Soot Formation and Oxidation

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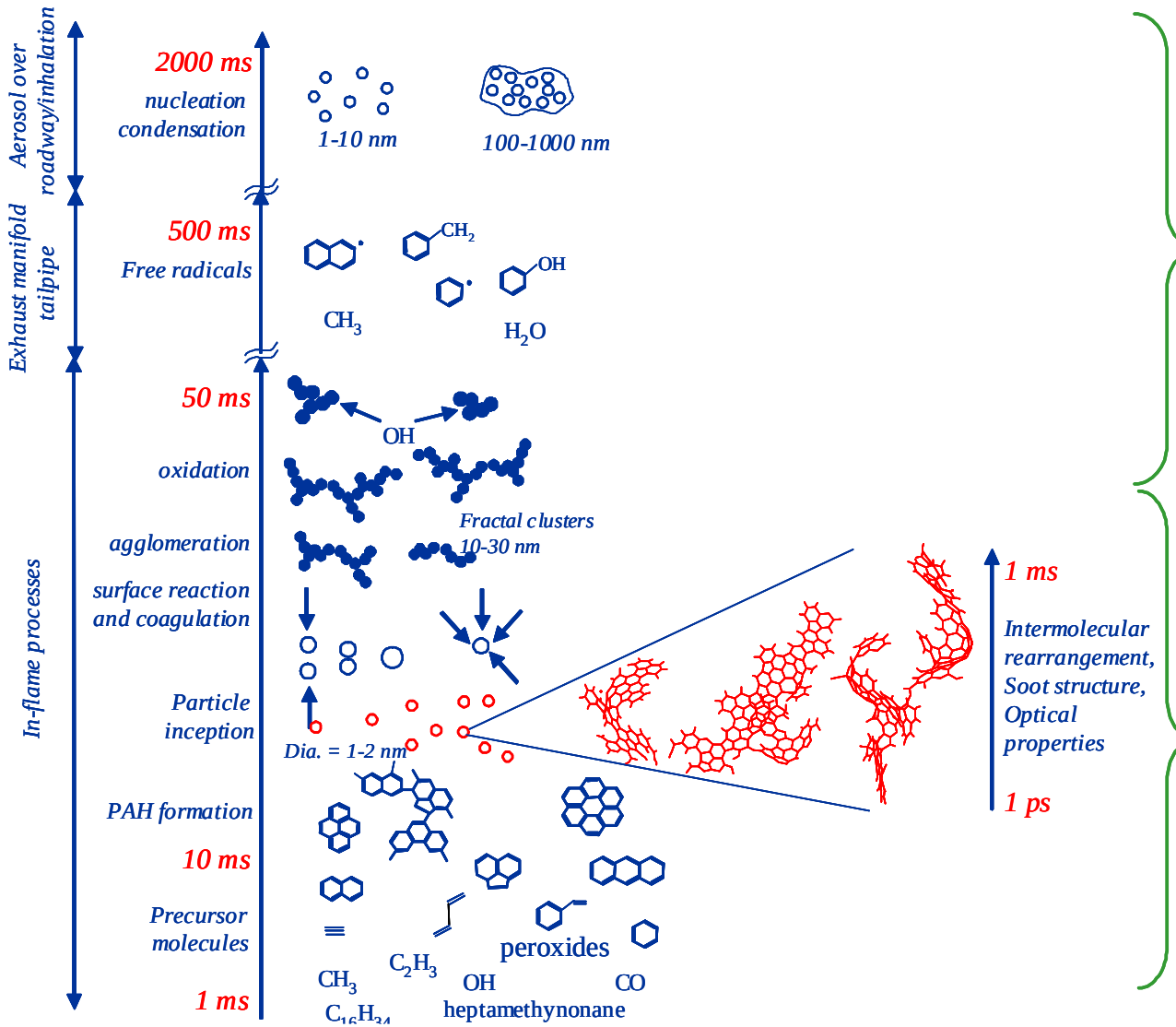
Objectives



Two-stage burner

NMR, SMPS

Inverse Diffusion Flame; Flat Flames

KMC/MD modeling

NMR, AFM, SMPS

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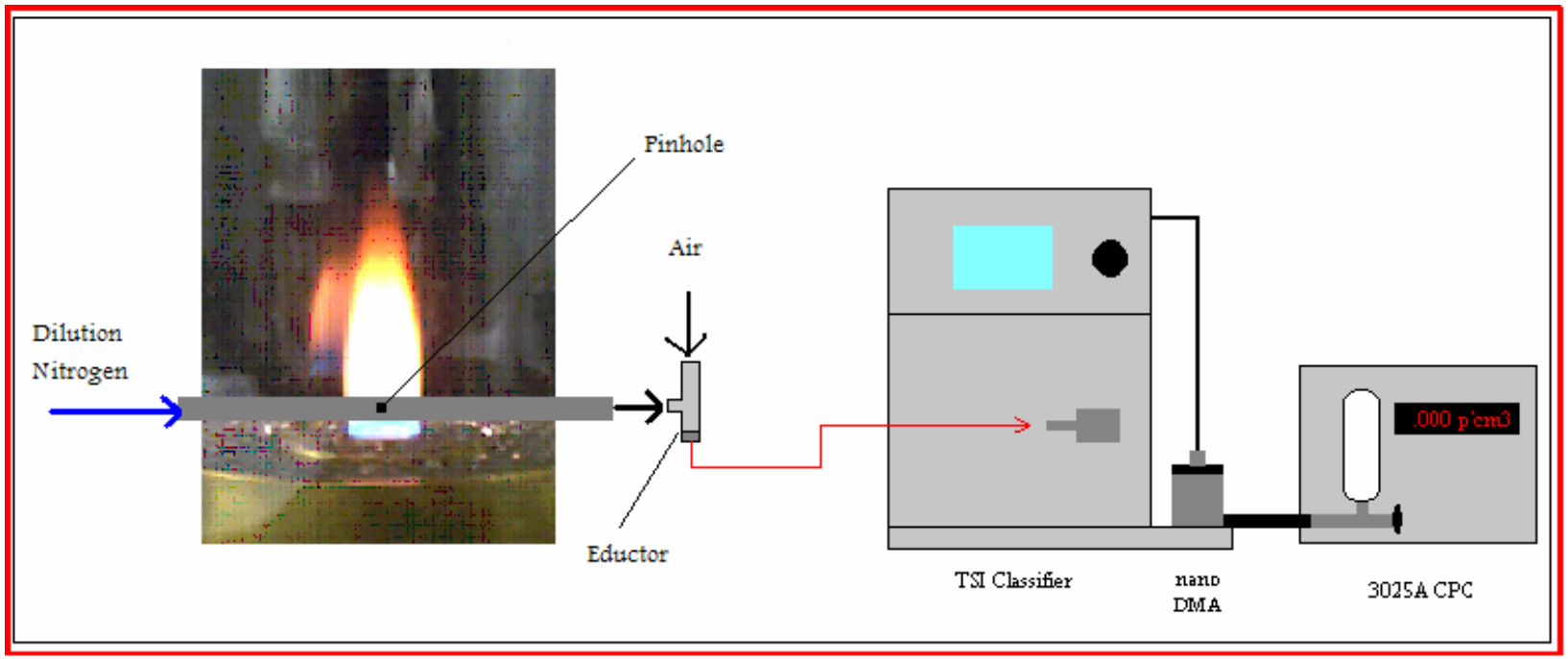
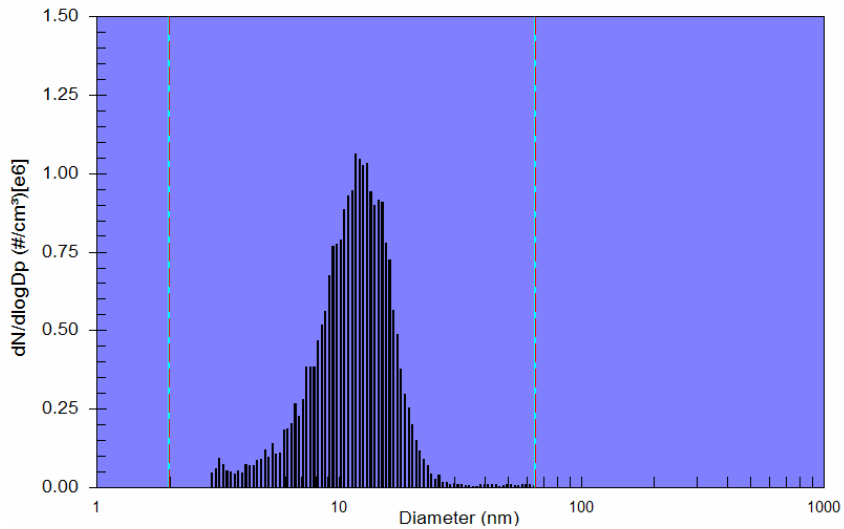
Experimental Systems

- Scanning Mobility Particle Sizer (SMPS)
  - Long-DMA
  - Nano-DMA
- Dilution System
  - Series of eductors
- Thermophoretic Sampling System
  - Rapid insertion device
  - TEM grid analysis
- Thermocouple Particle Densitometry
- Burner Systems
  - Inverse Diffusion Flame
  - Two-Stage Burner

SMPS





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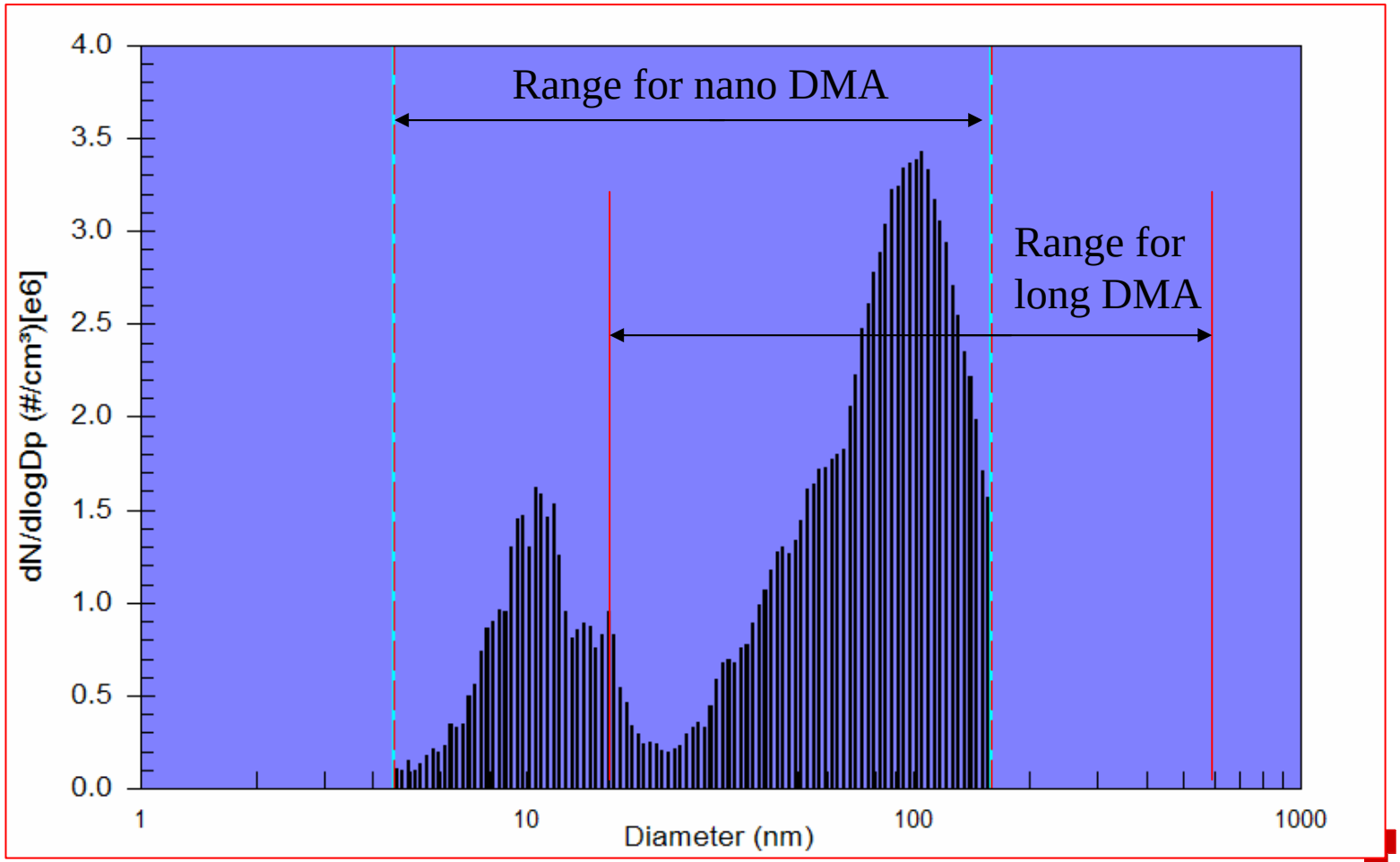
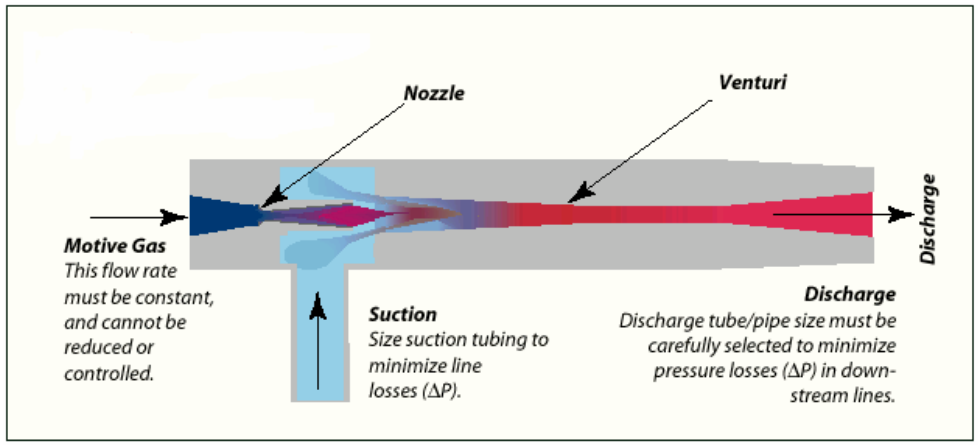
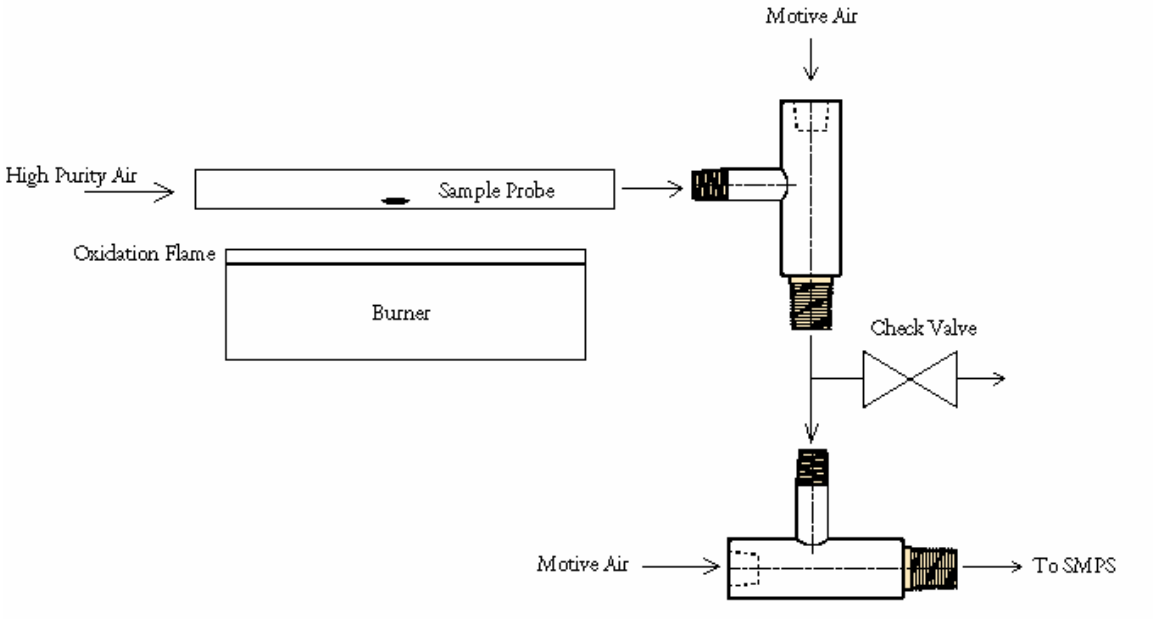
SMPS
Nano Results

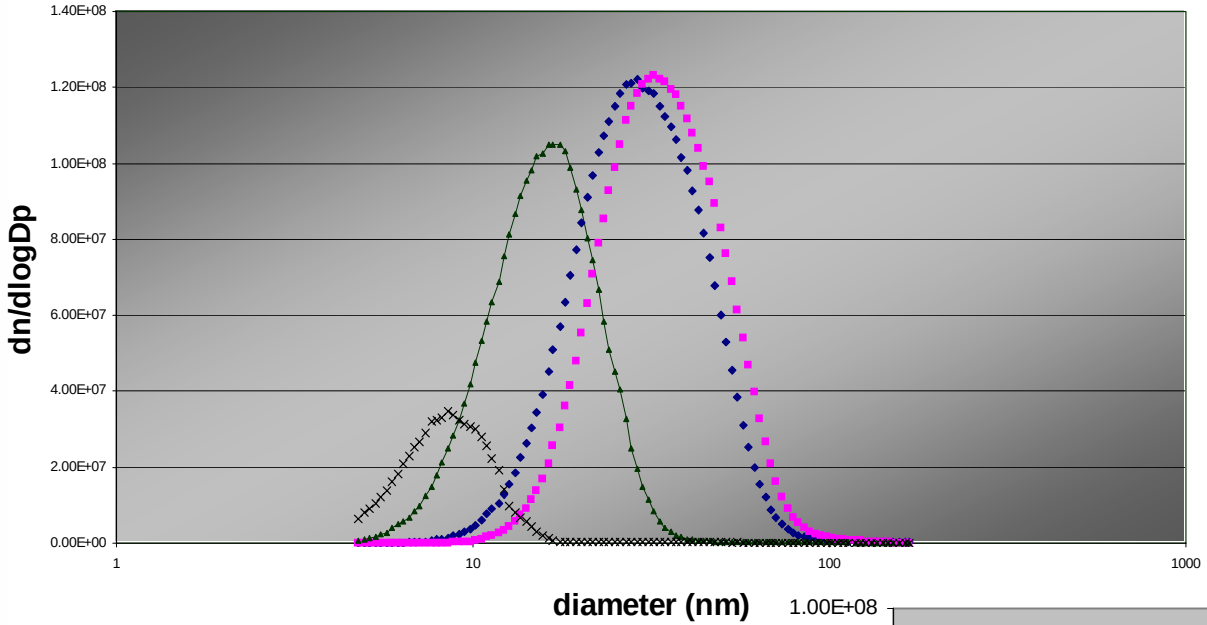


Dilution System



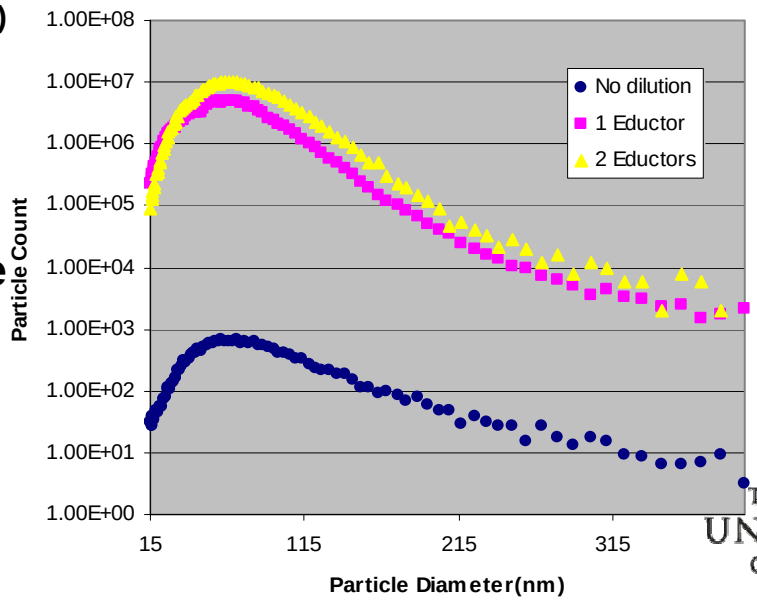


Importance of Dilution on PSD



Data are not correct for dilution but show a shift in diameter

Data show an increase in particle number with dilution increase; diameter changes are difficult to determine on the scale



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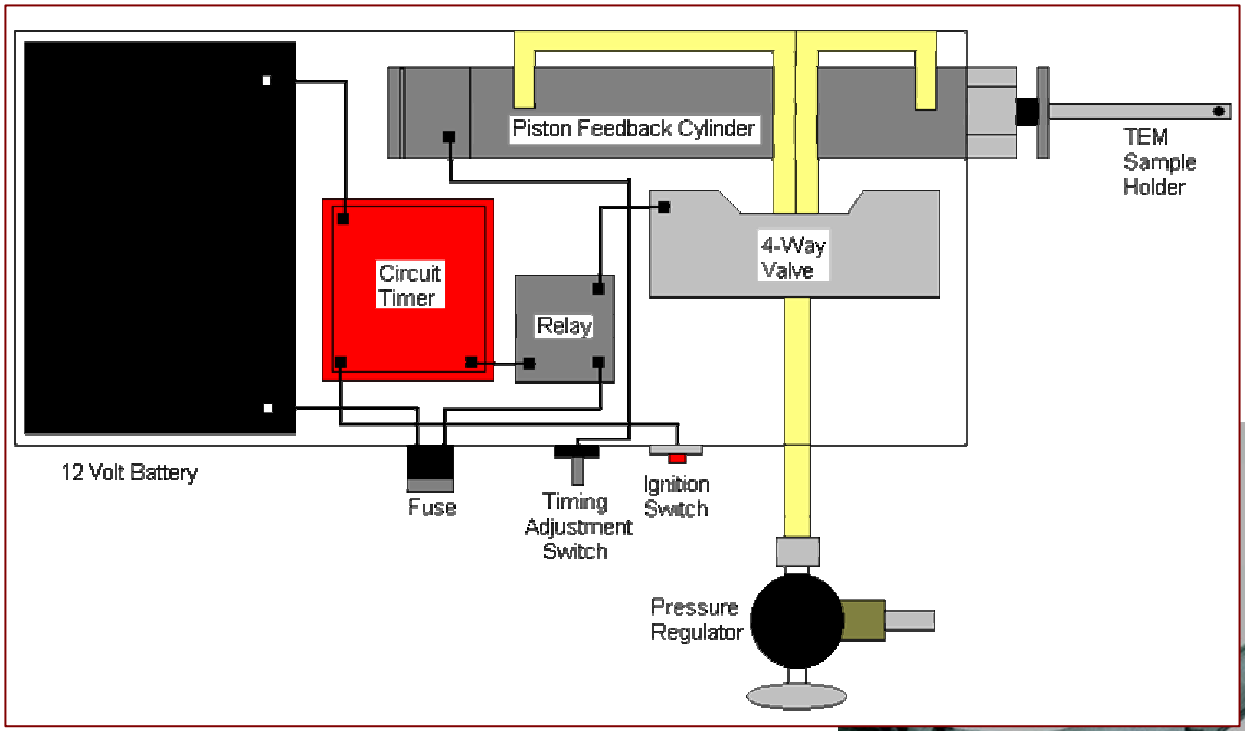
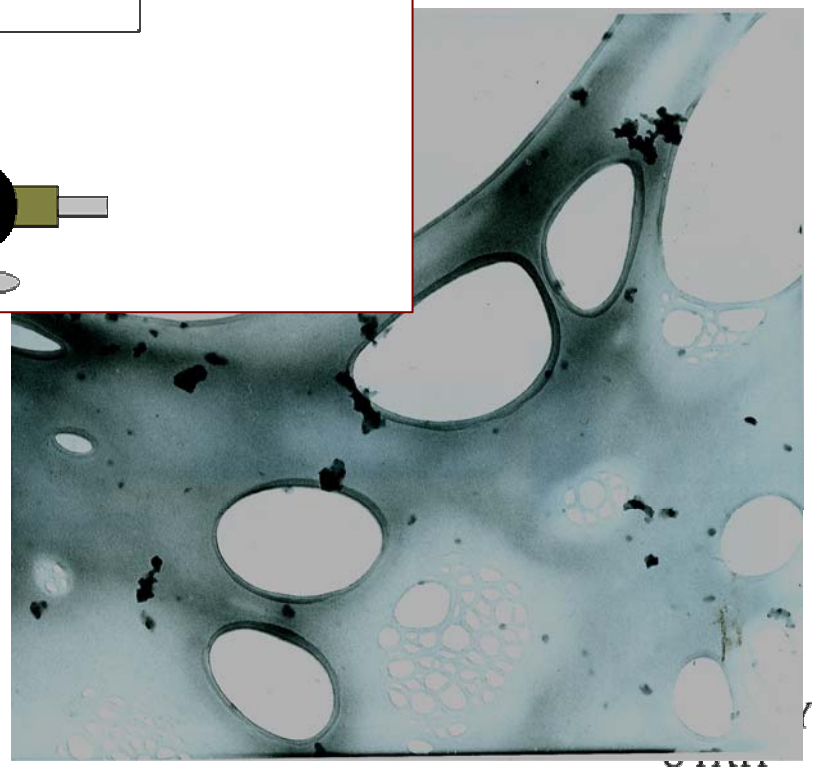
Thermophoretic Sampling





Thermophoretic Sampling provides TEM samples which allow for determination of soot morphology.

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Soot Formation

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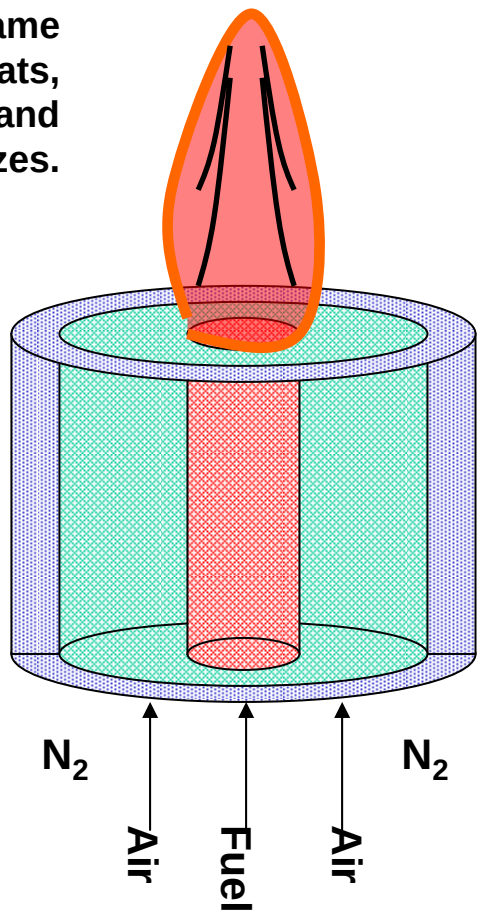
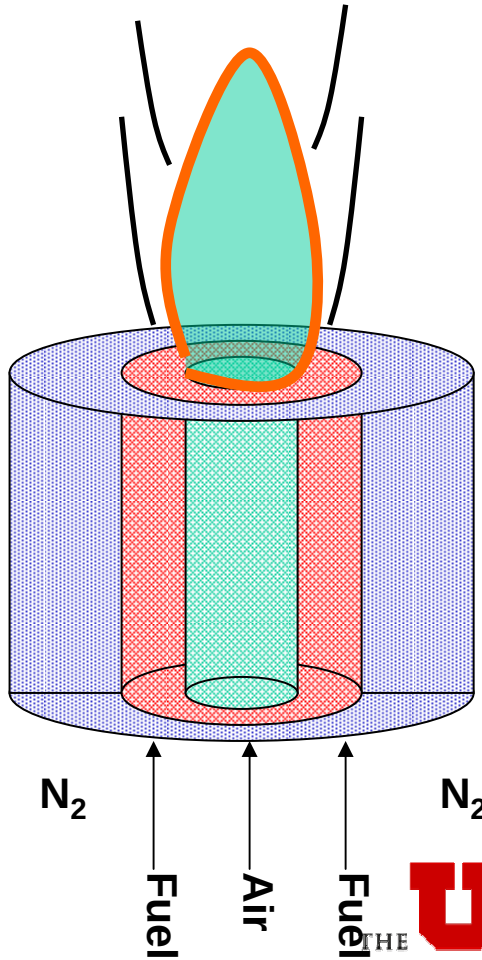
Soot Formation
Inverse Diffusion Flame (IDF)

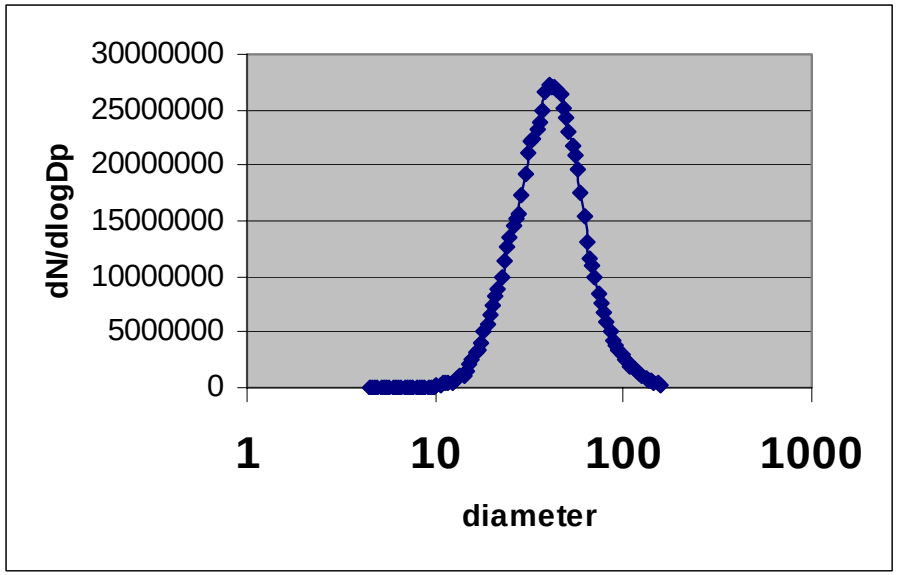
Normal Diffusion Flame: Soot passes through the flame and heats, carbonizes, and oxidizes.



Inverse Diffusion Flame: Soot is driven away from the flame into cool fuel/N_2: no carbonization or oxidation.

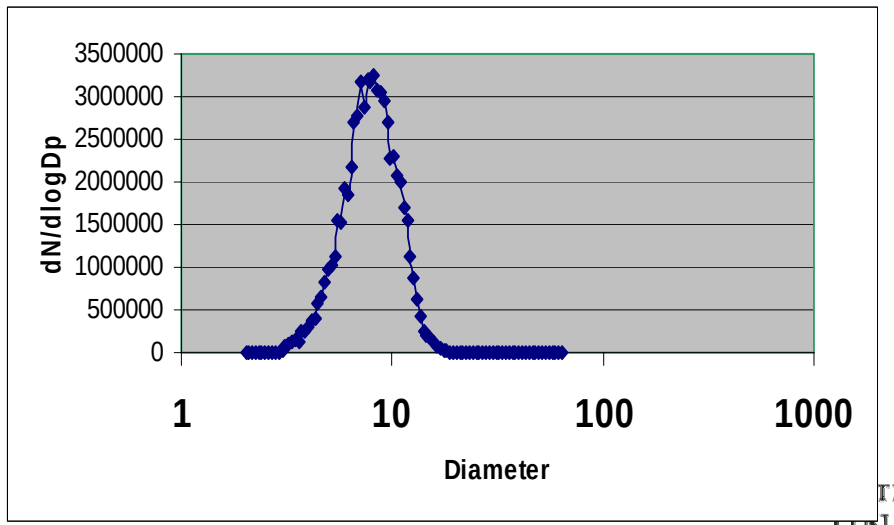


PSD from IDF

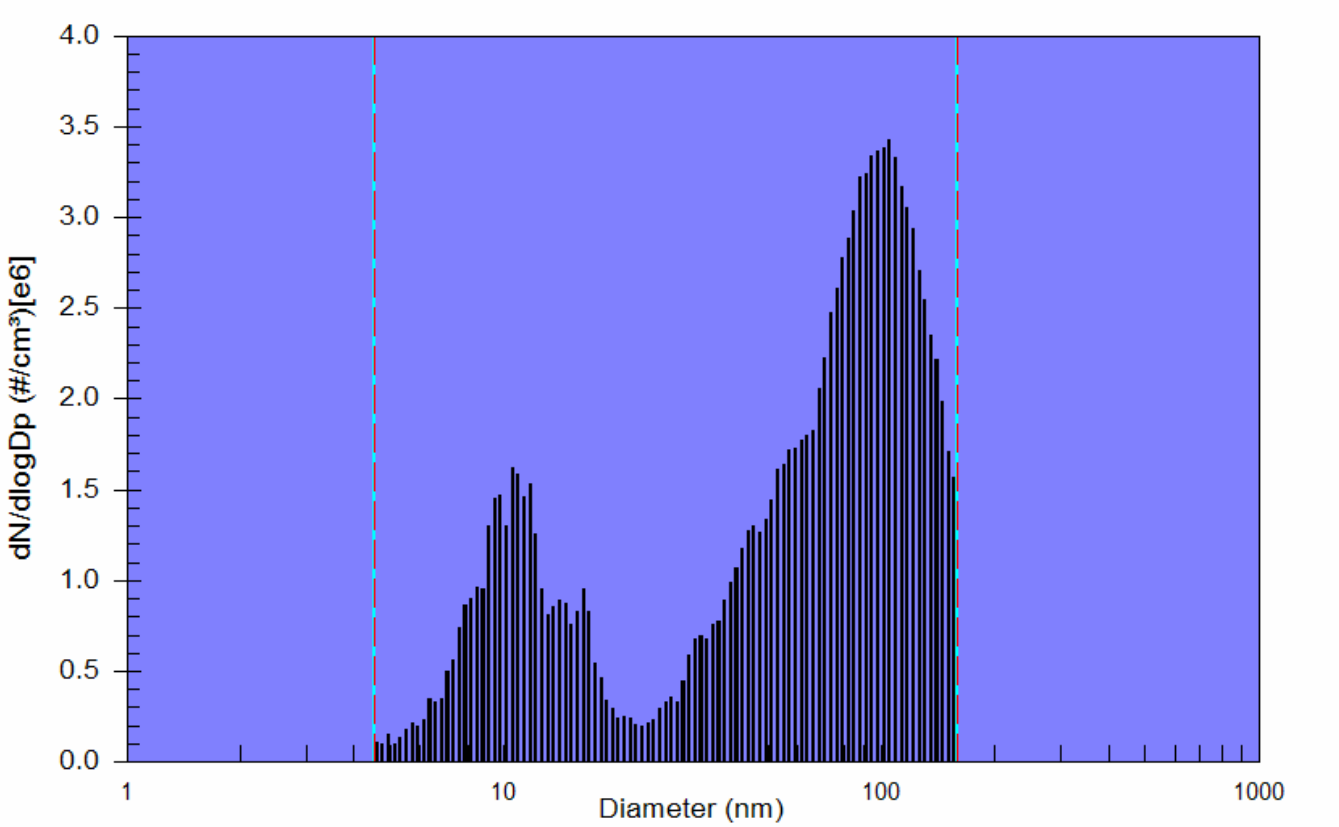


Peak at about 50nm for a premixed ethylene flame.

Peak at 9 nm for an ethylene inverse diffusion flame. IDF provides much younger and smaller soot.



PSD from IDF

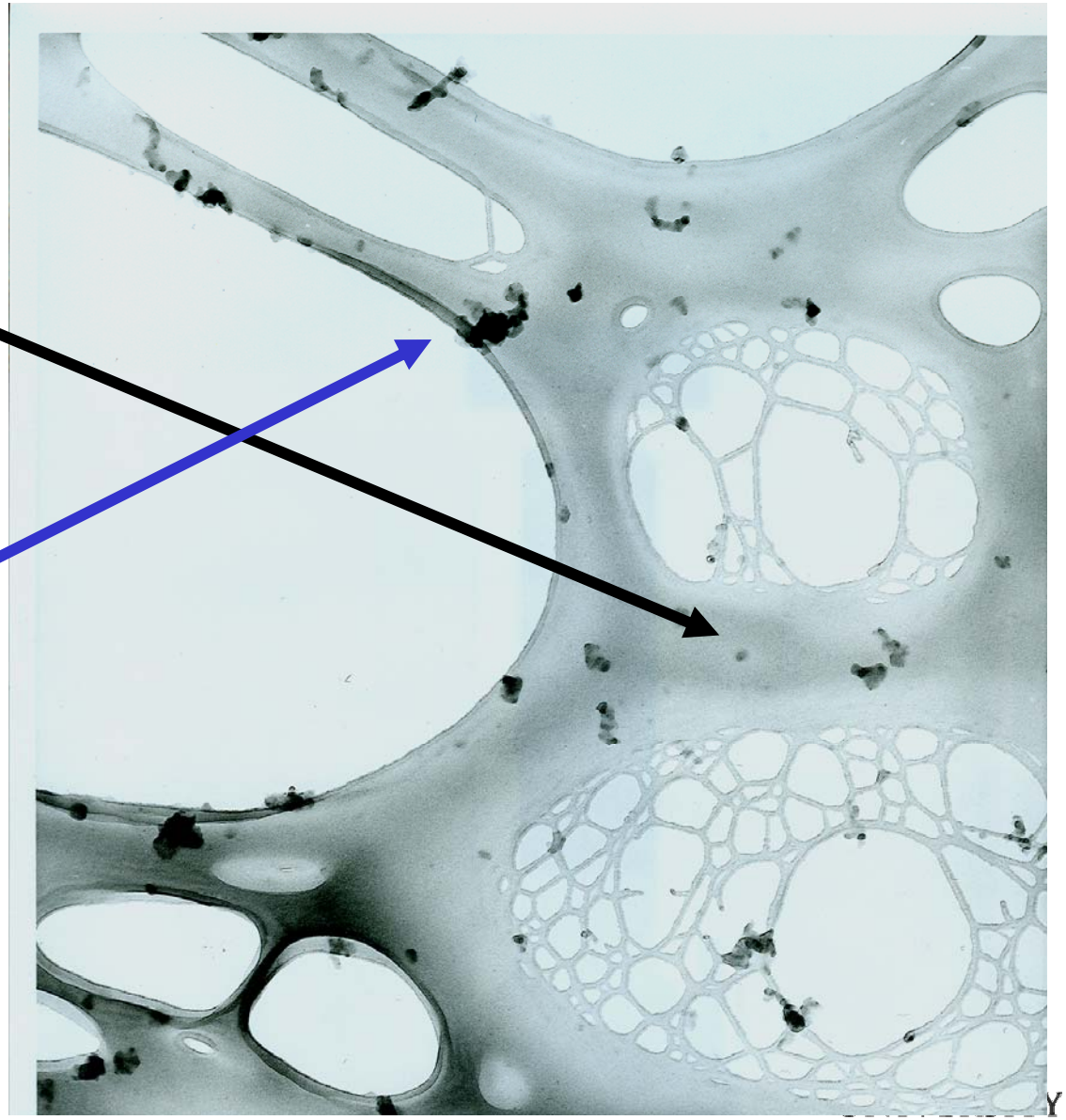


Above the flame, there appears to be a bimodal distribution. The primary particle size is still near 10 nm and the coagulation peak is about 90 nm.

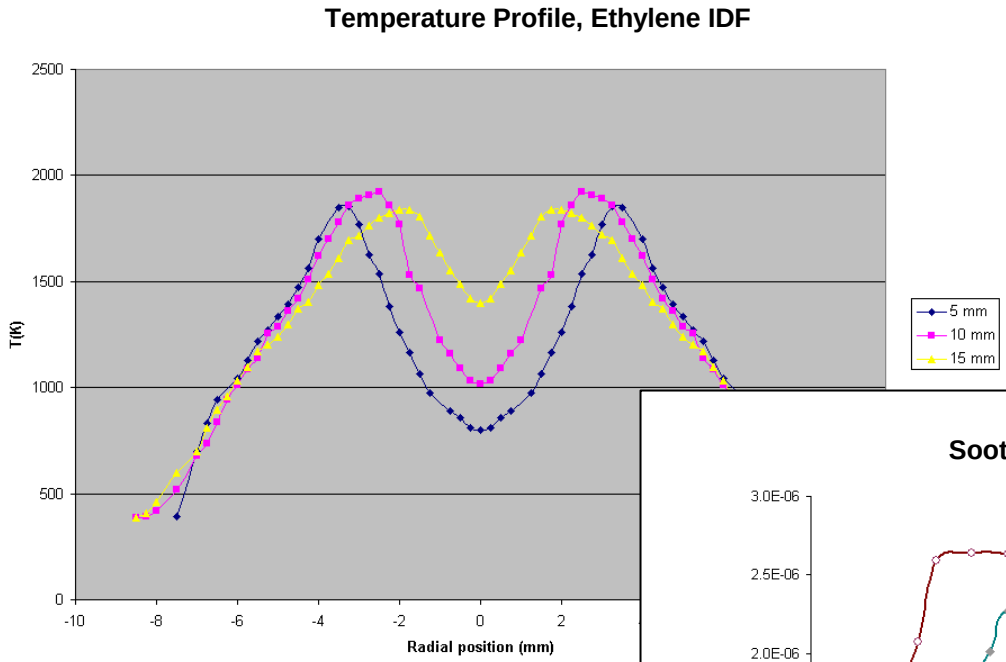
TEM from IDF

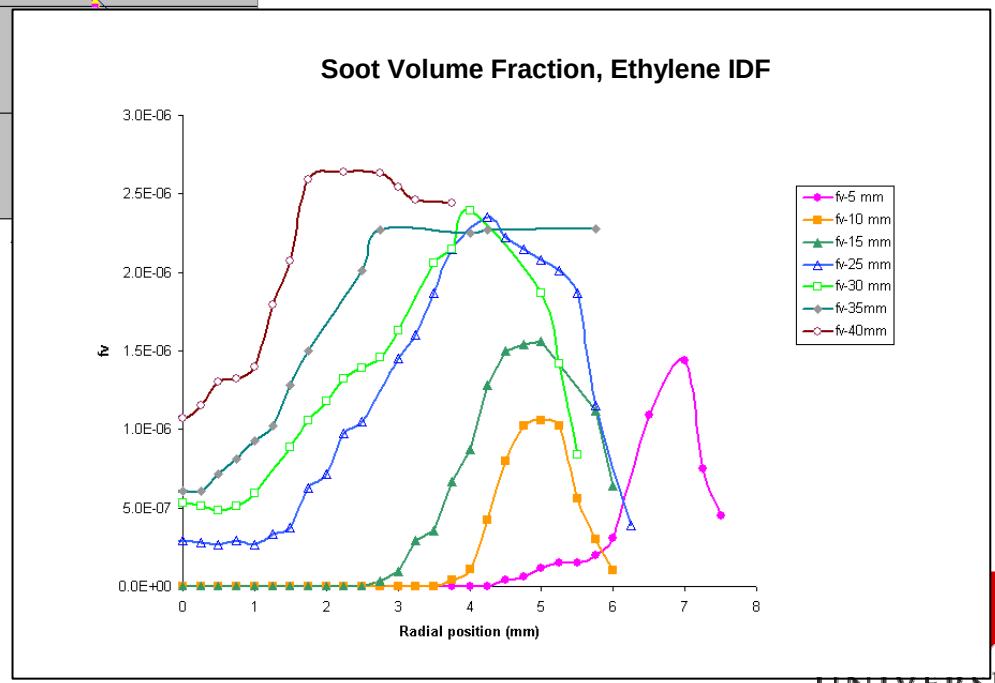
The majority of the soot particles are small, young soot as viewed by the small, spherical particles.

A few larger soot agglomerates can also be seen.



Temperature and Soot Volume Fraction
IDF Flame





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Soot Oxidation

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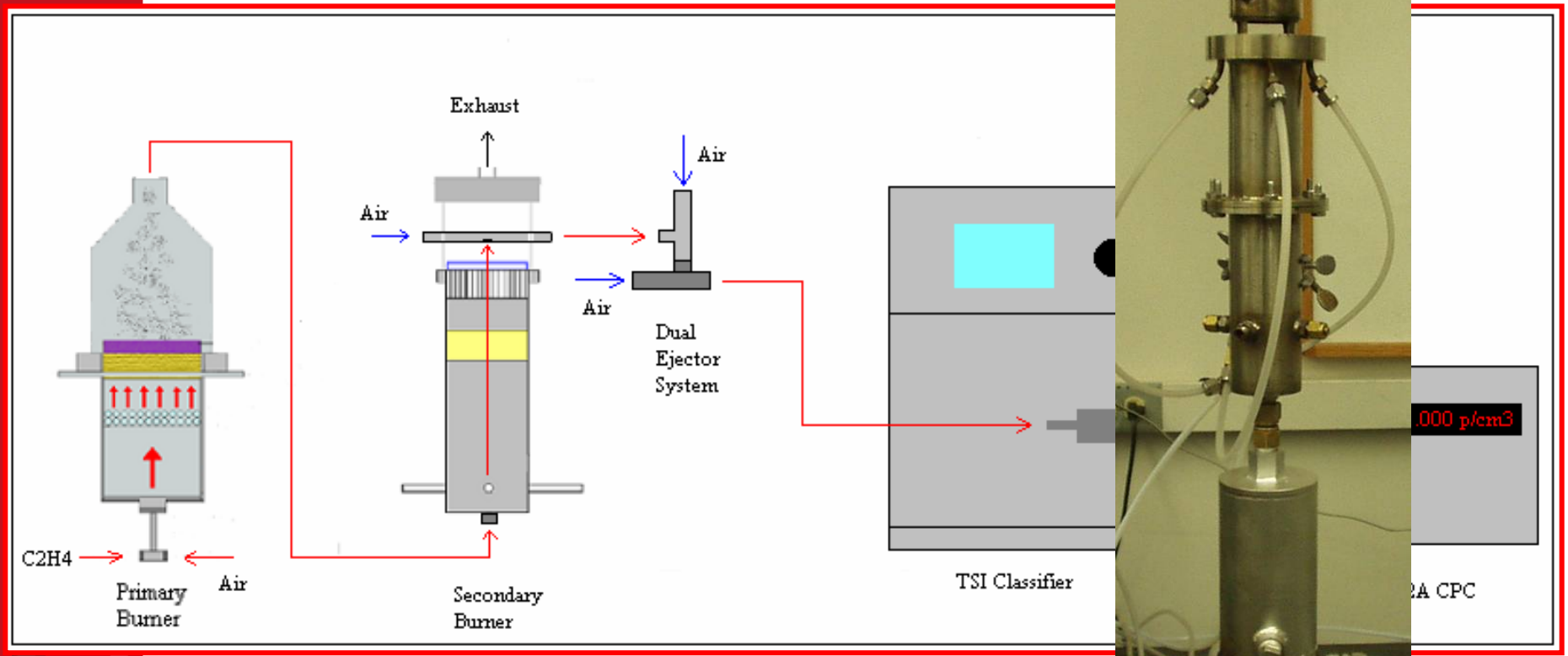
Soot Oxidation
Two-Stage Burner



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Oxidation Kinetics

OH Oxidation

-Neoh Hydroxyl Oxidation Formula

$$r = 1.2731x10^{-2}\,\Gamma_{OH}\,P_{OH}T^{-1/2}$$

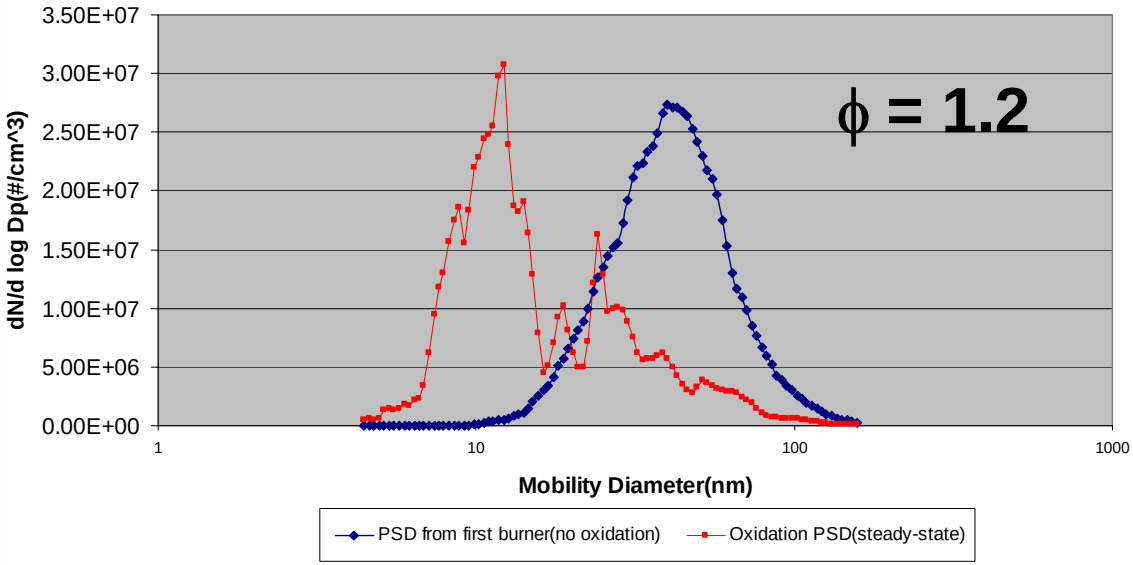
Γ_{OH} -OH collision efficiency

O_2 oxidation

-Nagle and Strickland-Constable Formula (NSC)

$$r = 120\frac{k_a P_{O_2}}{1+k_z P_{O_2}}x + k_b P_{O_2}(1-x)\left[\frac{kg}{m^2 s}\right]$$

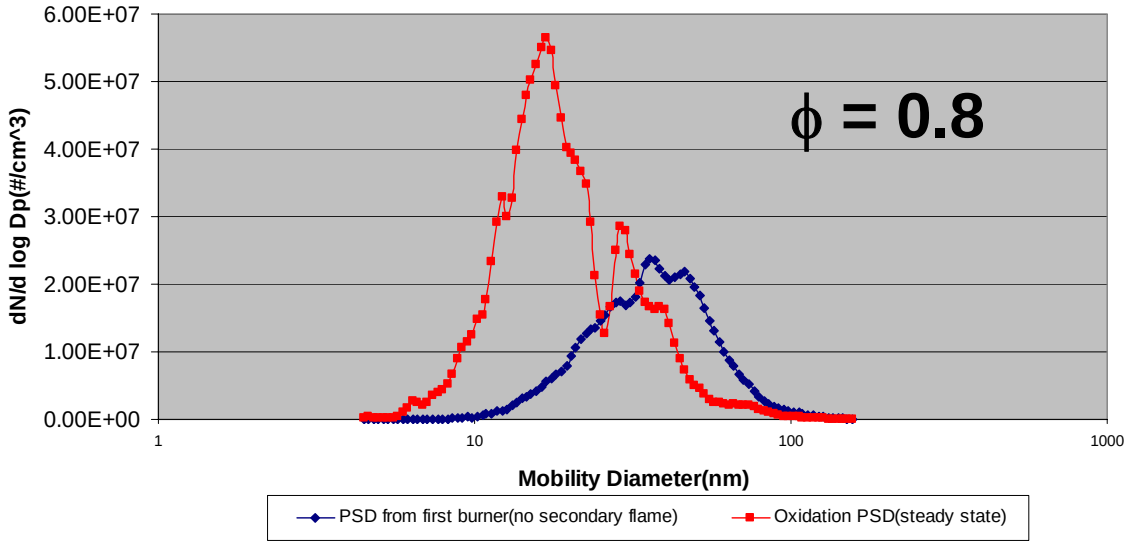
Oxidation – Fuel Rich vs. Fuel Lean



-Diameter decreased from 48.7 to 29 nanometers

-Particle No. DECREASED from 12,100,000 to 11,200,000 (correcting for counting efficiency)

-80% mass burnout



-Diameter decreased from: 43.7 to 26.7 nanometers

-Particle No. INCREASED from 10,500,000 to 21,600,000

-57% mass burnout

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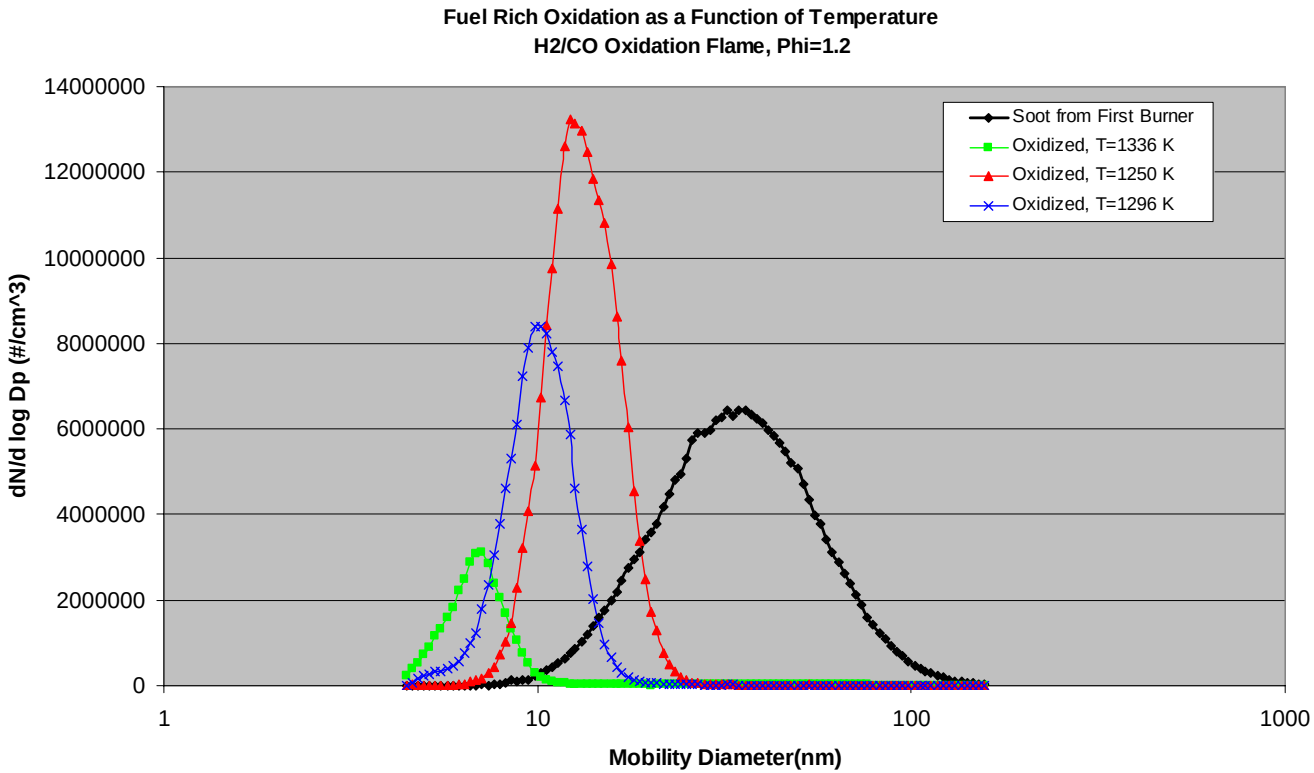
OH Rate Temperature Dependence



-Both particle diameter and particle number decrease with increasing temperature

-Burnout: 95-99%

-Rates were calculated from this data and were also modeled for these conditions

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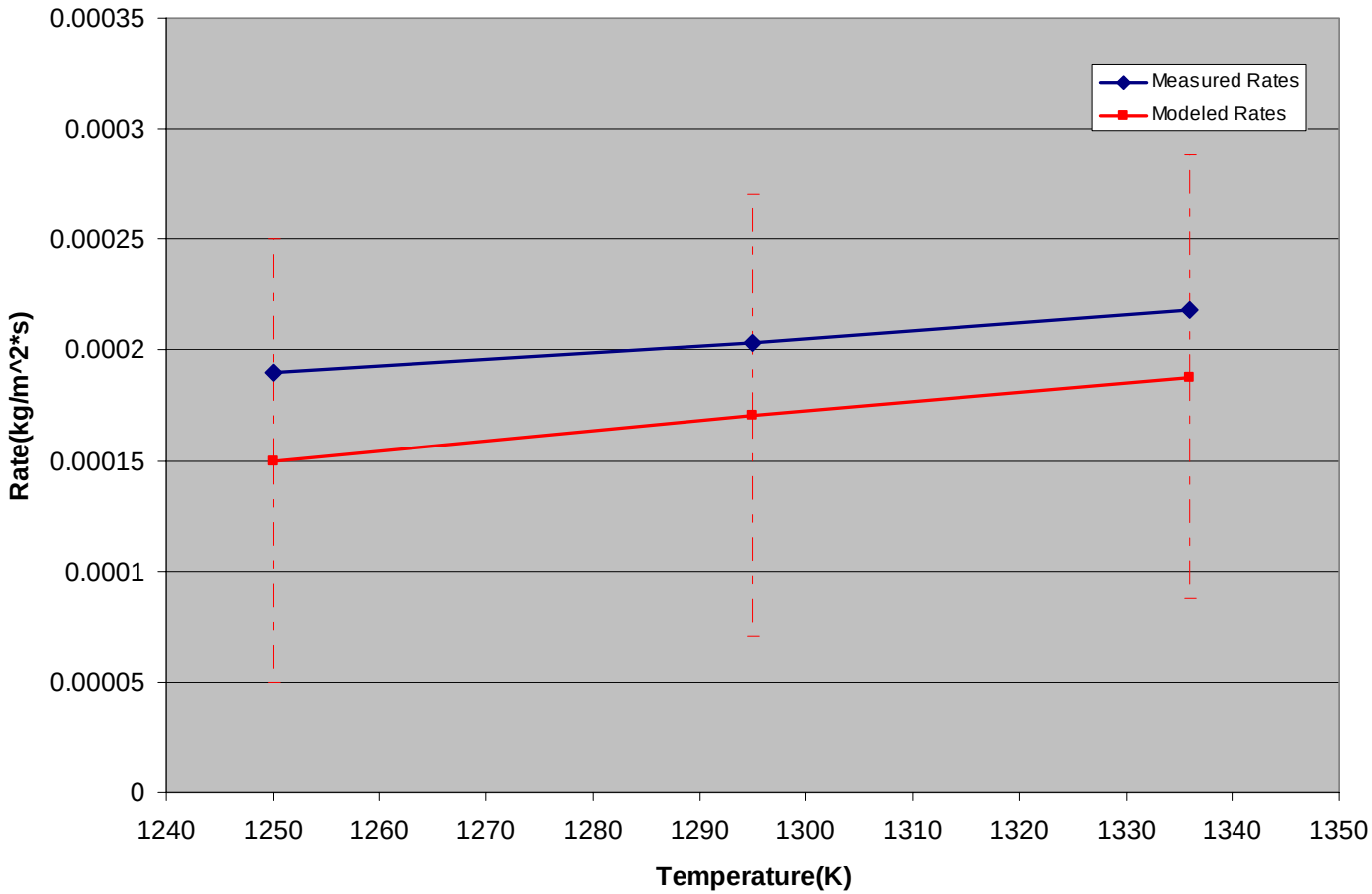
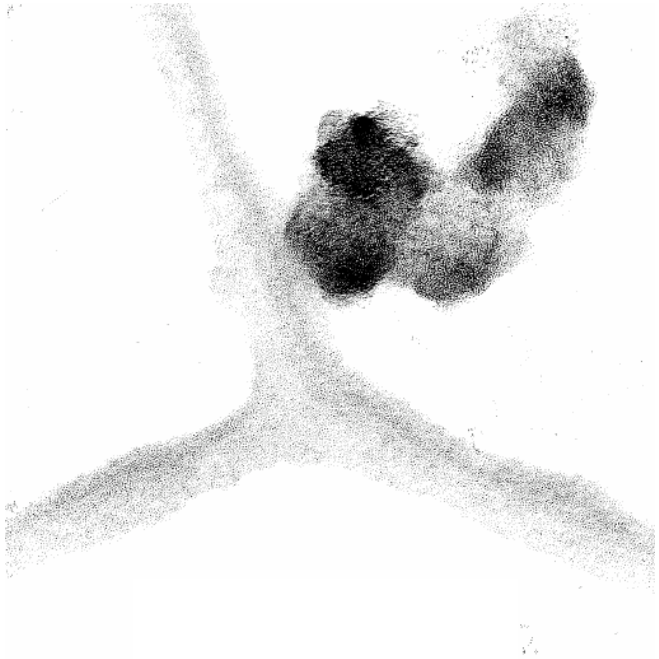
Measured OH Oxidation Rates



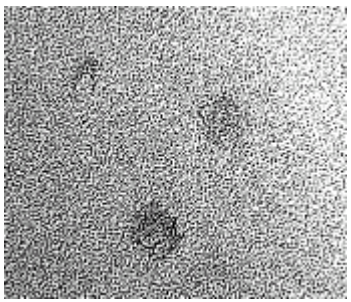
-The measured rates are slightly higher than those predicted by Neoh but have the same trend

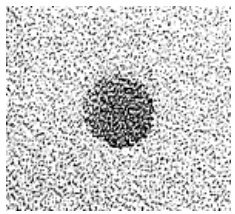
-Average OH Collision efficiency was 0.27 for these runs. Neoh reported values between 0.13 and 0.28.

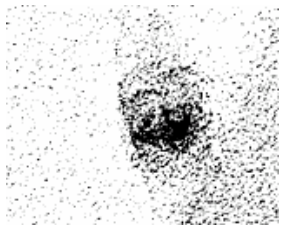
TEM from Two-Stage Burner



- Soot from 1st Burner
- Soot consists of agglomerates made up of primary particles
- Size: about 100 nm







- Soot from 2nd Burner
- Large soot agglomerates are not visible
- Size: about 20 nm

Conclusions

- Developed sampling methodologies for a range of particle sizes from 3 nm to over 100 nm
- Developed two burner systems for the study of soot formation and oxidation
- Soot generated in the Inverse Diffusion Flame appears to be “young soot” based on particle size
- Distributions above the burner show a coagulation mode
- Fuel Rich oxidation rates were measured and were consistent with the Neoh OH Oxidation Model
- Fragmentation of soot under fuel-lean conditions was evident from an increase in total number concentration
- TEM pictures from the two-stage burner show large soot agglomerates from the first flame and much smaller spherical oxidized particles from the second flame

Future Work

- **Perform additional SMPS and TEM measurements in the IDF flame**
  - **Locations and fuel dilution**
- **Perform Photoacoustic measurements to explore the optical properties of soot from the IDF**
- **Develop improved kinetic rate constants for OH oxidation and incorporate with NSC model**