INL/CON-24-78812-Revision-0

Molten Salt and Off-Gas Monitoring Using Combined Spectroscopic Methods

June 2024

Robin Virginia Roper, Micah Edward Raab, Ruchi Gakhar, Ammon N Williams, Qiufeng Yang

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Prepared for the
U.S. Department of Energy
Under DOE Idaho Operations Office
Contract DE-AC07-05ID14517, DE-AC07-05ID14517

June, 2024

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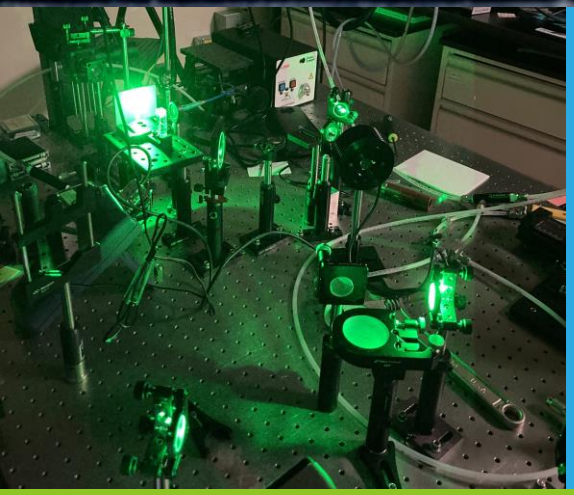


Molten Salt and Off-Gas Monitoring Using Combined Spectroscopic Methods

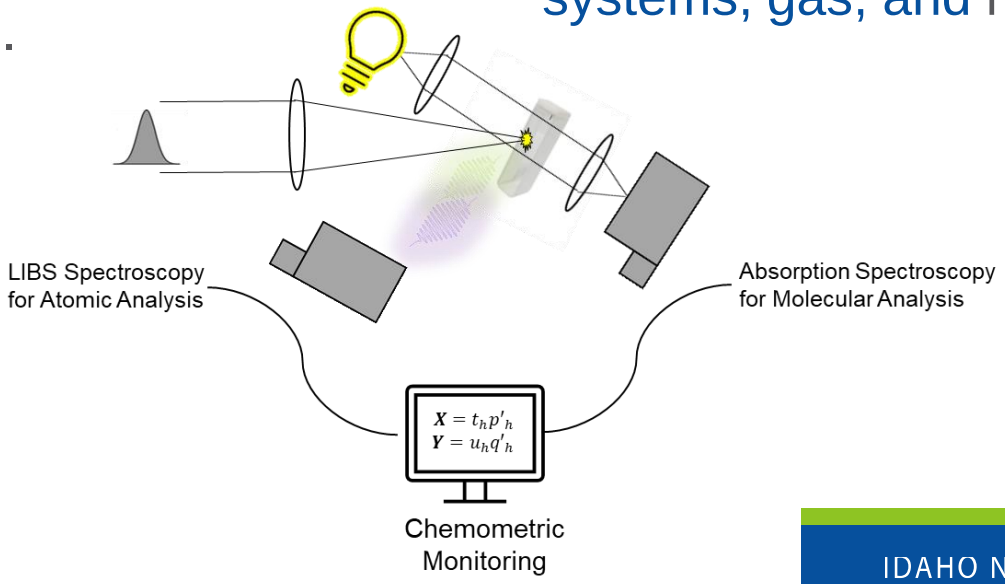
INL PI: Ruchi Gakhar and Ammon Williams

Idaho National Laboratory

Motivation and Approach

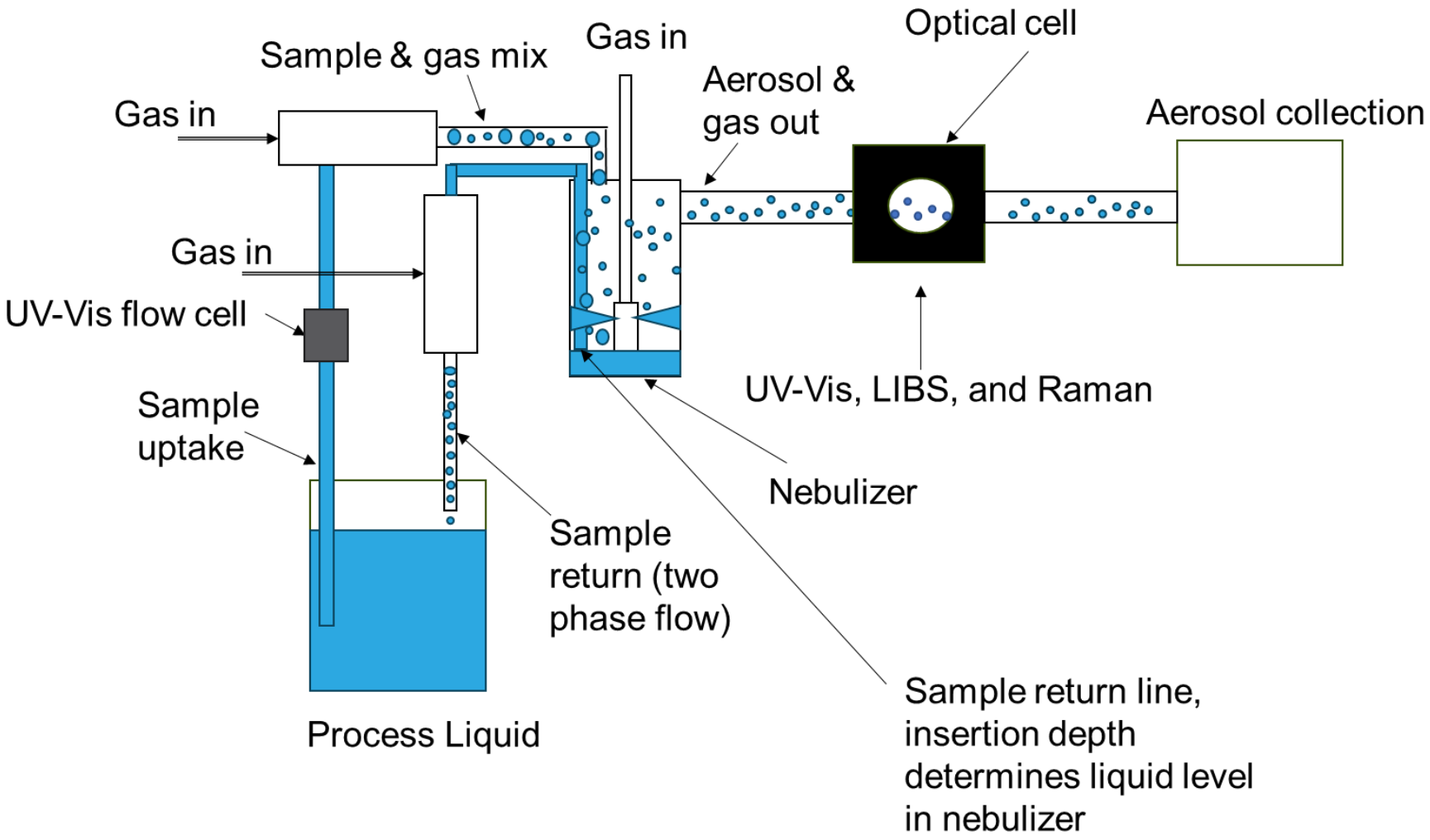
- Nonproliferation and safeguards is a challenge for liquid fueled MSRs as most current research is focused on discrete sampling and offline analysis leads to delays.
- The physical and chemical properties of molten salts are not well understood.

- Real-time atomic and molecular analysis using combined traditional spectroscopies: LIBS, UV-Vis absorption, and Raman (underway)
- Partial least squared (PLS) regression for precise quantification
- Speciation in molten salts
- Three phases: aerosol/flowing systems, gas, and molten salt

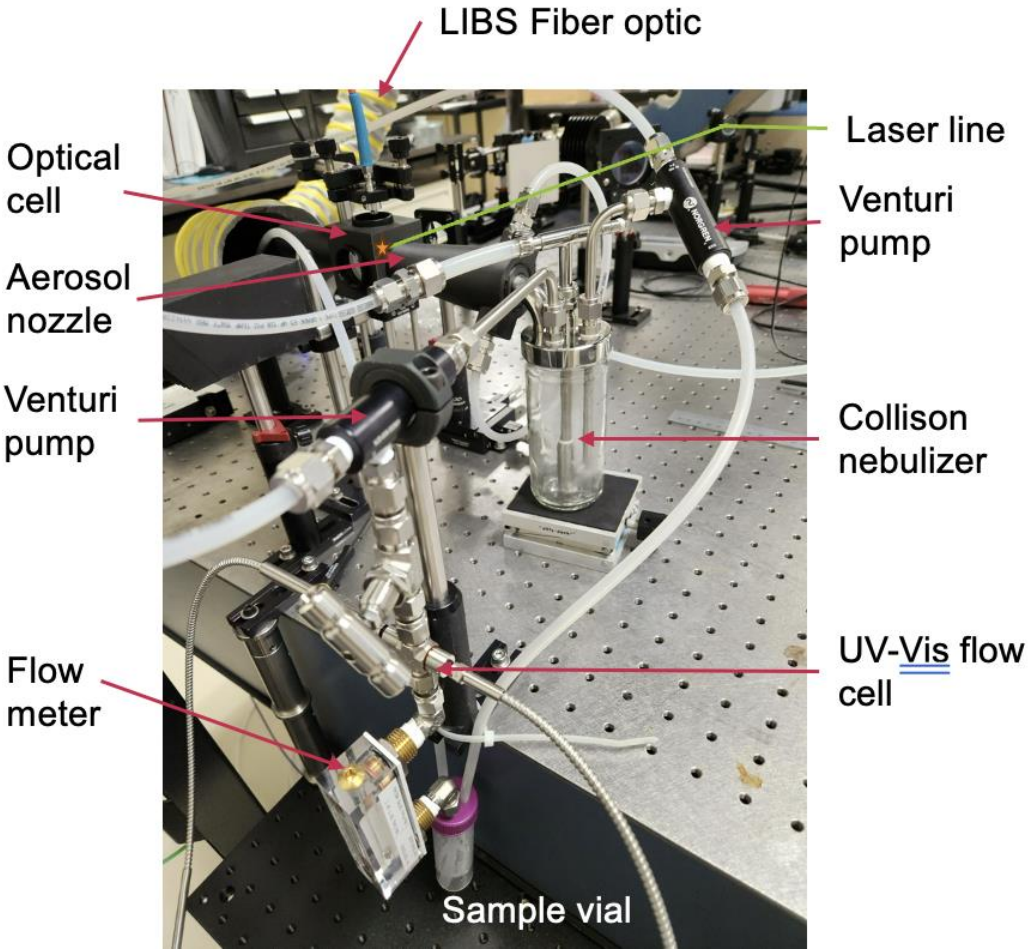


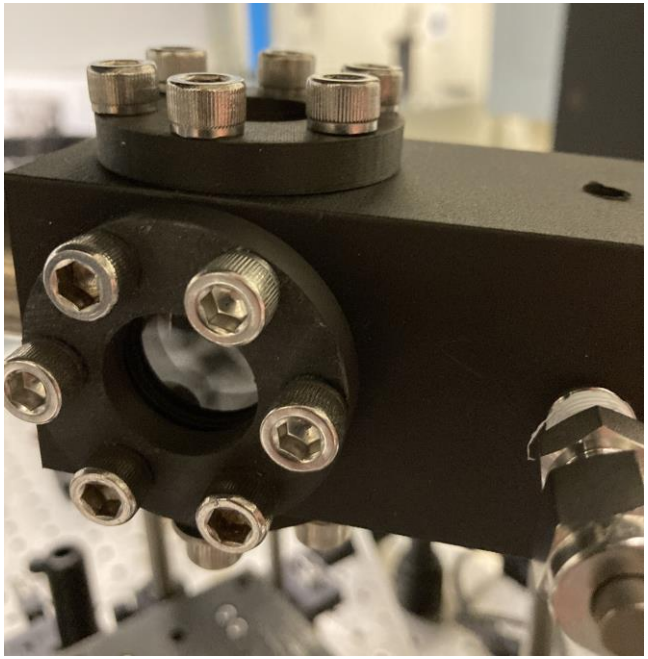
Concept & Design

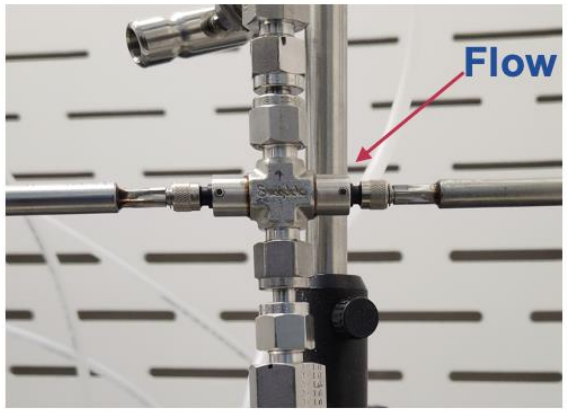
LIBS in aerosol circumvents splashing caused by produced shock waves in molten salts



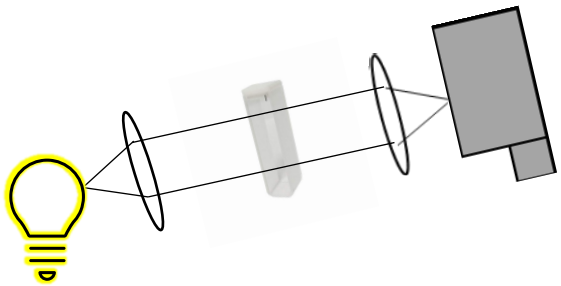
Experimental Setup



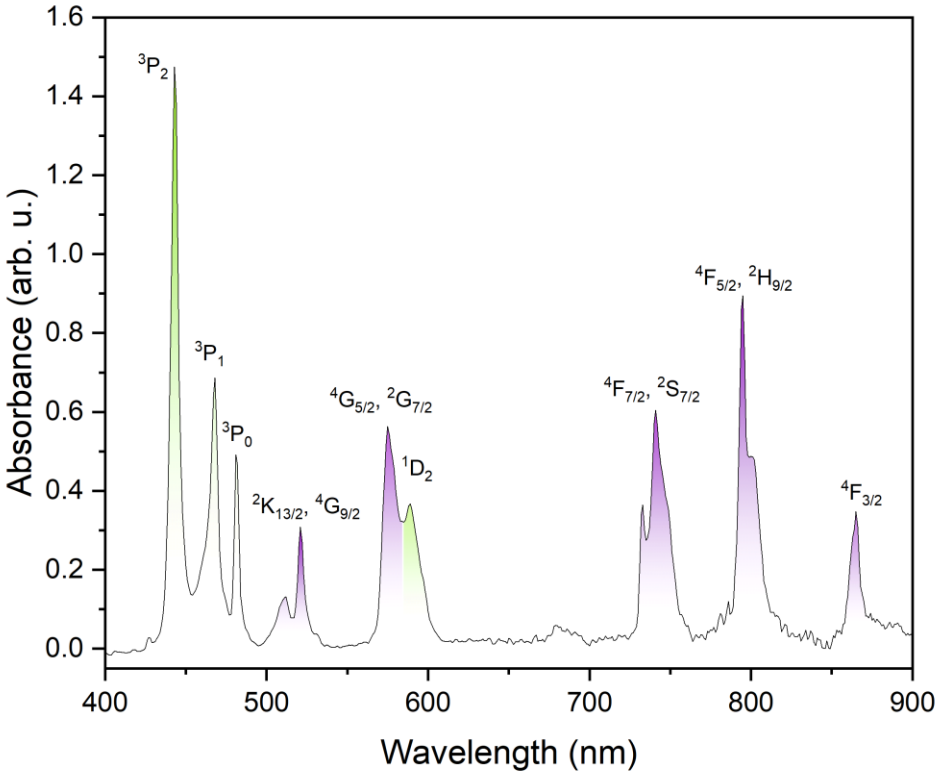


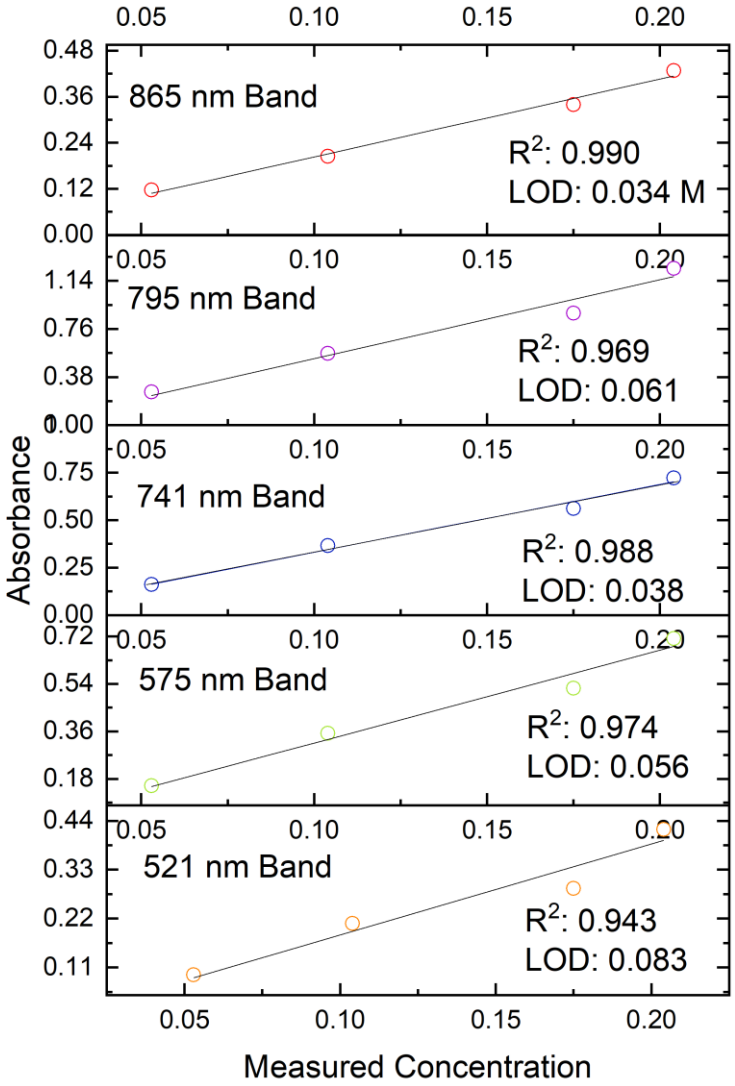


Calibration Curves Generated by Absorption Spectroscopy in Liquid

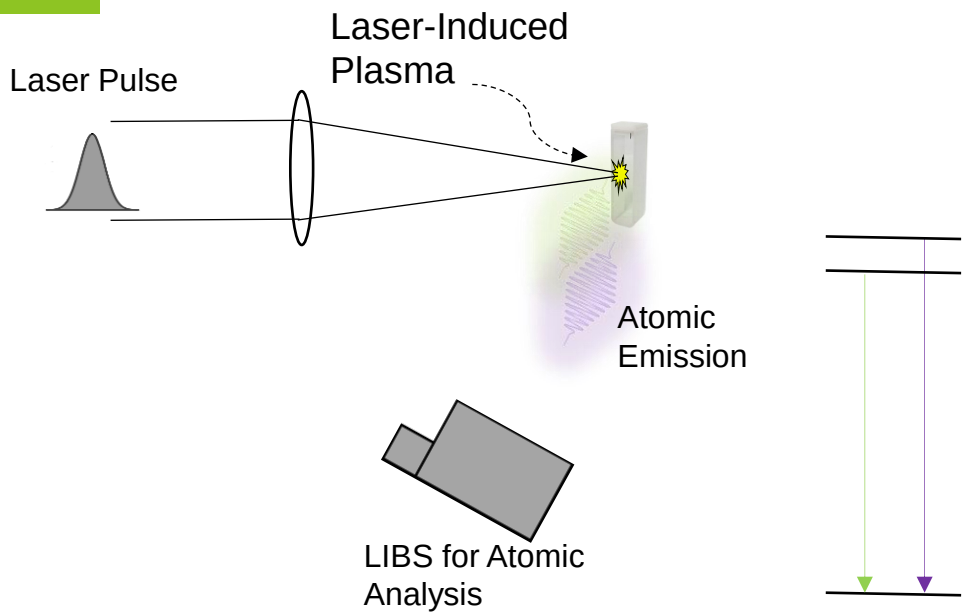


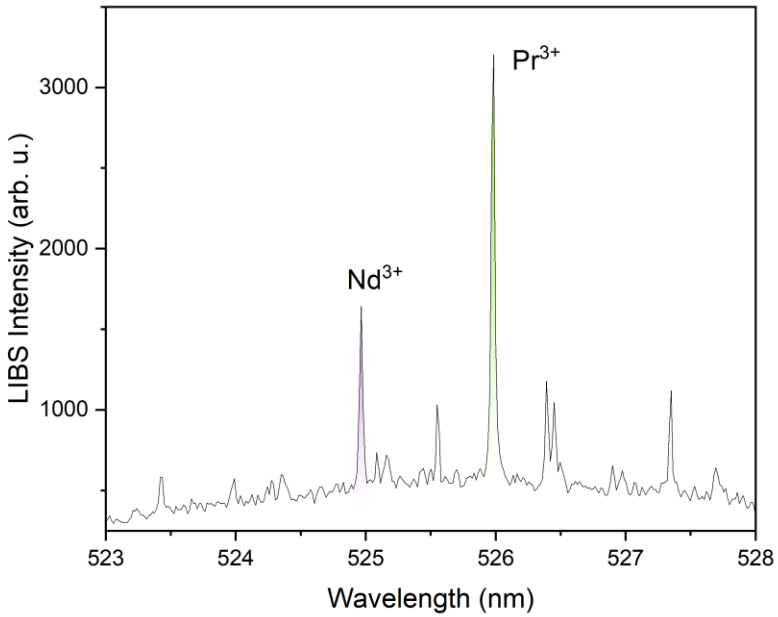
Absorption Spectroscopy for Molecular Analysis

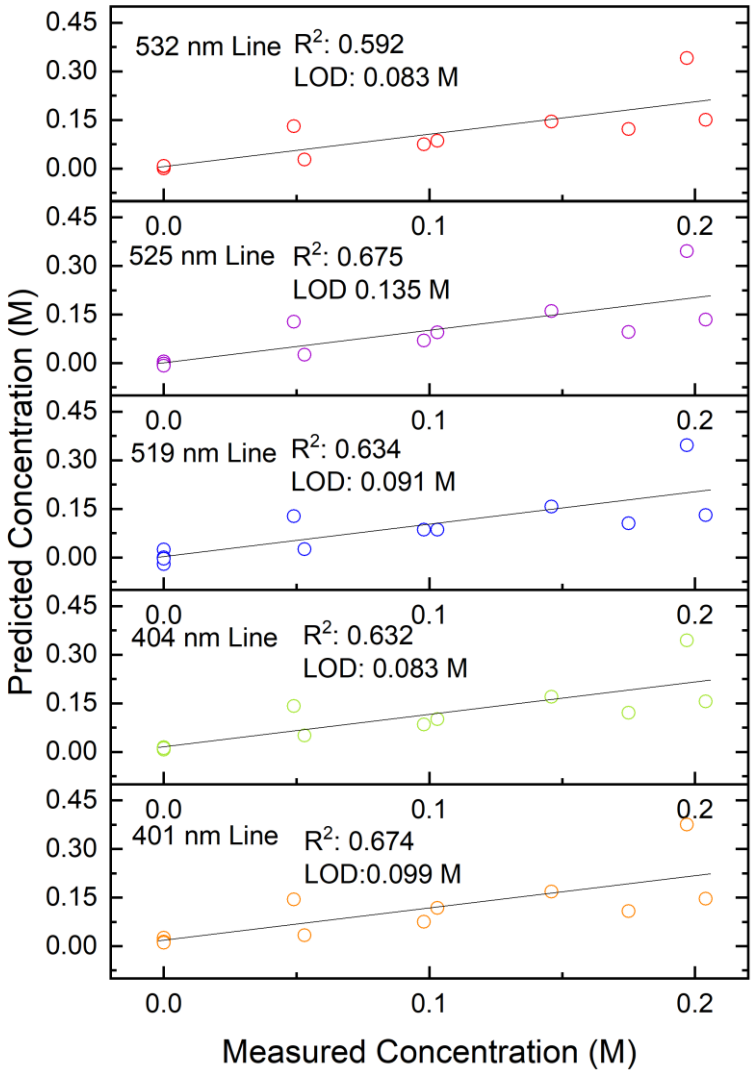




Univariate Calibration Curves Generated by LIBS







[Nd]: 0.05-0.2 M

Multivariate Calibration Curves

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

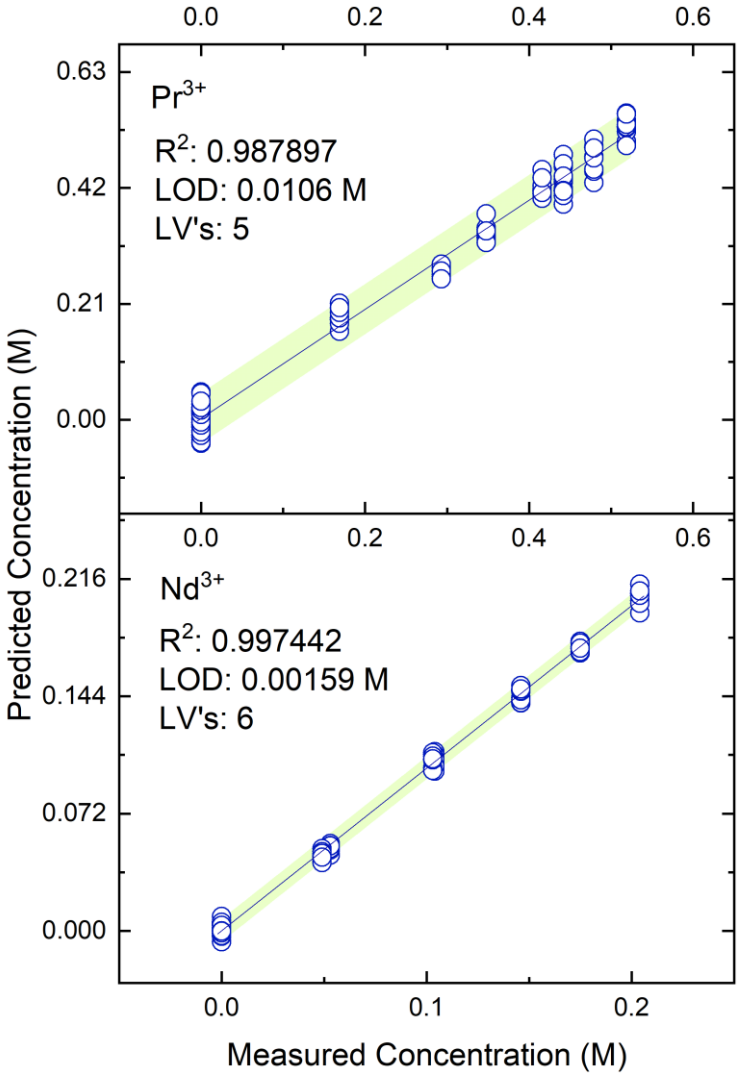
LIBS analysis of Pr^{3+}

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

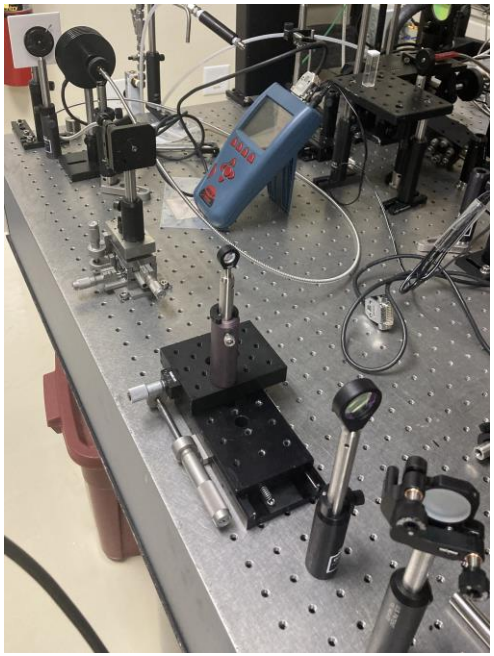
Multivariate analysis for LIBS gives higher R^2 and lower LOD

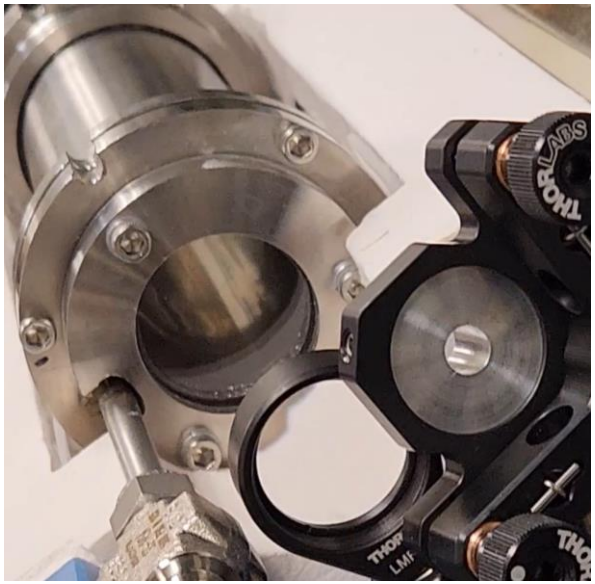


UV-VIS and LIBS Optical Configuration

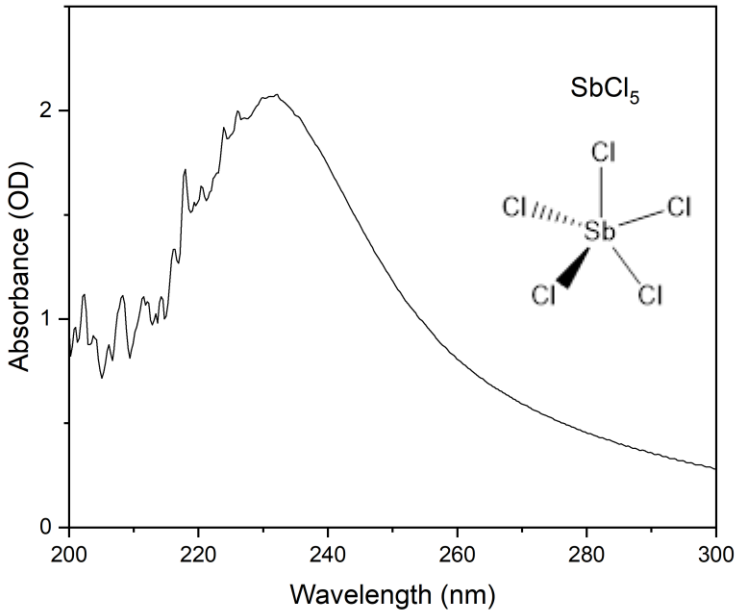
TerraPower seeks to apply chlorine-based volatility (CBV) to fission products. Here, uranium will be volatilized and separated based on boiling point difference.

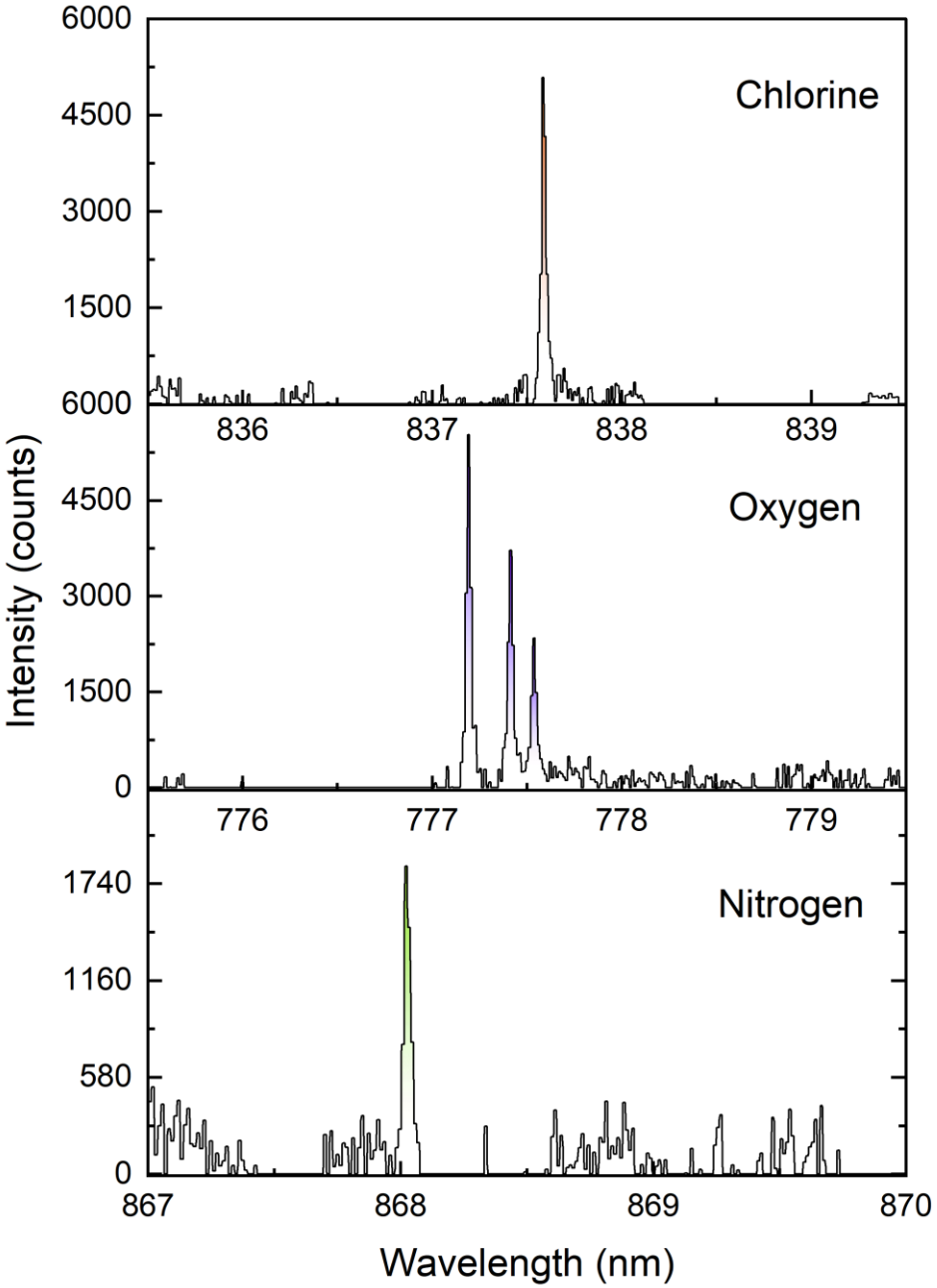
INL seeks to use combined spectroscopy for atomic and molecular analysis before and after separation.





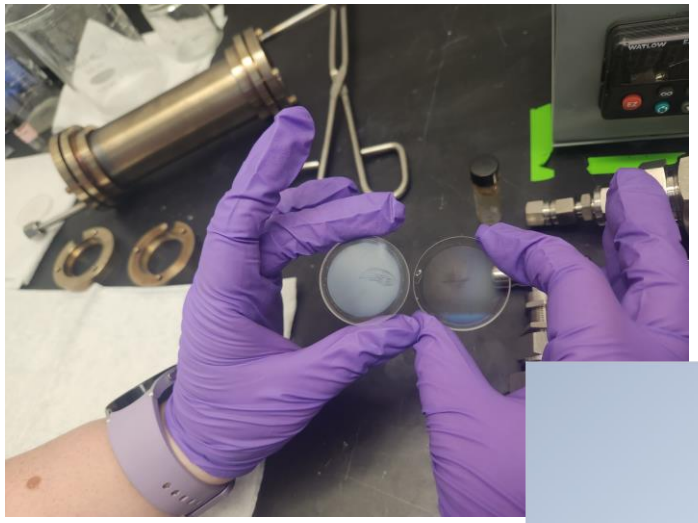
Preliminary Spectra of $SbCl_5$





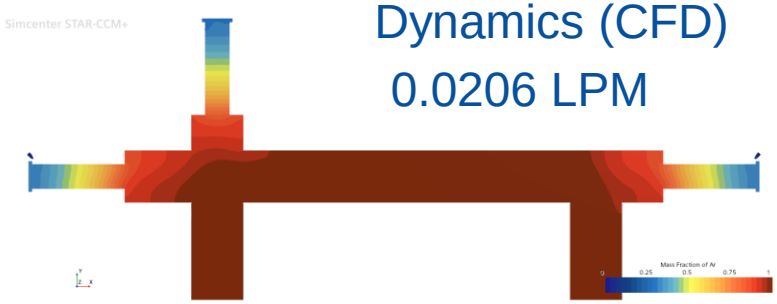
Mitigation of Material Deposition

Window Deposition

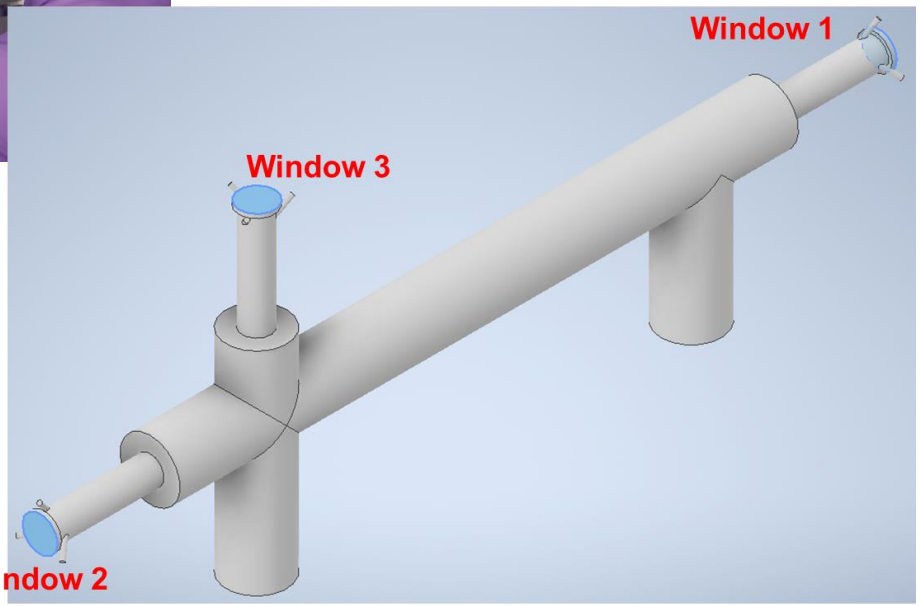


Computational Fluid Dynamics (CFD)

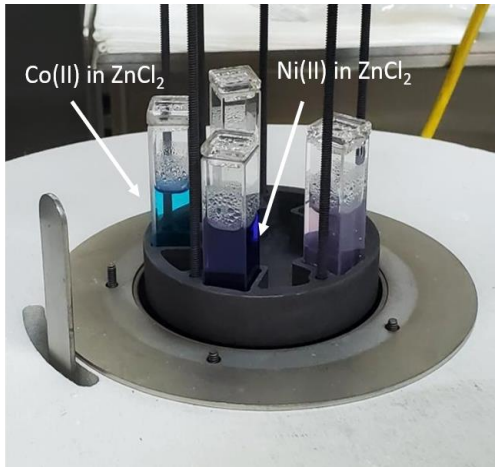
0.0206 LPM



New Cell Prototype



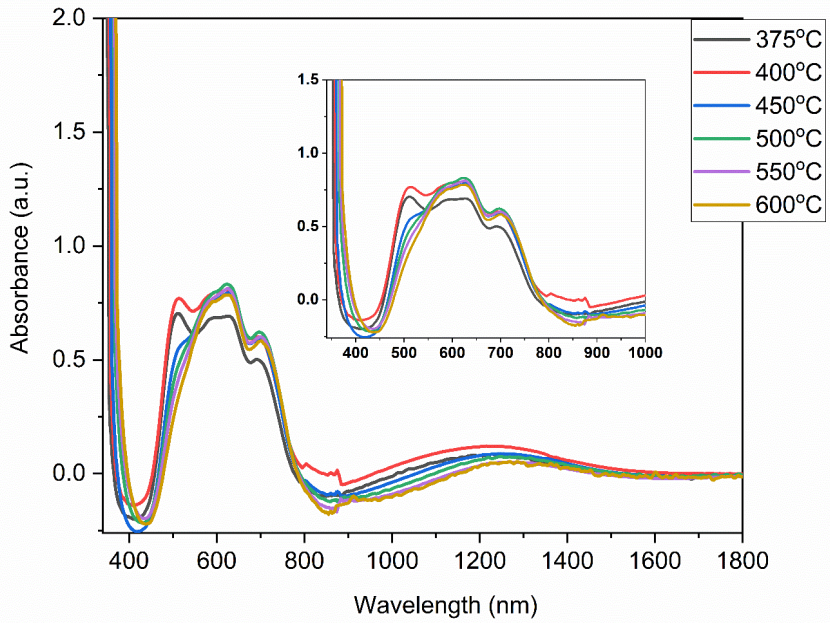
Advanced Spectroscopy Furnace Design



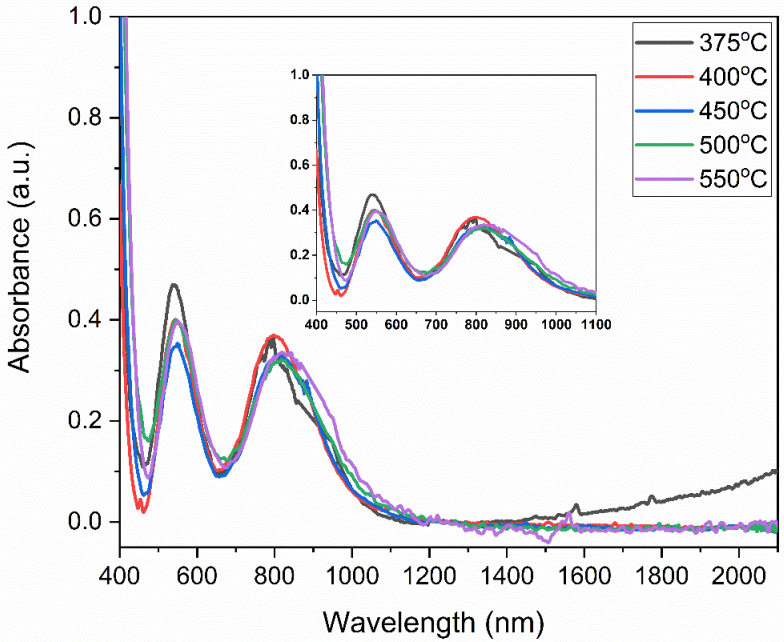
- Agilent Cary 5000 UV-Vis-NIR
  - 175-3,300nm spectral range
  - Up to 0.01nm resolution
- Custom fiber optics by Fiberguide
  - 200-1400nm (UV-Vis)
  - 300-2300nm (Vis-NIR)
- Custom designed furnace for *in-situ* spectroscopy and electrochemistry
  - Five sample slots
  - Accommodates standard quartz cuvettes

Rev. Sci. Instrum. 91, 083105 (2020)

Effect of temperature on metal speciation



Ni(II) in LiCl-KCl eutectic melt (M. Pt. 352°C)



Cr(III) in LiCl-KCl eutectic melt (M. Pt. 352°C)

Clear differences in coordination geometry and heterogeneity observed for Ni(II) environment, however Cr(III) exhibits homogeneous environment

Thanks to All Collaborators, Contributors, and Sponsors!

This work was supported U.S. Department of Energy (US-DOE), under these programs:

- ARPA-E ONWARDS program in collaboration with TerraPower
- INL LDRD program under Contract No. DE-AC07-05ID14517 at INL

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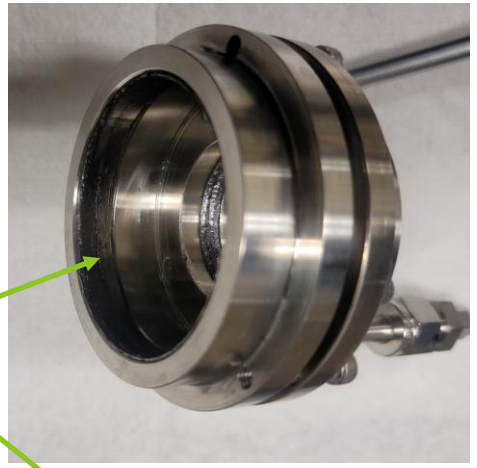
Robin Roper

Micah Raab

Postdoctoral Fellows

Optical Cell Components

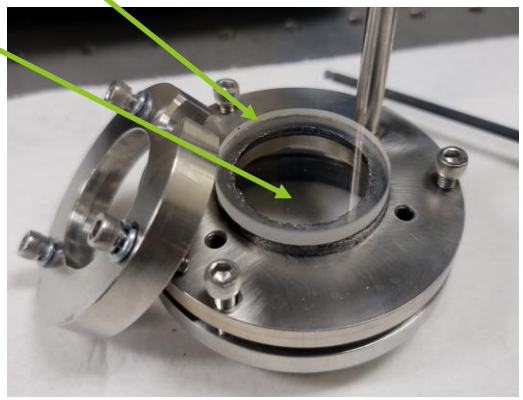
Inside Seals

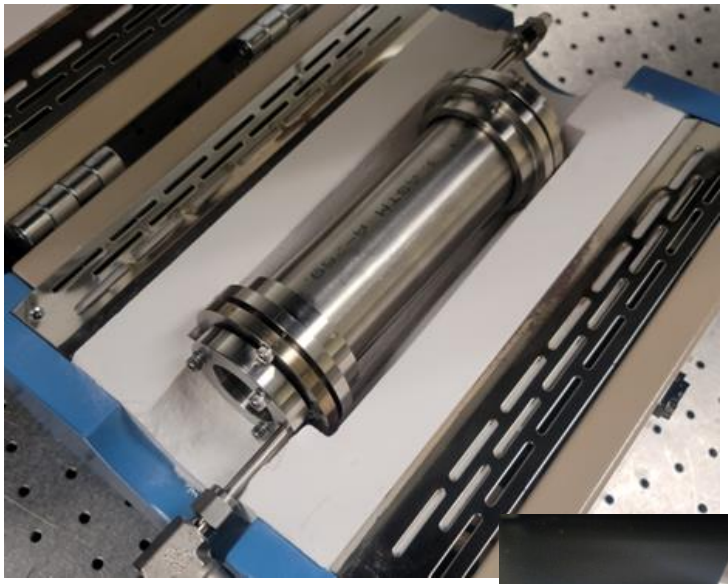


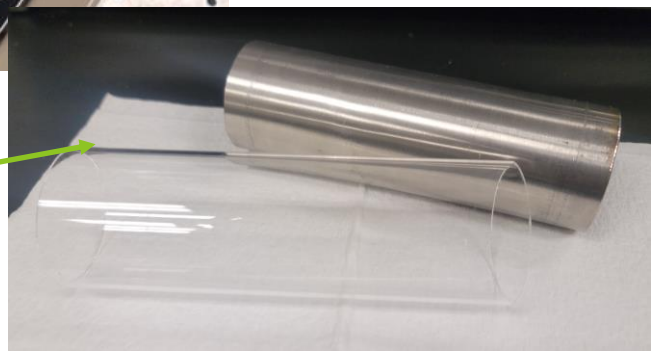
Graphite Seals

Quartz or sapphire windows

Window Seals



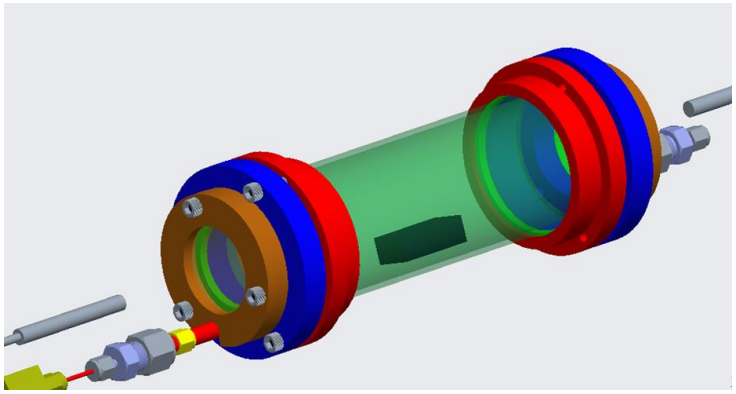


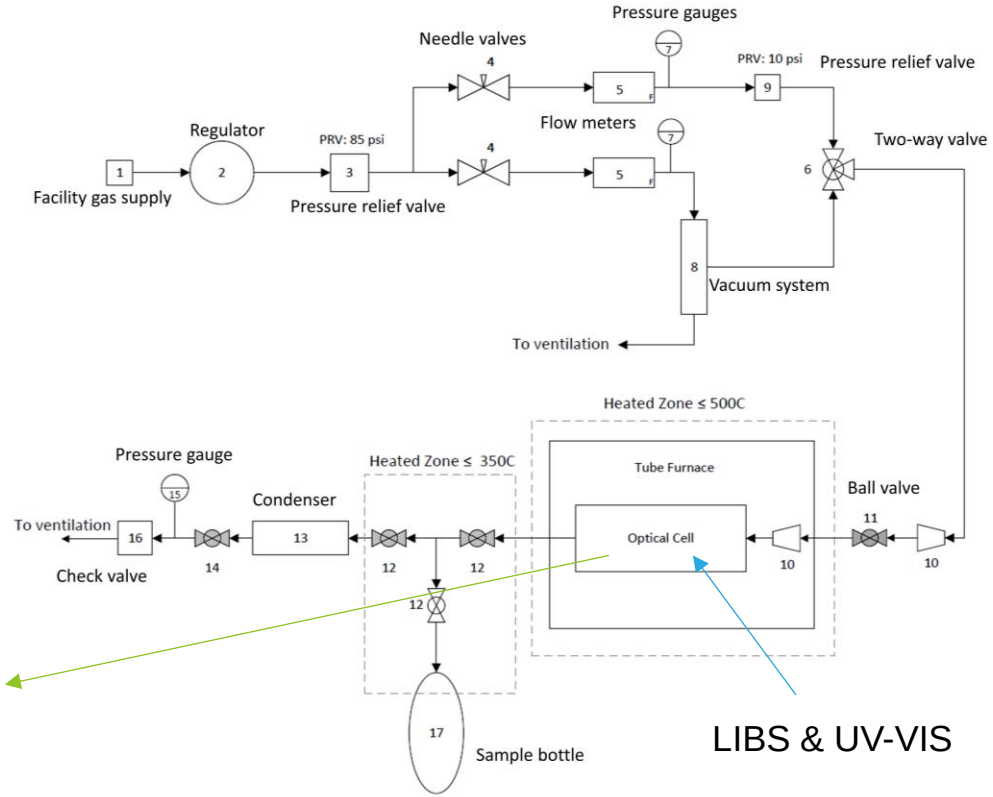


Stainless steel or Quartz body (length can vary)

Optical Cell and Testing Apparatus Design

- Ability to operate from vacuum to <45 psi
- Operate as open system (gas flowing through) or closed system (valve off optical cell)
- Ability to sample the gas phase for off-line characterization





- LIBS: EMU echelle spectrograph with an emccd from Catalina
- Raman: Czerny-Turner spectrograph with a CCD from Andor