

Understanding Soot Formation, Graphitization, and Oxidation

• Science Challenge/Opportunity

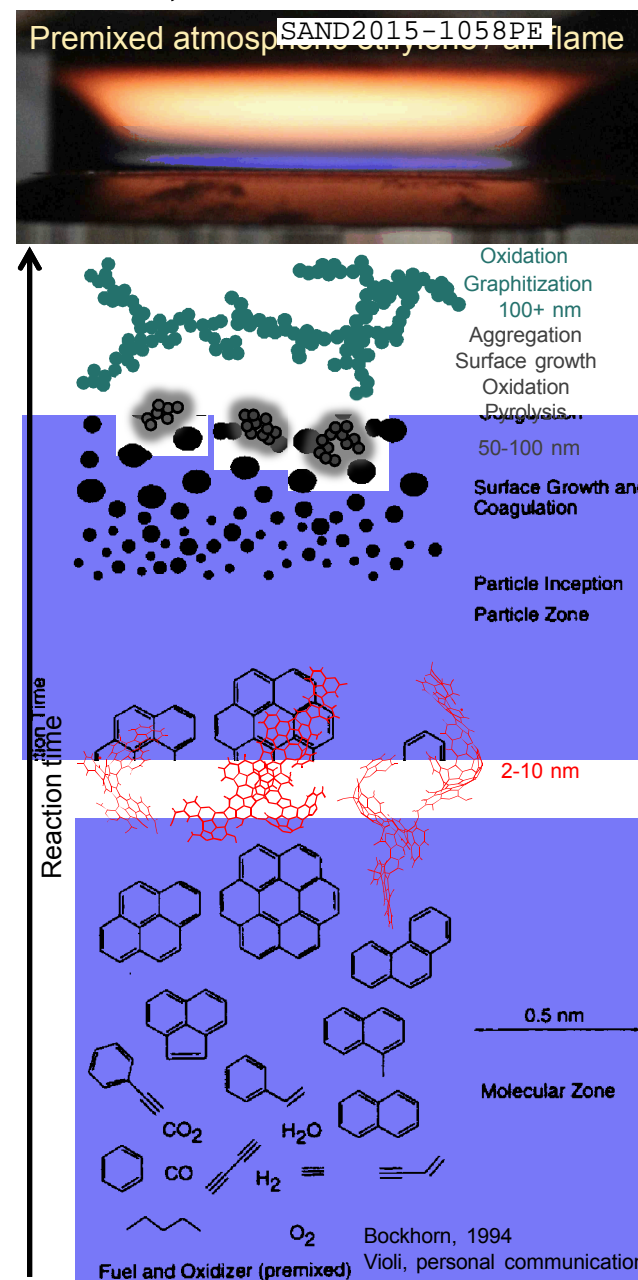
- Soot chemistry is not well understood.
- There is limited experimental data to constrain models.
- Extractive sampling is often used for particle composition, size, morphology, and fine structure.
- Extractive sampling perturbs combustion conditions.
- Need *in situ* diagnostics for composition, size, morphology, and fine structure.

• Significance & Impact

- Soot has significant negative impact on human health, agriculture, air quality, and global climate.
- Mitigation requires understanding formation and destruction.
- *In situ* measurements of composition, size, morphology, fine structure enables model development needed for mitigation.

• LCLS-II Strengths & Challenges

- High photon flux, short pulse duration enables *in situ* X-ray diffraction to measure morphology from single particles
- Narrow-band, coherent, high photon-flux pulses enable *in situ* X-ray coherent anti-Stokes Raman spectroscopy for soot / flame composition



Experimental Approach

• Techniques

- Soot particle size and morphology – *in situ* (in flame) XRD
- Soot / flame composition - *in situ* (in flame) XCARS

• Tools

- XRD: Fast, imaging detector with high spatial resolution
- XCARS: 2-color, 1 tunable, separated by ~280 eV
- Hard X-rays to allow probing of combustion at pressures \geq atmospheric

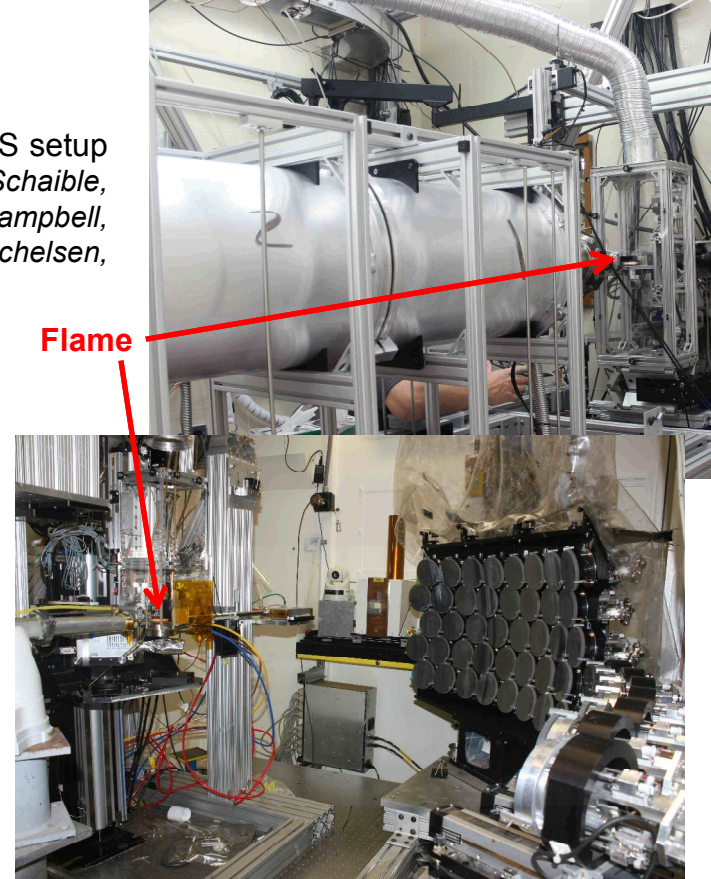
• Alternatives

- SAXS / WAXS
- Visible laser elastic scattering
- Spontaneous X-ray Raman spectroscopy
- Extractive sampling and analysis, e.g., NEXAFS

ALS SAXS setup

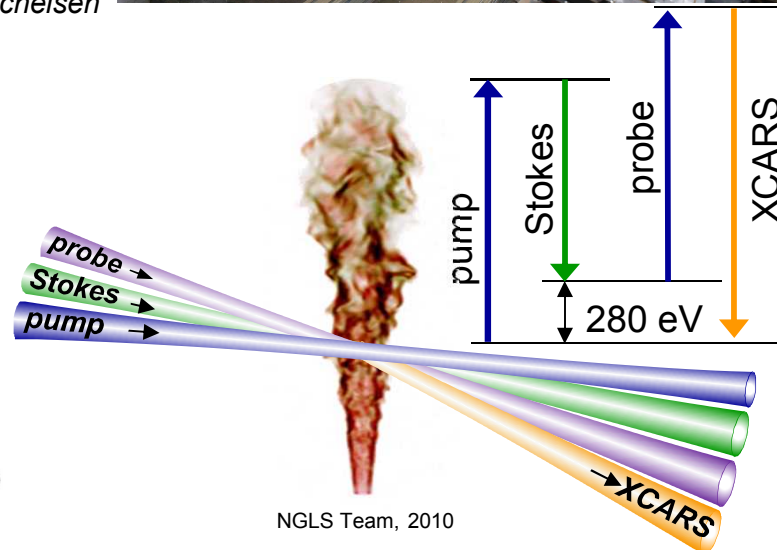
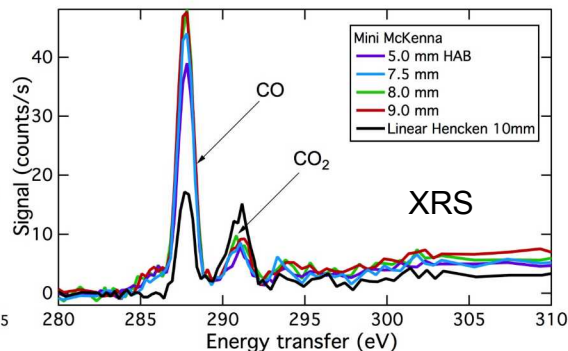
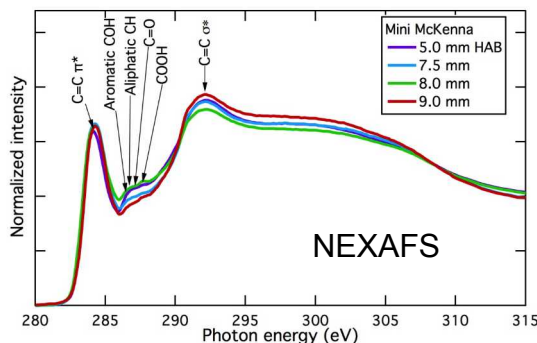
Tran, Van Buuren, Willey, Schaible,
Zhu, Stewart, Hexemer, Campbell,
Johansson, Schrader, Michelsen,

Flame



SSRL XRS setup

Tran, Van Buuren, Willey, Weng,
Nordlund, Sokaras, Campbell,
Johansson, Schrader, Bambha,
Rieken, Michelsen



Hope Michelsen, Tony Van Buuren, & Team

Measurement Parameters (fill in the key parameters)

Parameter	Units	Range	Comment
X-ray Photon Energy (wavelength)	keV		
X-ray Pulse Repetition Rate	kHz		
X-ray Photons per pulse (Energy)	N or mJ		
X-ray Pulse Bandwidth (FWHM)	meV		
X-ray Polarization (vert., horz., circ.)			
Pulse length (FWHM)	fs		
X-ray pump / X-ray probe delay	fs		2- color
X-ray Focal Spot Size	μm		
Optical Pump Laser Synchronization	fs		
Optical Pump Laser Rep Rate	kHz		
Optical Pump Laser Energy per pulse	mJ		
Optical Pump Laser Wavelength	nm		
Area Detector Readout Rate			
Area Detector pixel size	(μm)		
Area Detector Array Size (in pixels)	NxM		