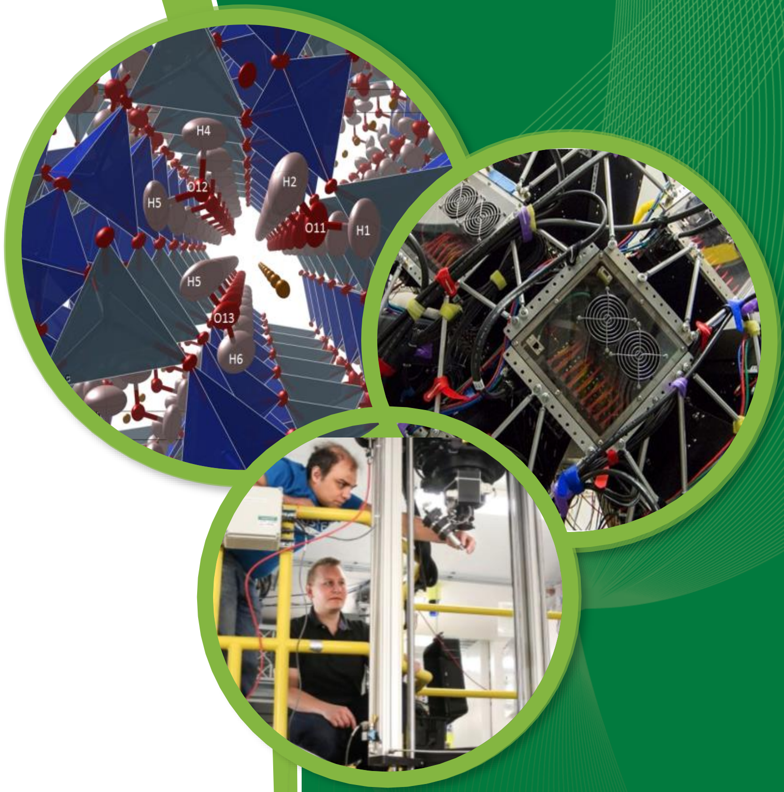
VISION

Neutron Vibrational Spectroscopy



Luke L. Daemen

Spallation Neutron Source

Oak Ridge National Laboratory

Oak Ridge National Laboratory | High Flux Isotope Reactor | Spallation Neutron Source

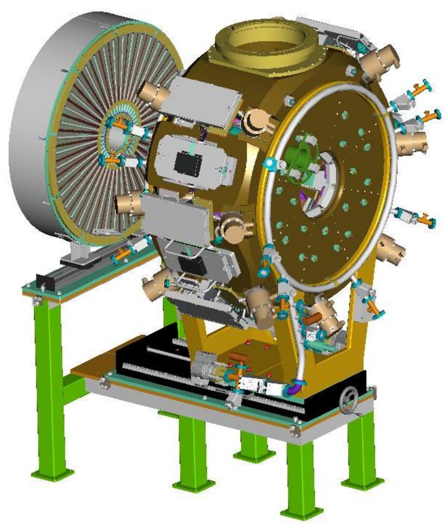
Outline

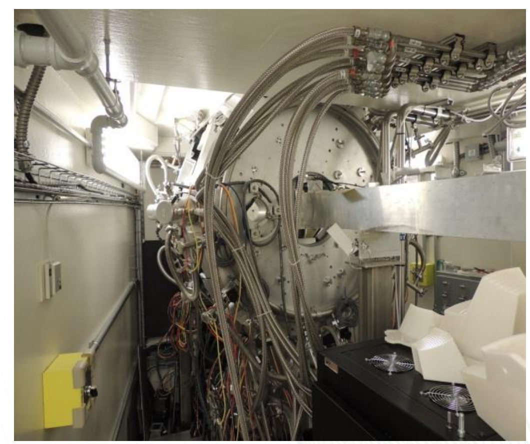
- VISION overview
- Scientific mission and impact
- Some statistics
- New developments since last review
- Science highlights
- Summary

VISION overview

- VISION is an inverse geometry, time-of-flight spectrometer used for neutron vibrational spectroscopy.

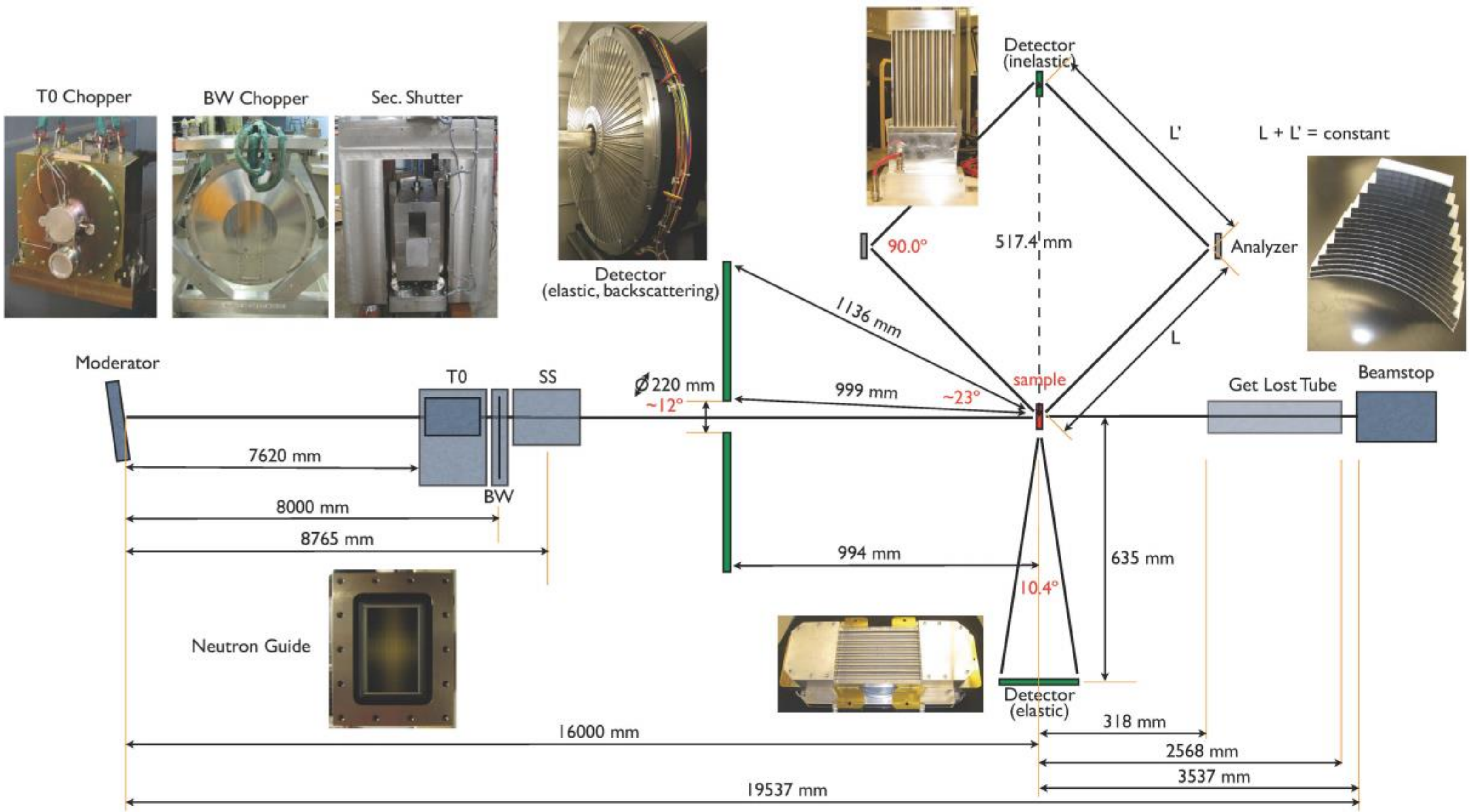
- 2001 - NSF funding for conceptual design
- 10/02 - EFAC meeting; VISION part of SING II
- 08/05 - SING II statement of mission
- 10/05 - CD-0
- 05/07 - CDR
- 02/08 - CD-2 start of construction
- 12/08 - Engineering design complete (design criteria document)
- 02/12 - Instrument completion of PEP parameters report
- 09/14 - Start of commissioning
- 01/15 - VISION enters user program





VISION overview: Instrument specifications

- White incident beam, fixed final energy (indirect geometry)
- High flux (~5×10^7 neutrons/cm^2/s) and double-focusing
- Broadband (-2 to 1000 meV at 30Hz, 5 to 500 meV at 60 Hz)
- Constant dE/E throughout the spectrum (~1-1.5%)
- Elastic line HMFW ~100 μs
- Backward and 90° diffraction banks



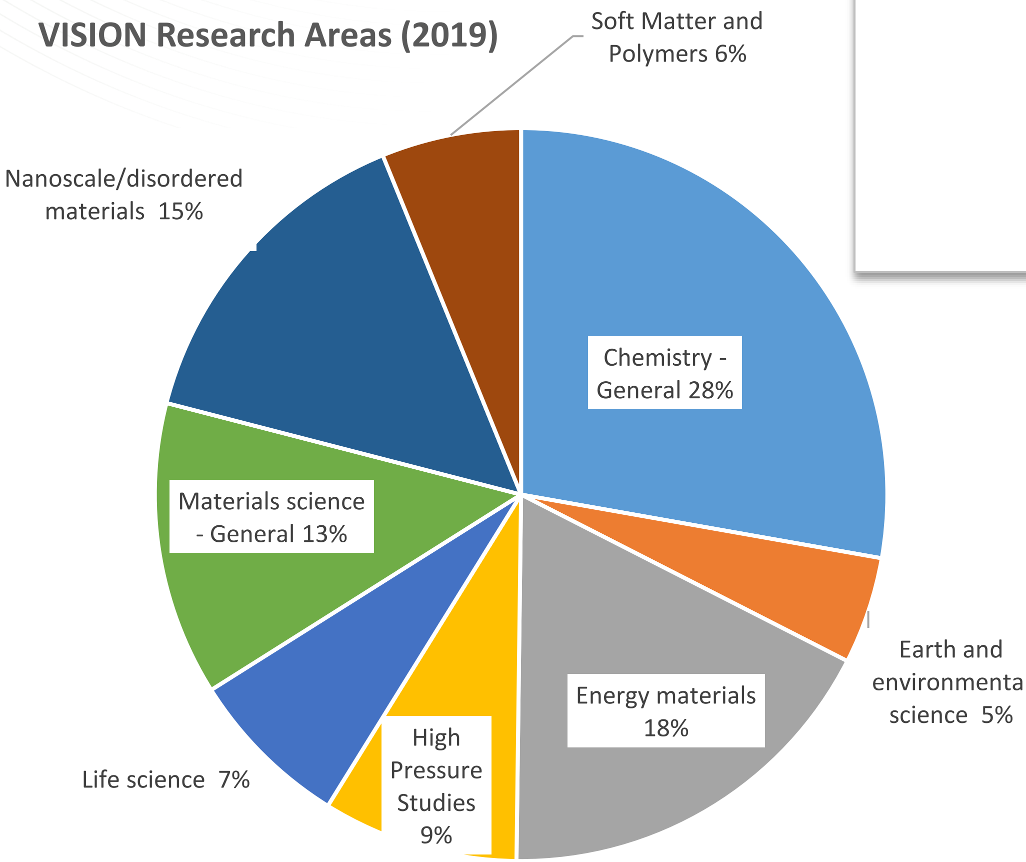
Scientific mission and impact

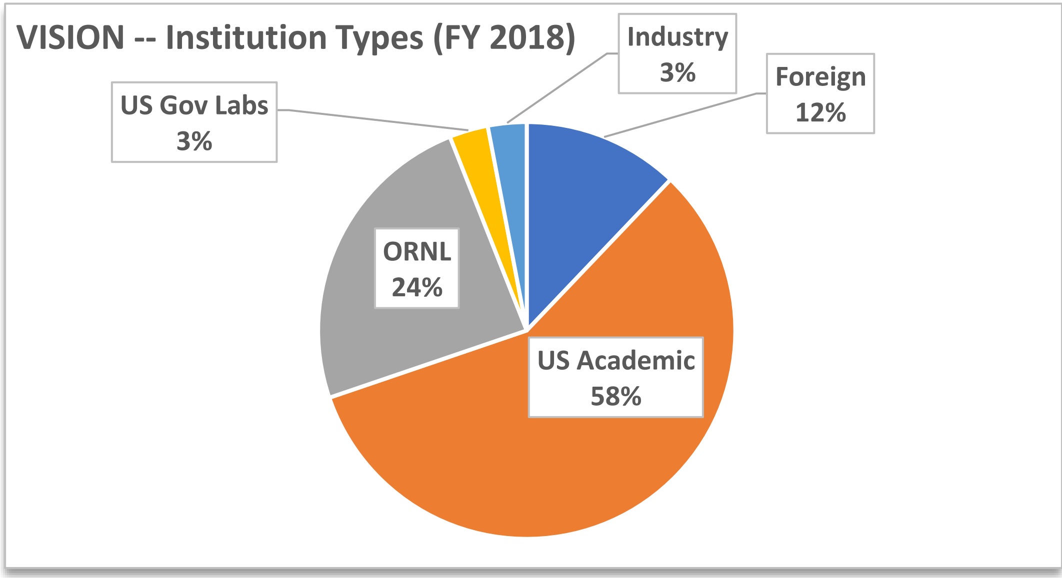
- VISION provides access to simultaneous neutron vibrational spectroscopy and powder diffraction to a diverse community of users in the fields of chemistry, physics, materials science, geology, and biology.
  - **Hydrogenous materials**
    - Metal Hydrides, biomolecules, water, organic/inorganic materials
  - **Functional/Advanced materials research**
    - Energy materials, catalysis, surface chemistry, porous materials geomaterials, thermoelectric, fuel cells, batteries, hydrogen storage, gas separation, natural materials
  - **Fundamental chemistry and spectroscopy**
    - PDOS, anharmonicity, modeling validation, intermolecular forces

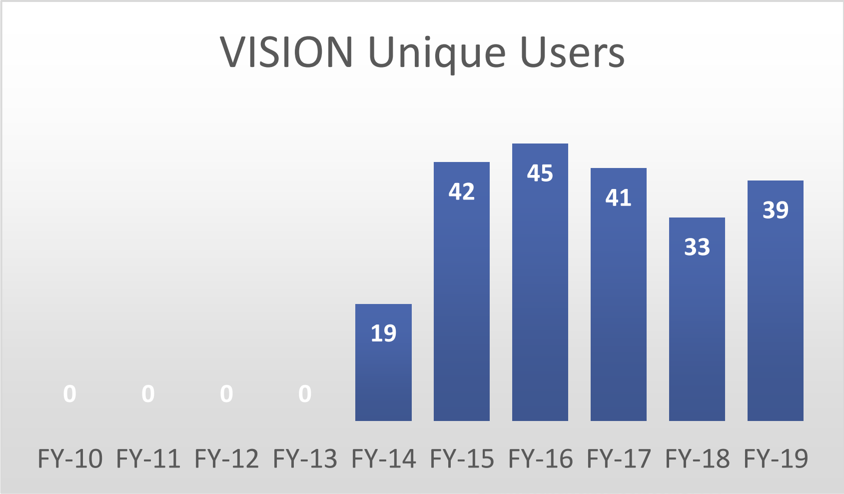
- Complements Raman/FTIR

The VISION user community

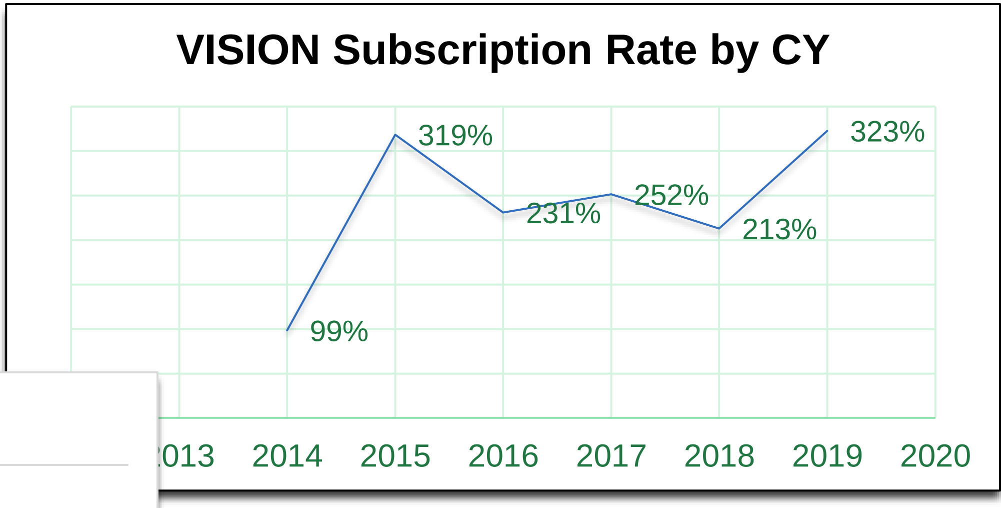
Greater science diversity

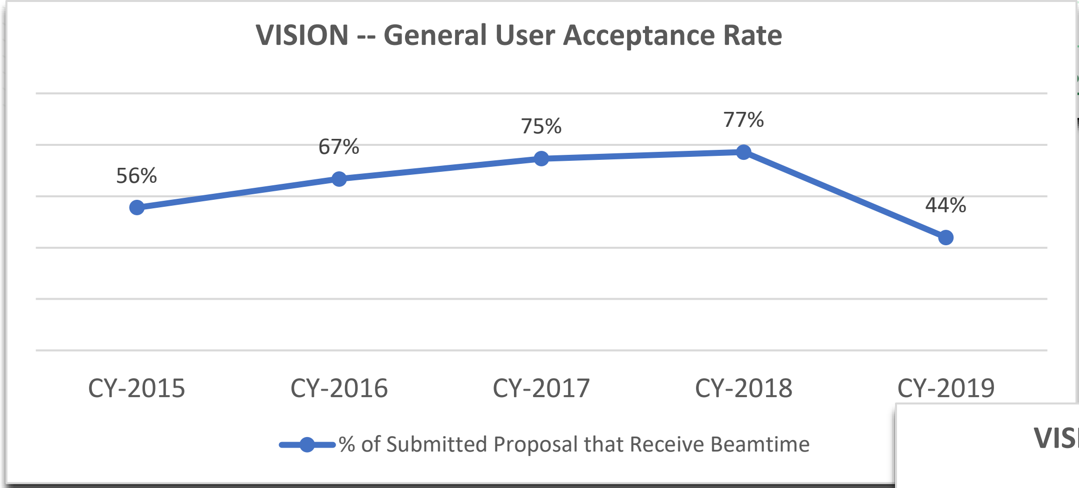


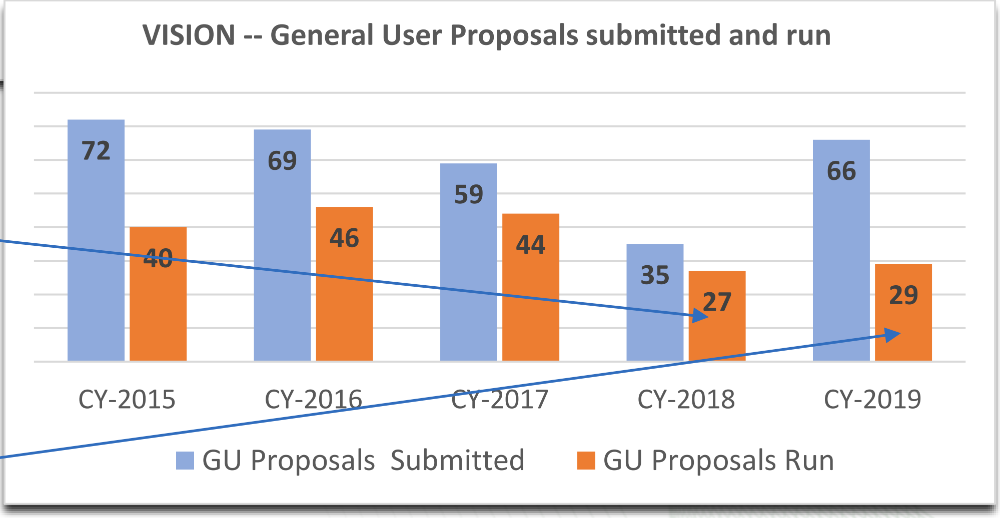




Some user statistics





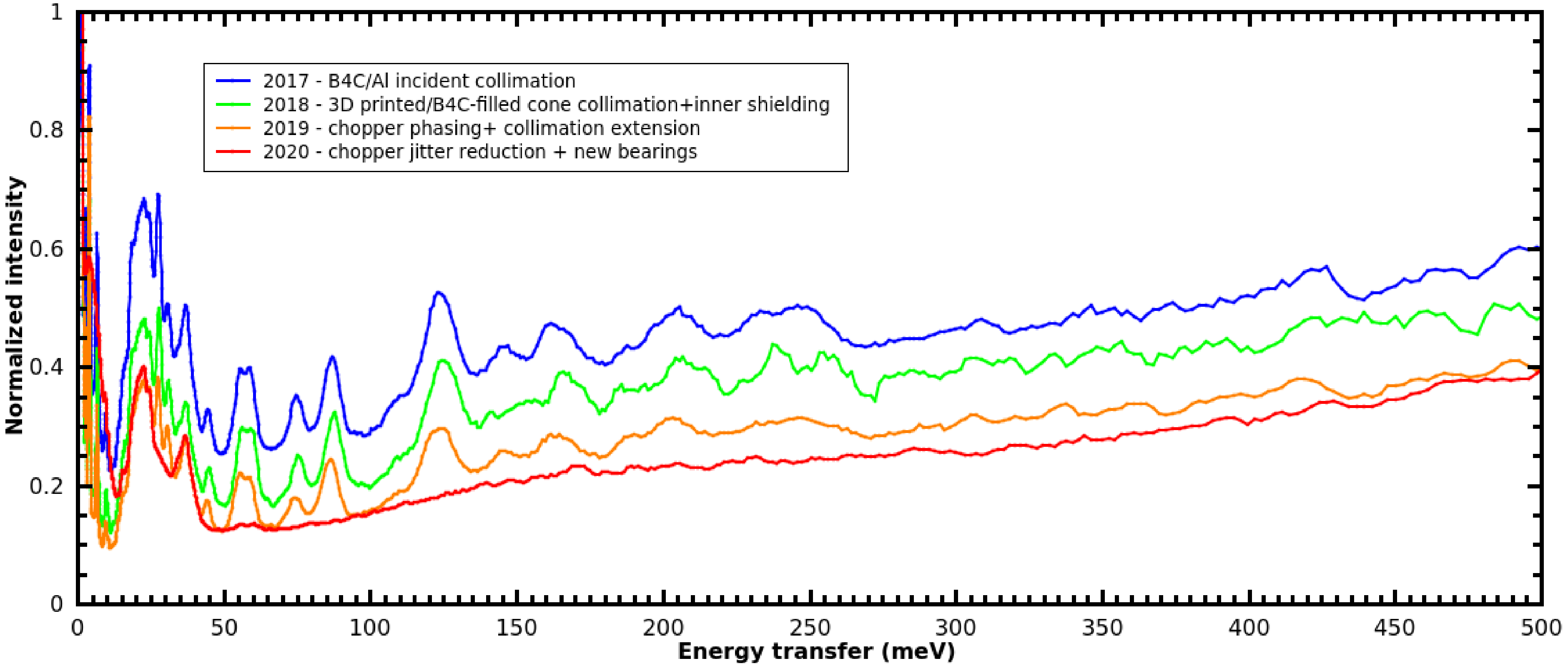


IRP replacement

62% beam availability in 2019

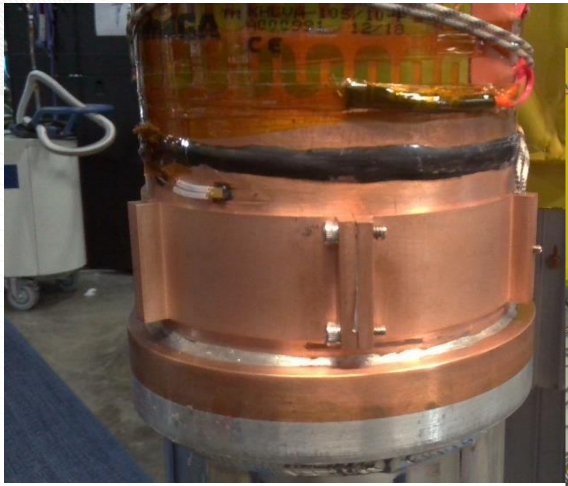
New developments

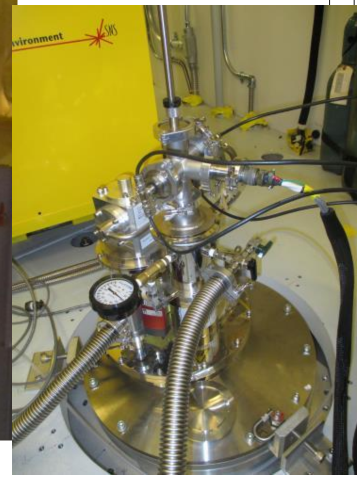
Background reduction

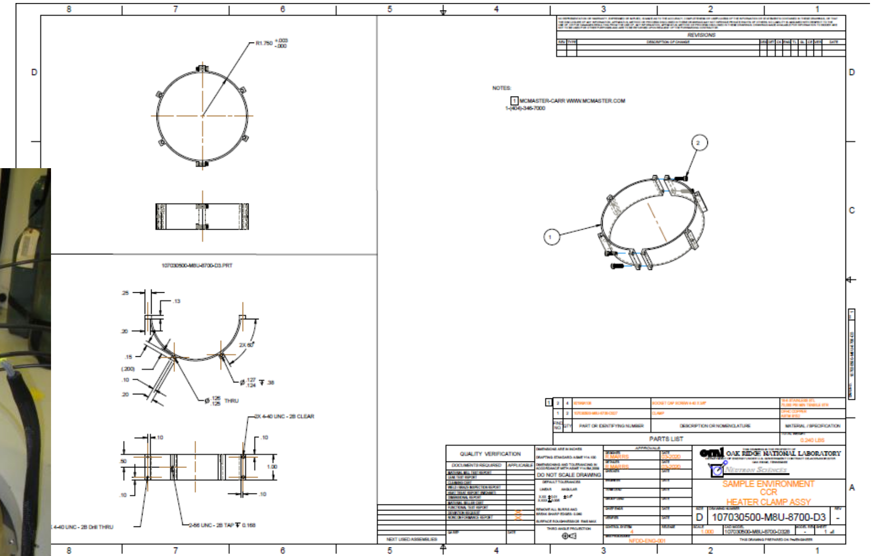


Low, structureless background essential for accurate difference spectra and high S/B with small samples.

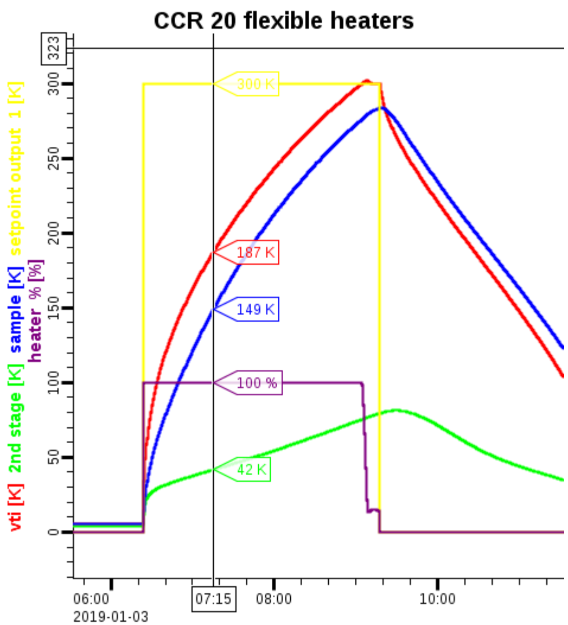
New developments



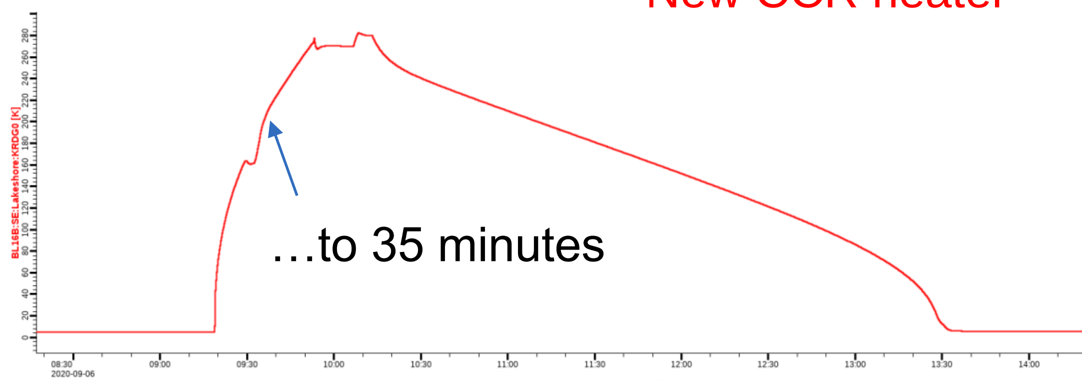




New CCR heater



from 165 minutes…



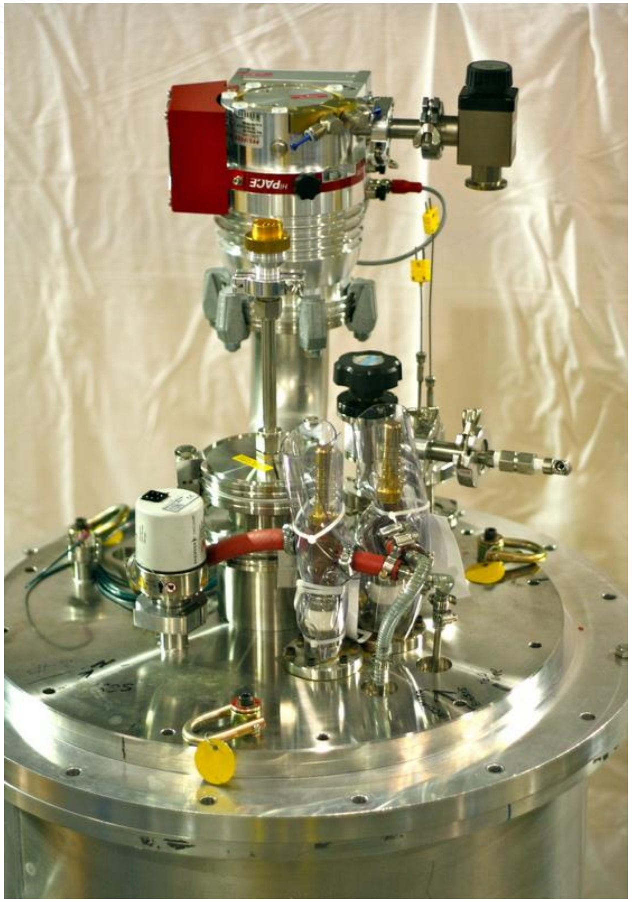
…to 35 minutes

5K →300K

w/ Sample Environment Randy Sexton and Robbie Marrs

New developments

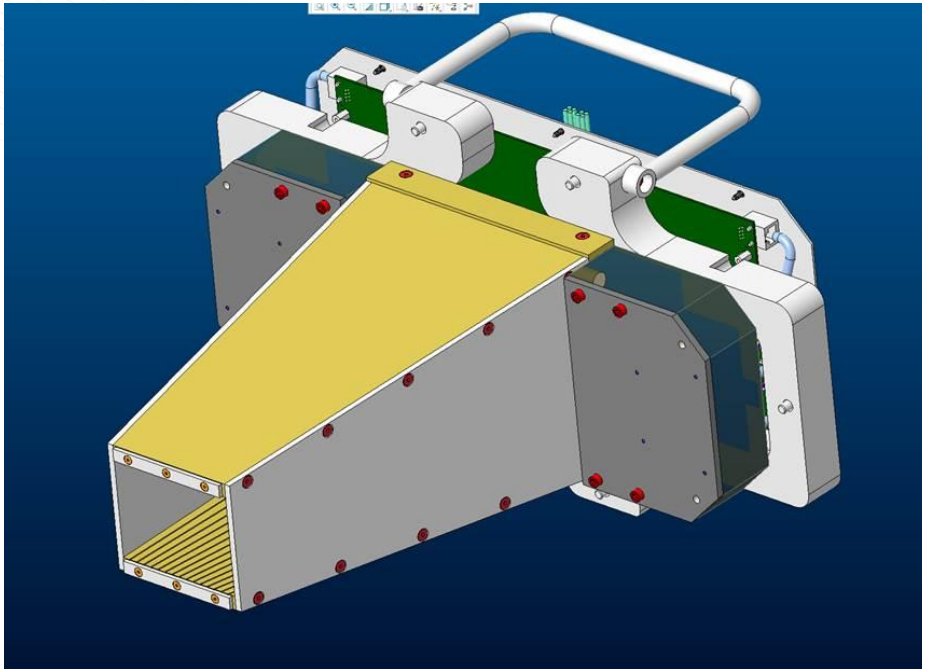
Mid-level funding was obtained in July 2019 for a
compact vacuum furnace for VISION MICAS design

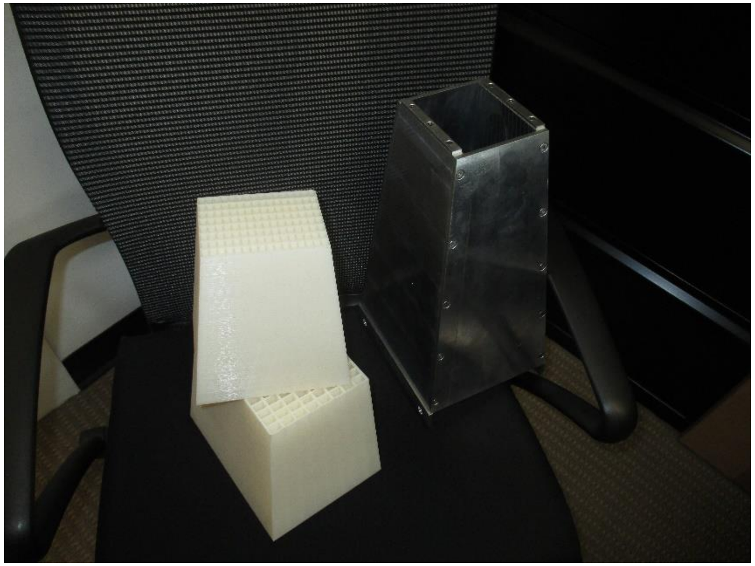


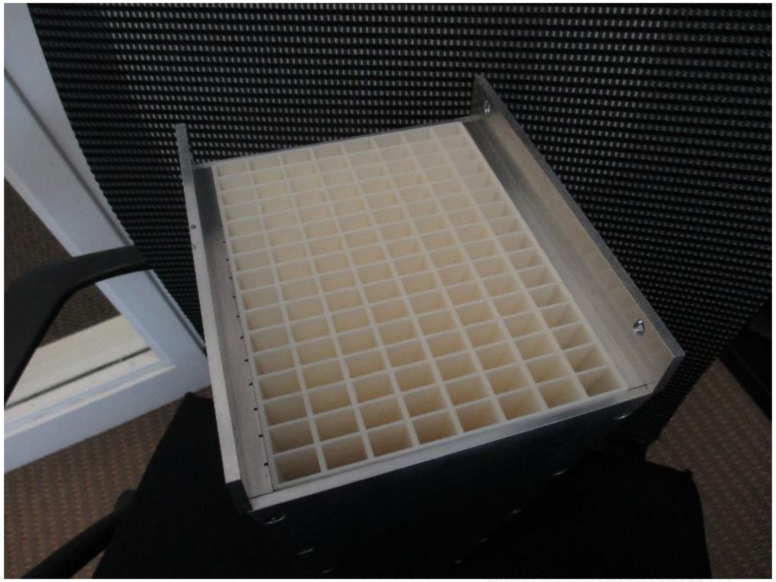
- 30^{o}C to 1000^{o}C
- Uniform heating; no T gradients
- Scaled-down version of existing MICAS design
- Compact design usable at other beam lines (e.g., at HFIR)
- Fabrication drawings ready (Aug-Oct 2019)
- All parts procured (Sept 2019-Jan 2020)
- Assembly delayed by COVID
- This furnace will support, e.g.,
  - Catalysis research
  - Molten Salt Reactor Project
  - Transformational Challenge Reactor (TCR)
  - Advanced glasses development (Corning)

New developments

In the summer of 2018, new 3D-printed collimators for the 90° diffraction detector were installed and tested.



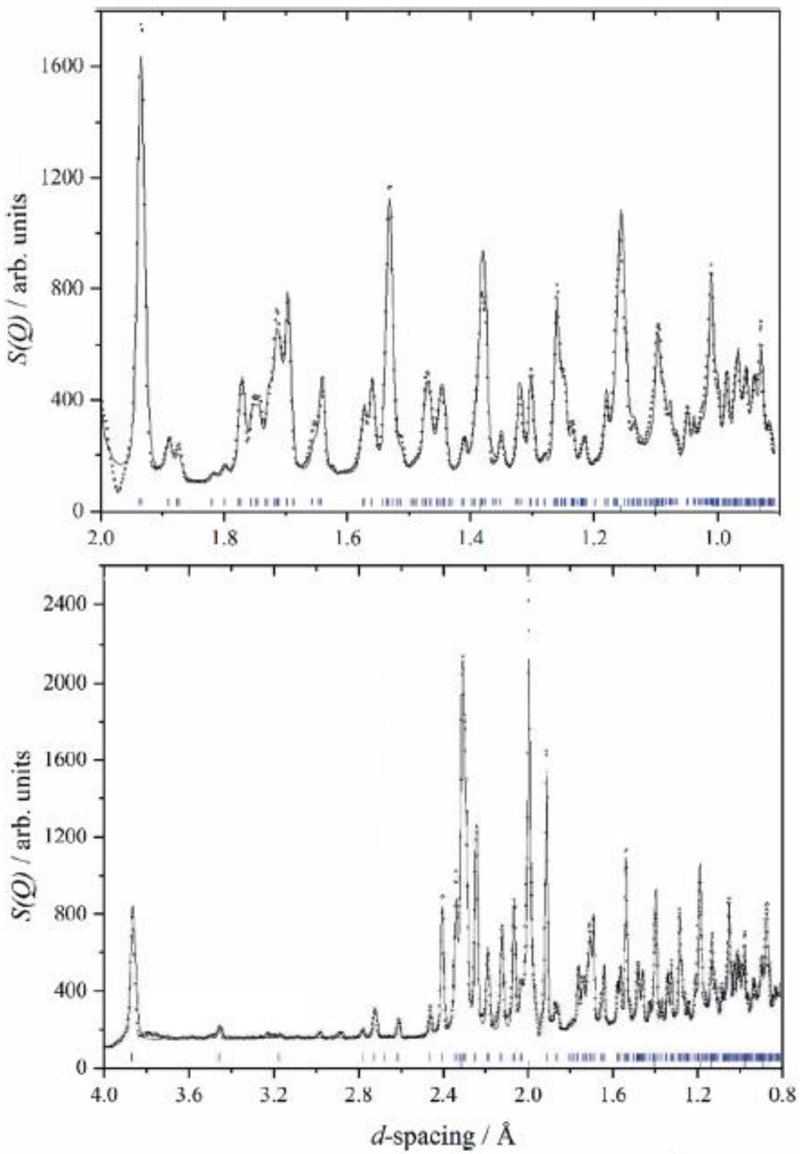




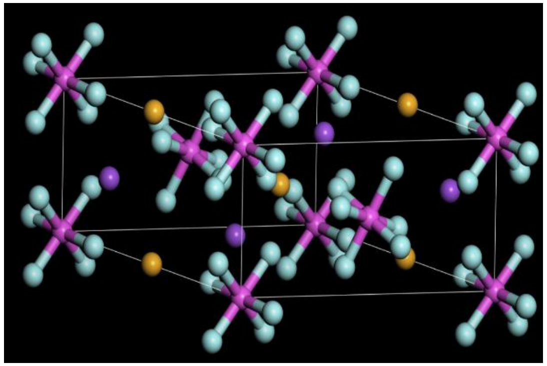
Automatic diffraction data reduction to multiple formats

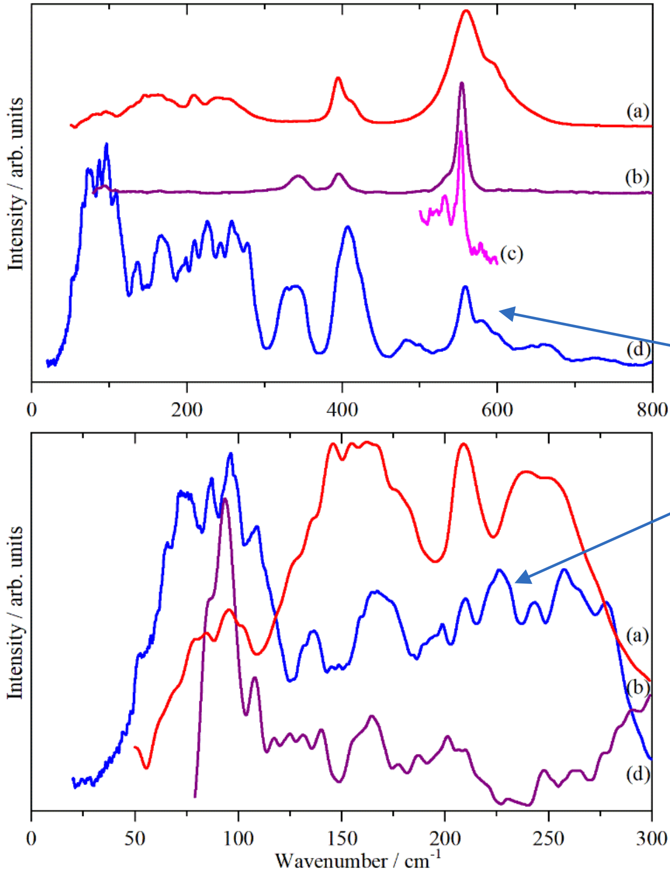
Diffraction data is now systematically collected with INS

simultaneous diffraction and ins



Na_3AlF_6, cryolite





VISION
INS

Parker et al., RSC ADVANCES, 10, 25856-25863 (2020)

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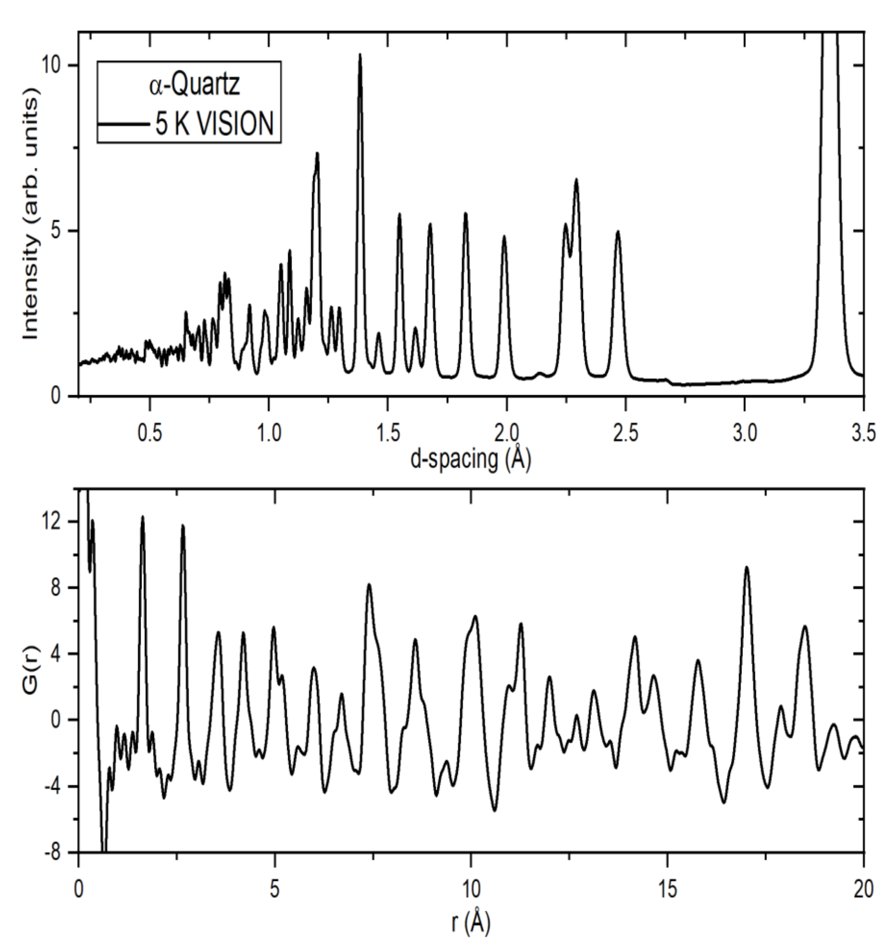
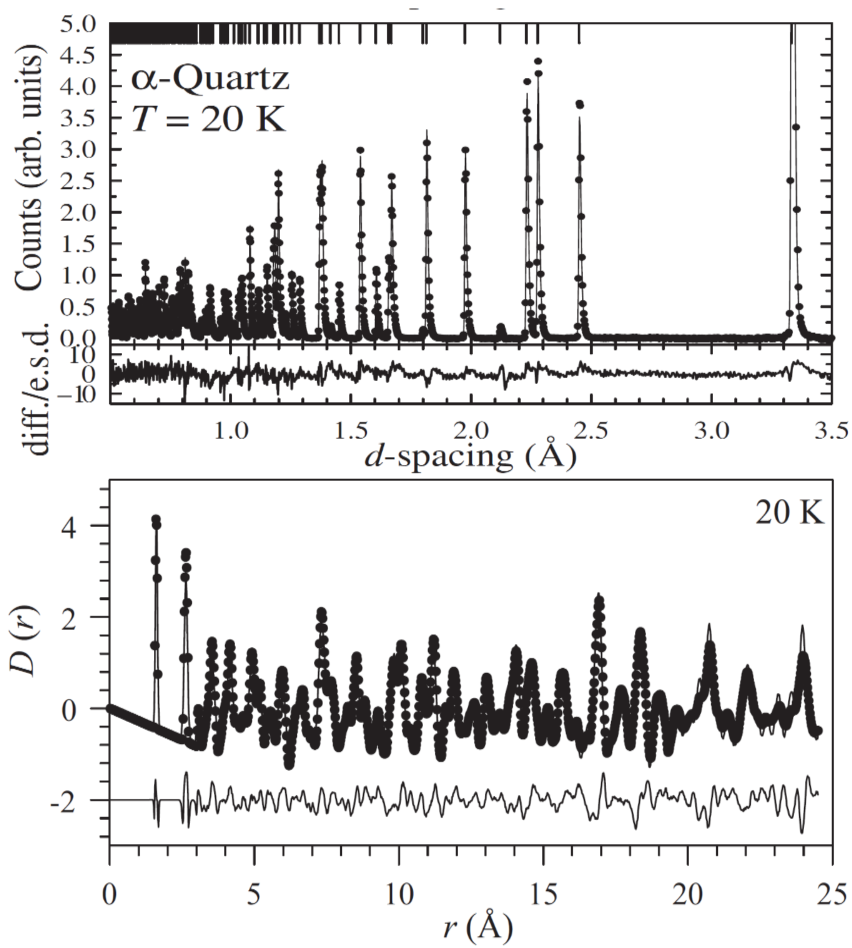
simultaneous pair distribution function and ins

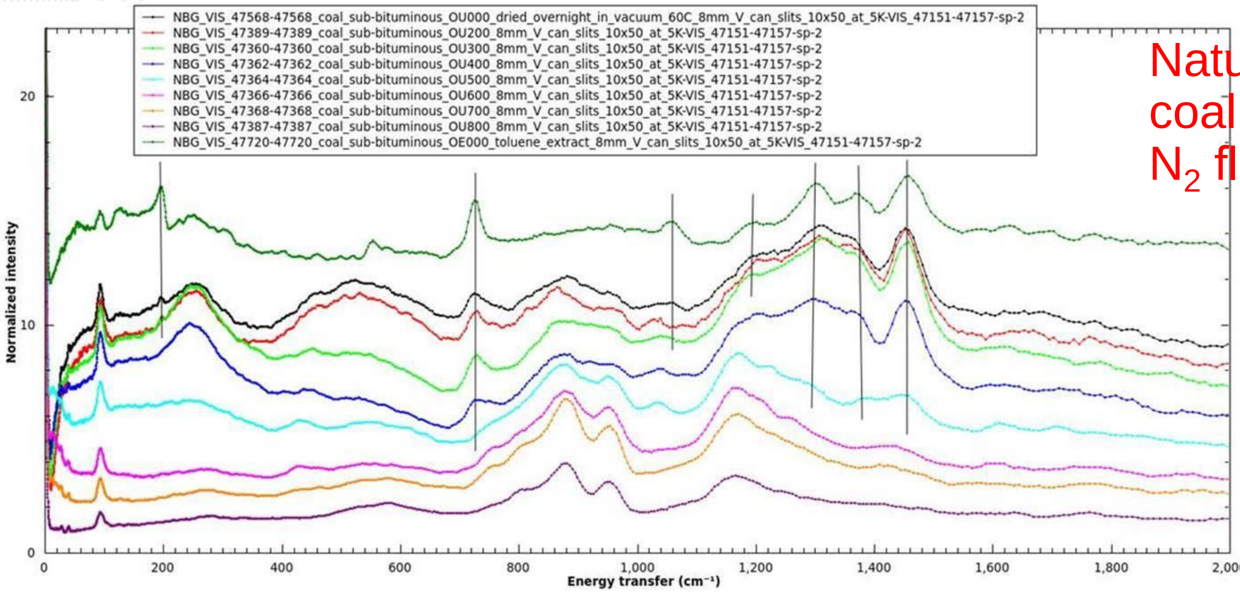


VISION (SNS)



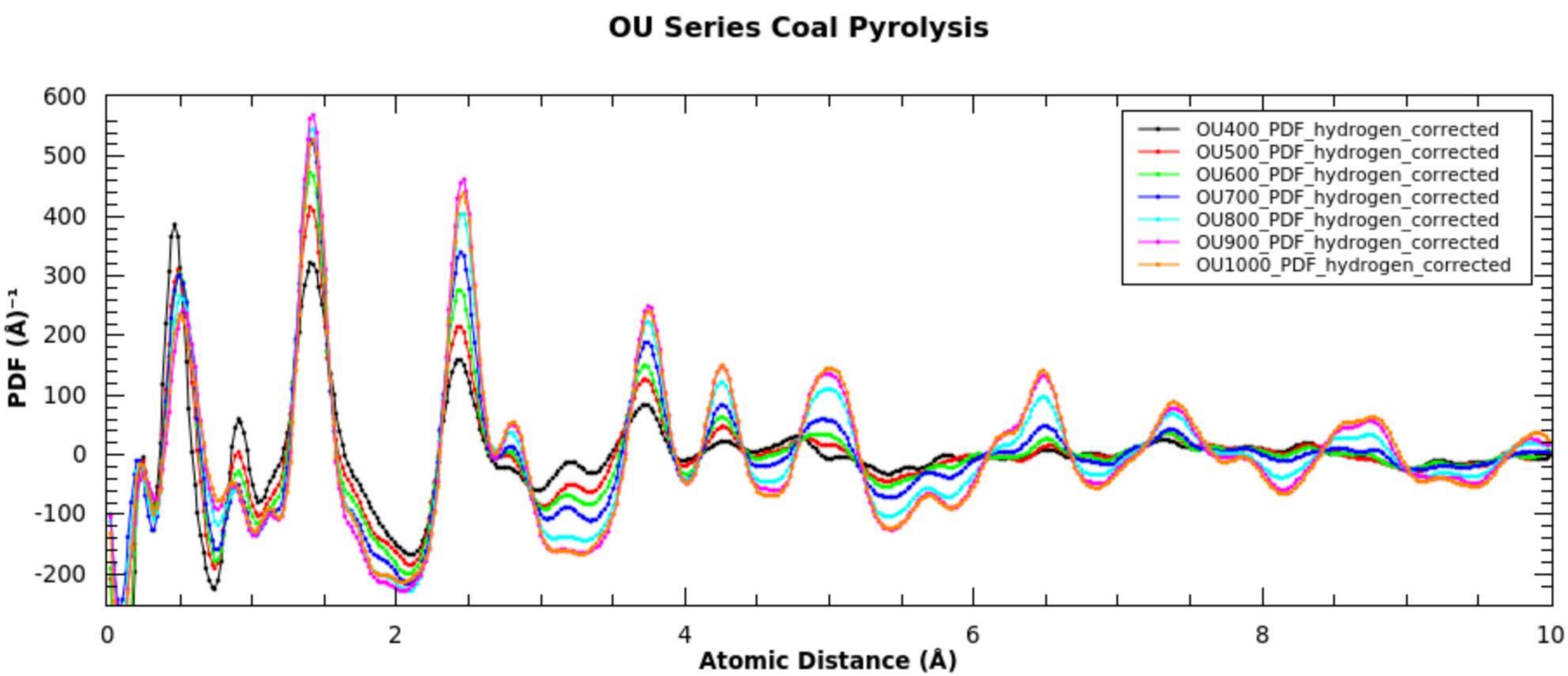
LAD (ISIS)

E. Novak, unpublished



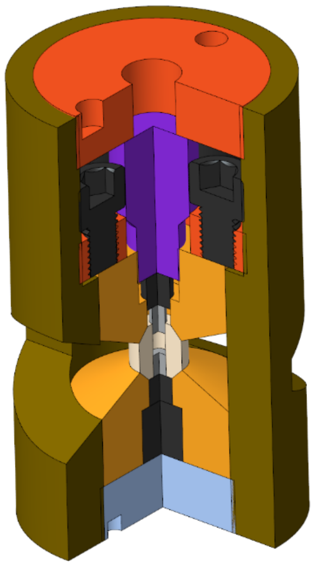
Natural product: sub-bituminous coal, pyrolyzed 10 mins under N_2 flow at T=400, …1000^{o}C

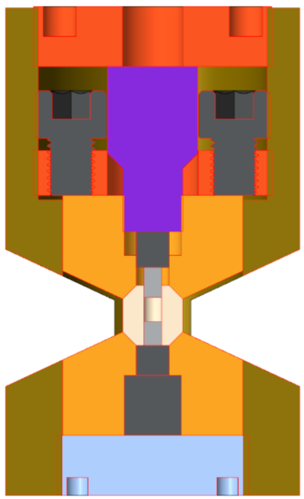
C. Tsouris, C. Finney
ORNL (NTRC)



New developments

High-pressure cells





New compact McWhan clamp cell design (R. Boehler, B. Haberl)

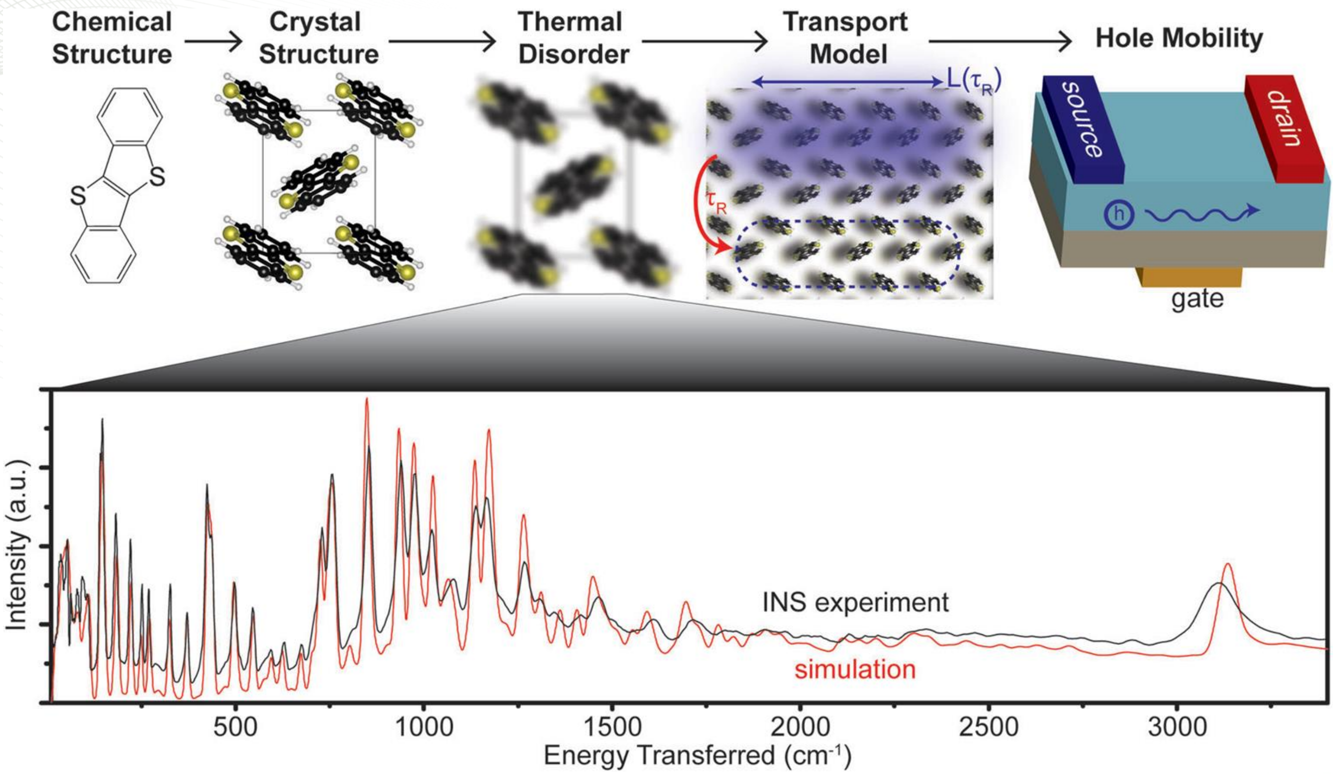
+ 35 mm^3
+ yttria-stabilized bicone
+ 10 Gpa at 7 tons loading

- New CuBe DACs with wide-angle opening for full illumination of analyzers and detectors.
- Versimax PCD is used for the anvils.
- Pmax = 30 GPa
- Sample volume is 1 mm^3

B. Haberl et al, **High Pressure Research 37**, 495 (2017) and B. Haberl et al, **Rev. Sci. Instr. 89** (9), 092902 (2018)

Science highlight

Nuclear modes limiting charge mobility in molecular semiconductors

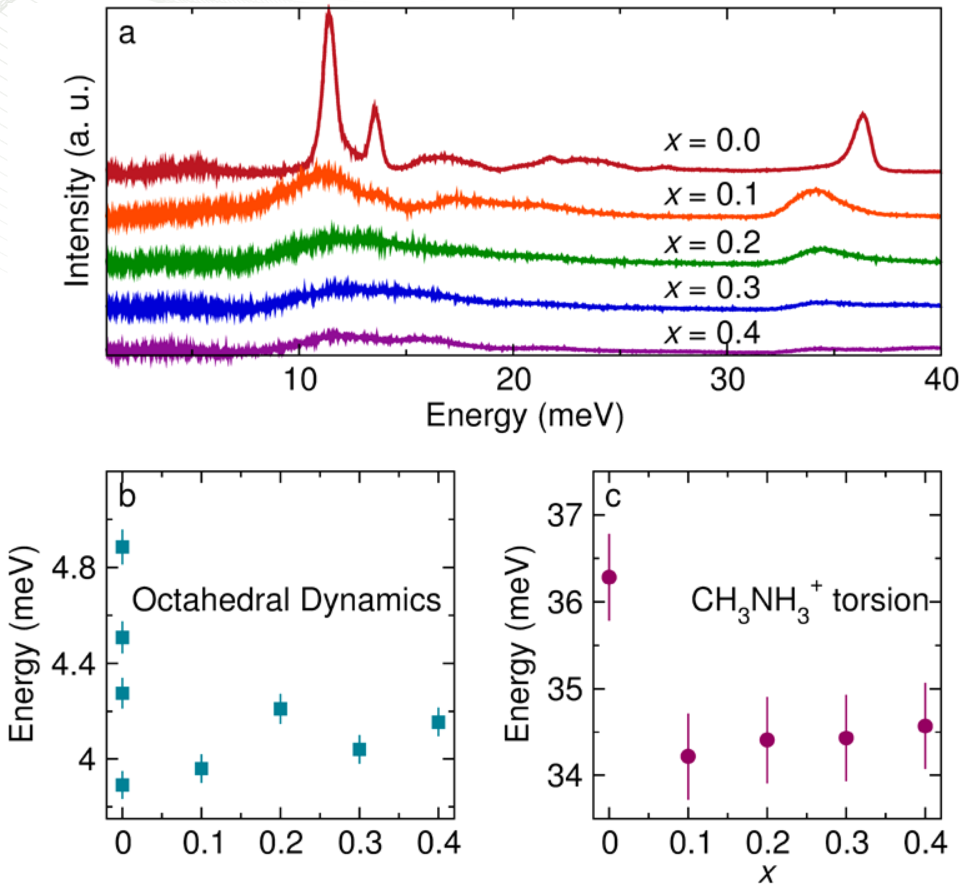


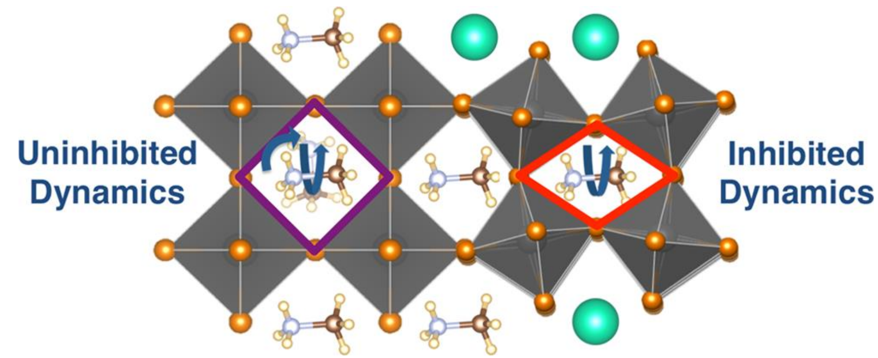
Schematic showing the connection between hole mobility, thermal disorder, crystal, and chemical structure in BTBT. The spectrum of motions leading to thermal disorder is measured using inelastic neutron scattering, which agrees with simulated modes over the full energy range measured (410–3500 cm^{-1}).

A. Moule et al., Materials Horizons, **6**, 182 (2019)

Science highlight

Orientational Glass Formation in Substituted Hybrid Perovskites $(CH_3NH_3)_{1-x}Cs_xPbBr_3$

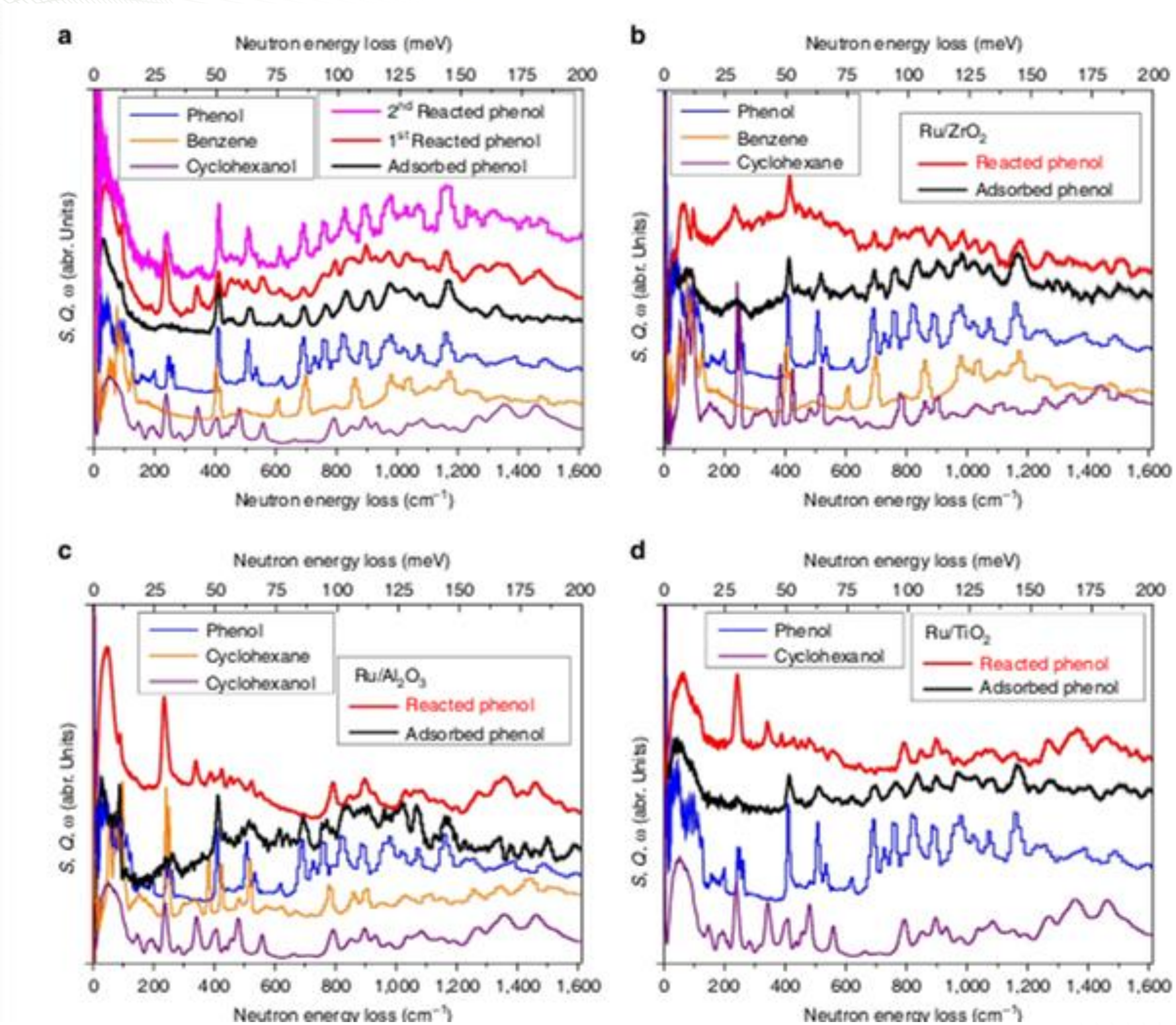




Reorientational dynamics of the methylammonium cation as a function of Cs doping as measured at VISION.

Science highlight

Production of arenes via direct lignin upgrading over a niobium-based catalyst



- Comparison of the INS spectra for the adsorbed and reacted phenol molecules on Ru/Nb_2O_5 (a), Ru/ZrO_2 (b), Ru/Al_2O_3 (c) and Ru/TiO_2 (d). The second reacted phenol curve in panel (a) shows the conversion of phenol to benzene and cyclohexanol, which does not occur on other supports in panels (b)-(d).

Summary

- Multiple improvements and additions: background reduction, chopper vibration, fast CCR heater, diffraction detector collimation, new CuBe DACs and new compact, high volume McWhan high pressure cell
- Reliable diffraction data, Rietveld refinements, d-spacing range of 0.2 – 5.5 Angstrom. Diffraction data reduction automated.
- Pair Distribution Function simultaneously with INS. Data processing methodology under development. Validation by comparison with other instruments and data sets done.
- New science areas: organic semiconductors, polymers, small pharmaceuticals, natural materials, biochemistry. Catalysis still going strong (30% of the beam time).
- Funding for new CCR (larger bore, faster cooling) secured.
- New Scientific Associate (Eric Novak) – started in March 2020.