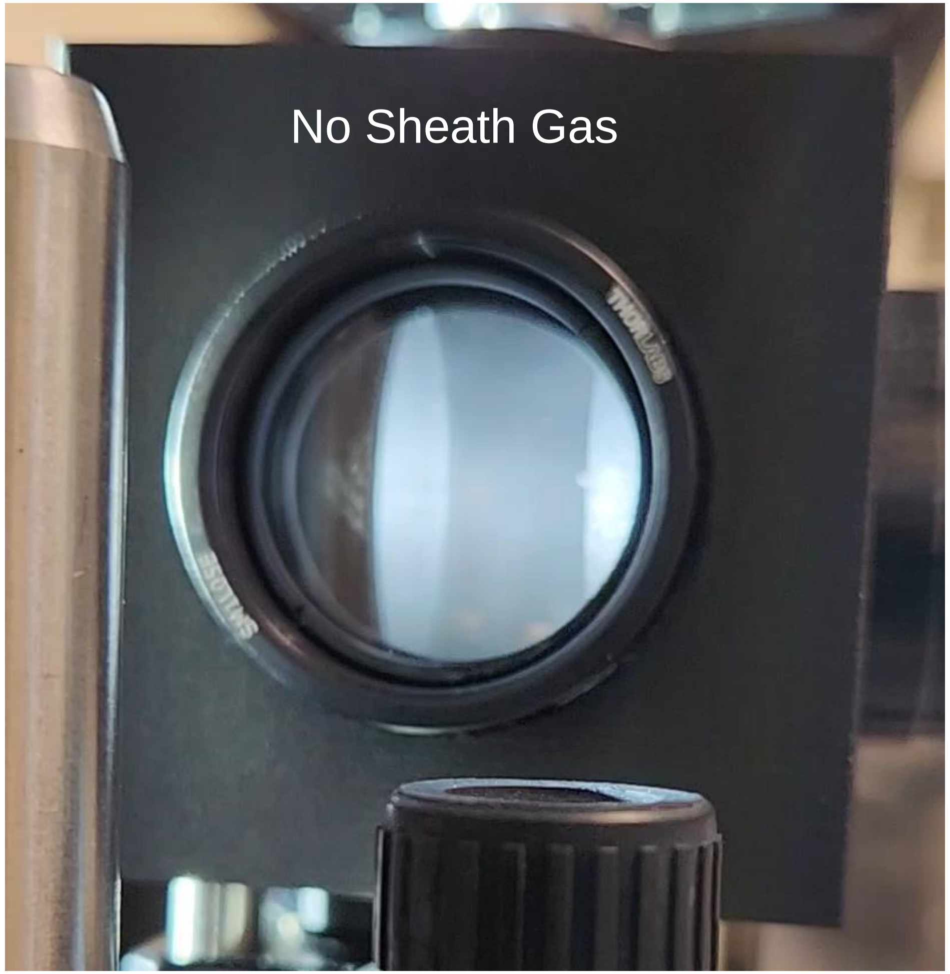



# INL Experimental Setup LIBS/UV-VIS



# Videos

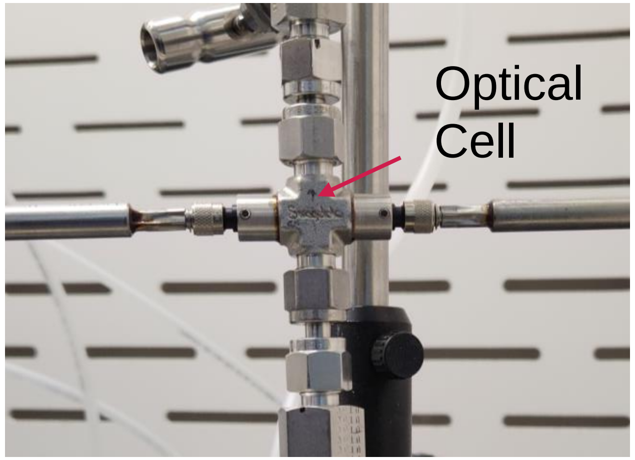

No Sheath Gas
THORLABS
SM1L05E


Sheath Gas @ 1 CFM
THORLABS
SM1L05E

# Initial Experiments: Conditions

## UV-VIS

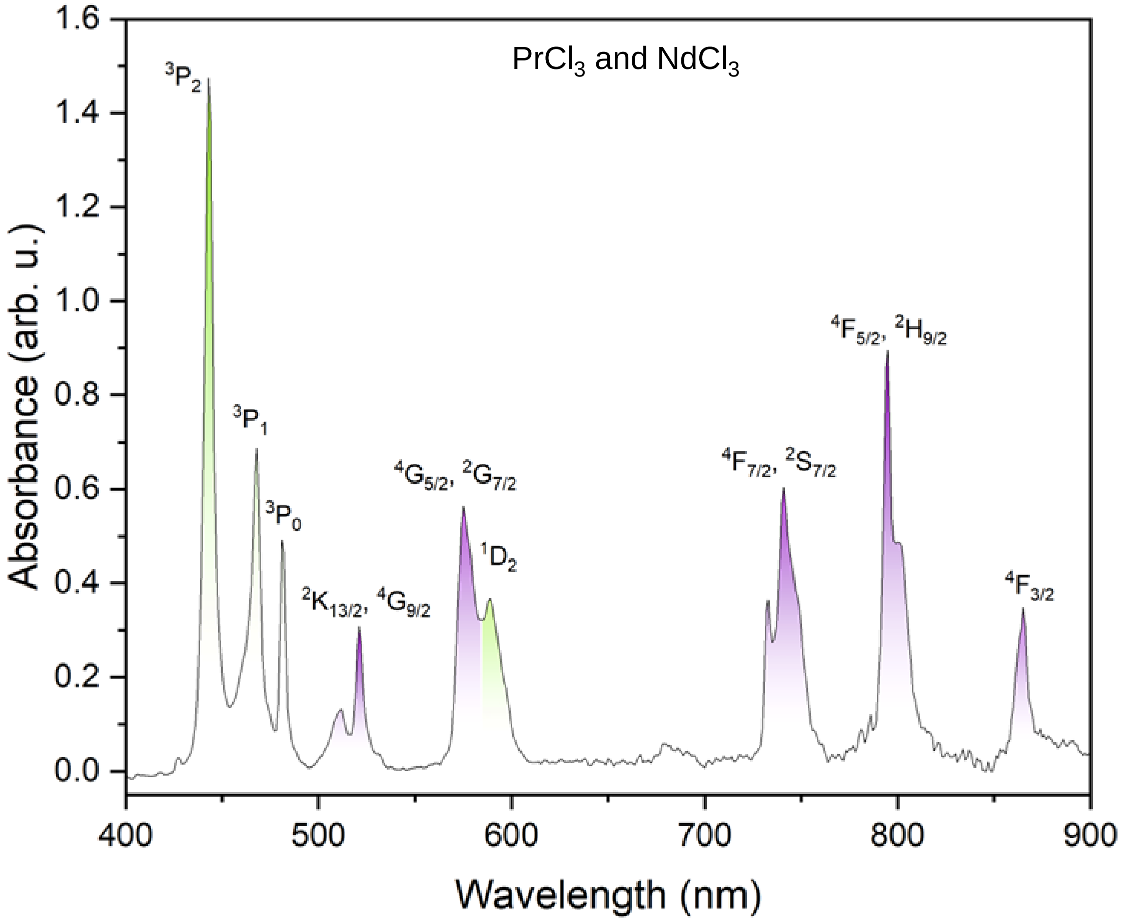
- Avantes 5 mm flow cell w/ SMA fiber optic connections
- Flow was set to ~1 GPH (controlled via venturi flow rate)
- Cary 60 Spectrometer (400-900 nm)
- 600 nm/min scan rate

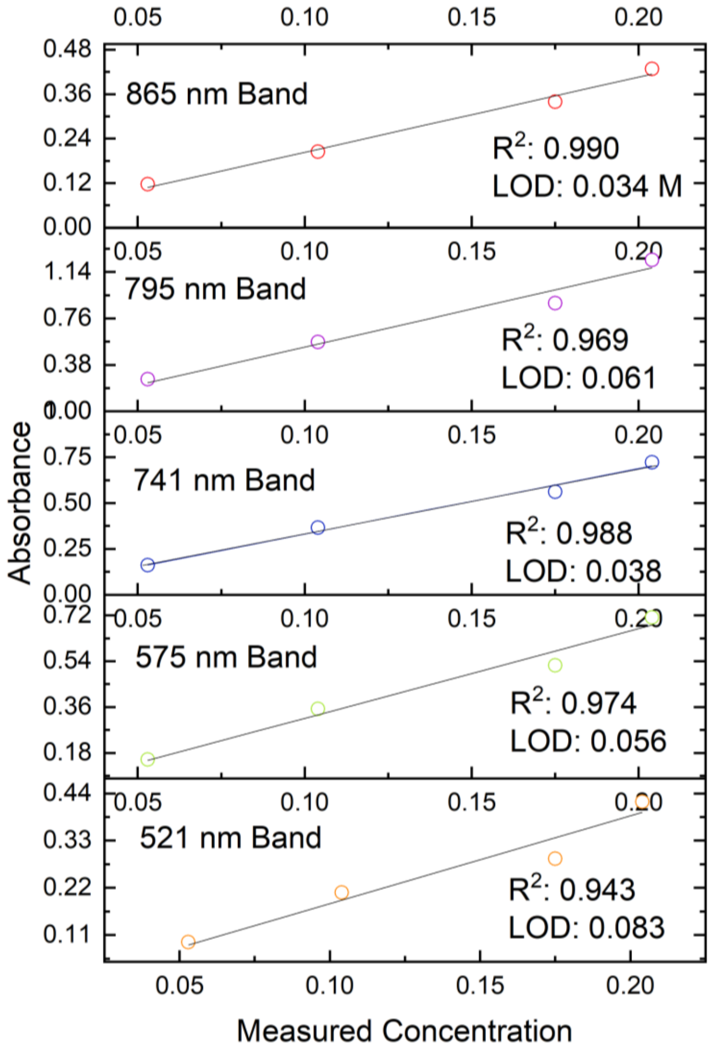



## LIBS

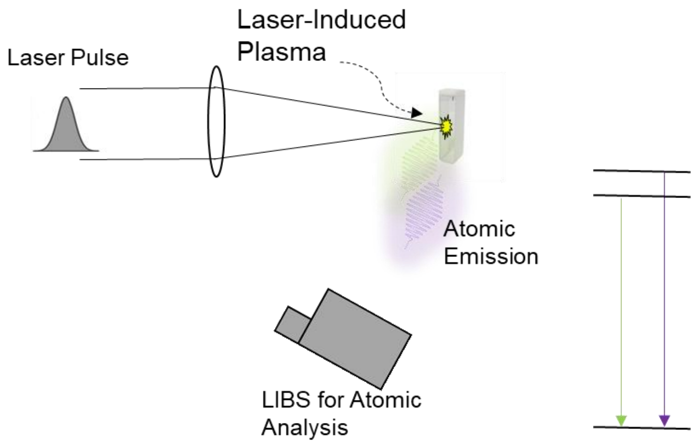
- Nd:YAG Q-SMART 450
  - 1064 nm
  - ~40 mJ
- 75 mm plano convex lens
- EMU 120/65 LIBS Spectrometer
- 3 μs gate delay
- 1 ms gate width
- 3-jet Collison nebulizer @ 40 psi

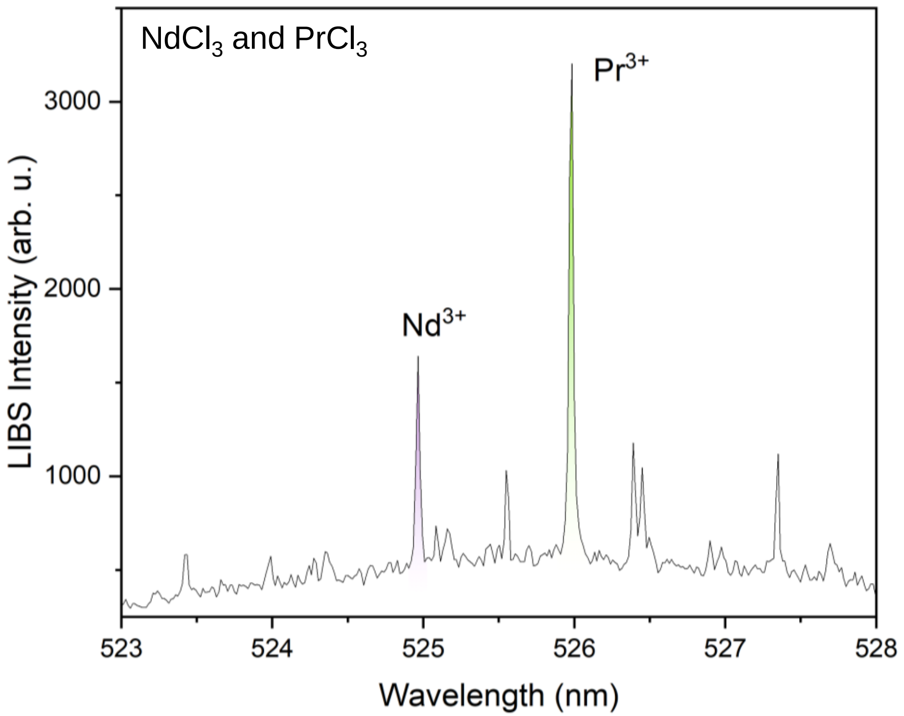
# Absorption Spectroscopy

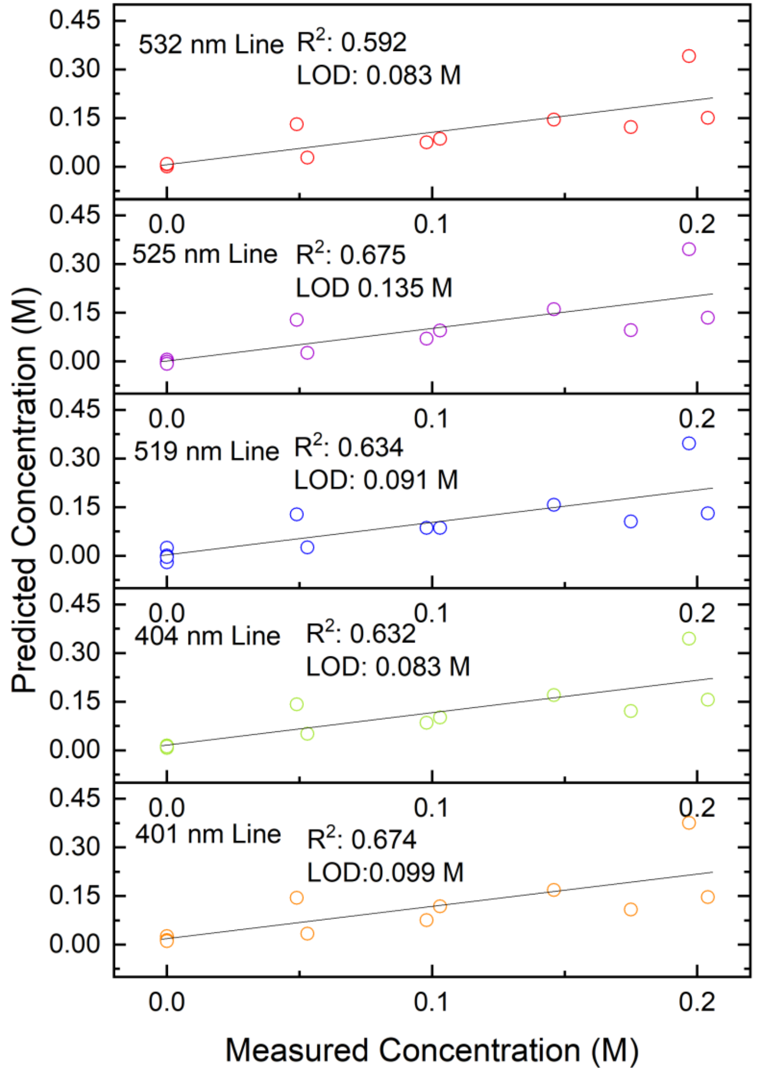



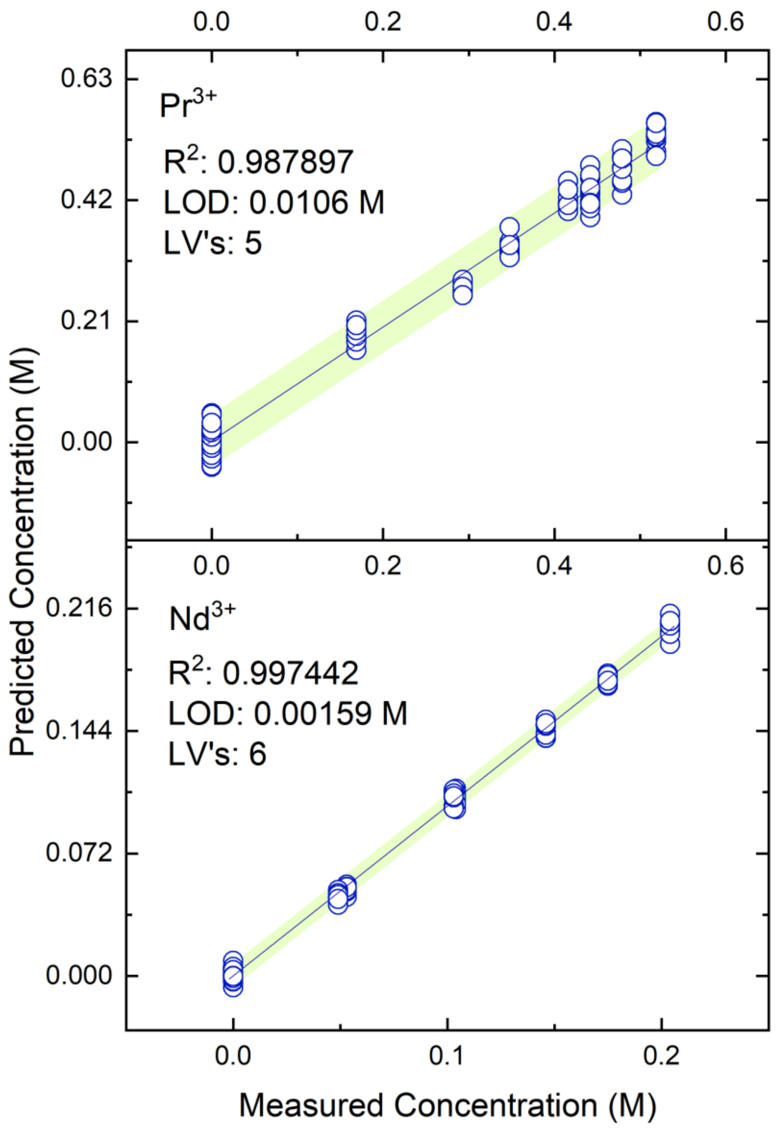
# LIBS Spectra



# LIBS Multivariate Calibration Curves




- PLS Toolbox Software utilized
- Multiple peaks are used through partial least square (PLS) regression to minimize unwanted statistical variation

LIBS analysis of $Pr^{3+}$

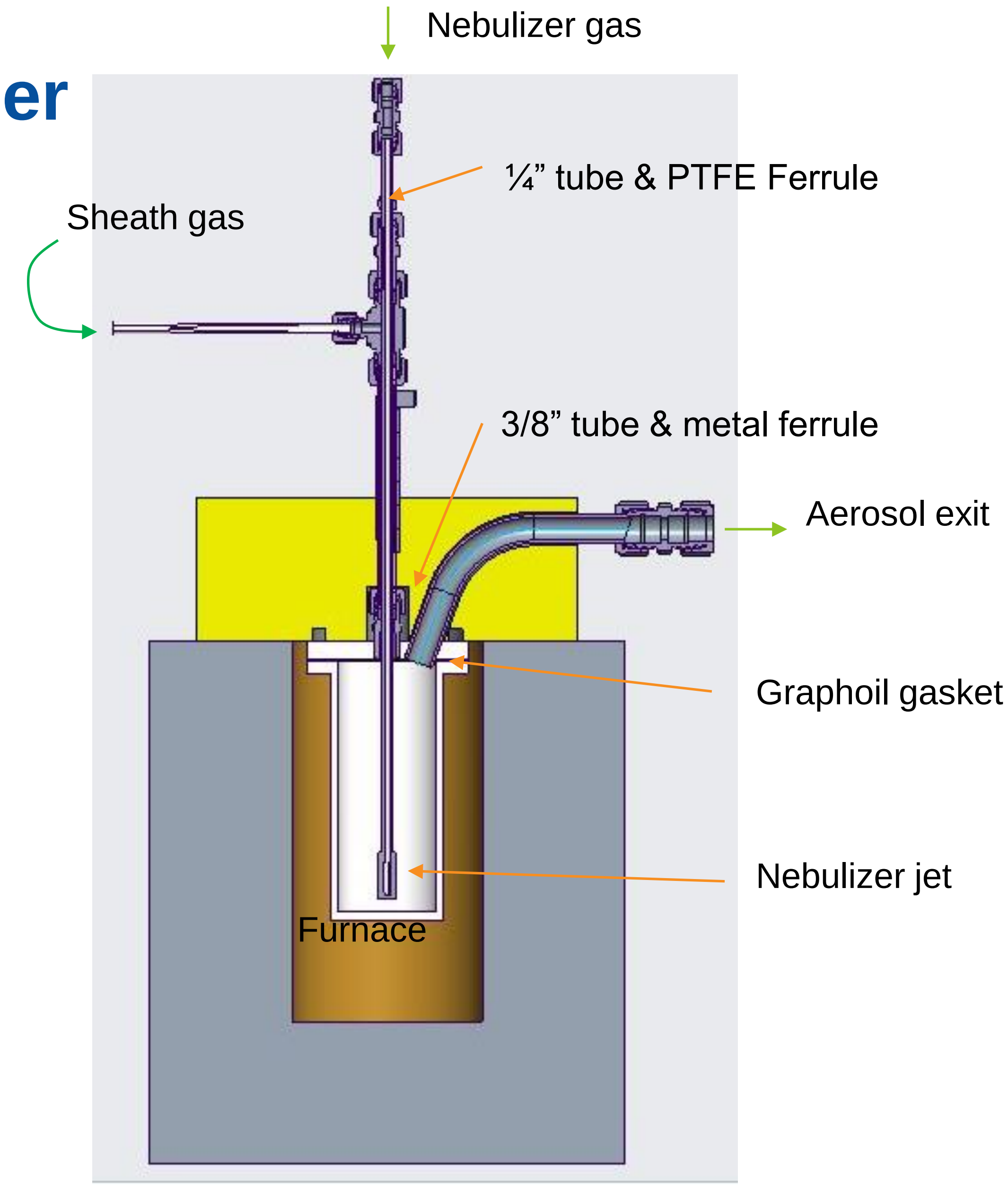
| Approach | $R^2$ | LOD (M) |
|---|---|---|
| Univariate | 0.837 | 0.129 |
| Multivariate | 0.988 | 0.0106 |

LIBS analysis of $Nd^{3+}$

| Approach | $R^2$ | LOD (M) |
|---|---|---|
| Univariate | 0.675 | 0.091 |
| Multivariate | 0.997 | 0.00159 |

# Scaling up to Molten Salts: Nebulizer Design

- Challenges:
  - High-temperature seals?
  - Difficult to adjust the depth of nebulizer tip:
    - Nebulizer jet locked due to metal Swagelok ferrules in high temperature zone
    - Freeze the tip into the salt? No, couldn't remove jar
- Solution:
  - Custom stainless-steel jar with flange & graphoil gasket
  - PTFE ferrule to seal the nebulizer jet tube above the heated zone
  - Set of concentric tubes with purge gas to prevent vapor & aerosol from freezing the tubes together



# Scaling up to Molten Salts: High-temperature Nozzle Design

- The 1/4” exit nozzle used in the aqueous study difficult to manufacture and led to holdup in tubing
- Made 3/8” and ½ inch nozzles and tested flow conditions:
  - Sheath gas flow 0-4 cfm
  - Nebulizer pressure 20-60 psi
  - Ranked based on degree of mist on the windows
- Both nozzles had regions where they worked well
  - 3/8” was more forgiving and would be easy to manufacture
  - Both showed less holdup in the tube
- In the process of fabricating a 3/8 nozzle from stainless steel

3/8” 40 psi, 1 cfm

1/2” 40 psi, 0.4 cfm

# Upcoming work:

- Explore Raman in aqueous aerosol
- Absorption in the aerosol phase?
- Metal nozzle and optical cell fabrication
- Setup furnace & heating
- Perform LiCl-KCl molten salt tests with:
  - $CeCl_3$
  - $PrCl_3$
  - $NdCl_3$
  - $RuCl_3$



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# Summary

- Circulating sampling and aerosol system can be operated for long periods without issues
- Simultaneous LIBS/UV-Vis achievable in a circulating system
- Larger nozzles don’t negatively effect aerosol stream but are easier to make and lead to less holdup

# Thanks to All Collaborators, Contributors, and Sponsors!

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