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Soot Nanostructure: Definition, Quantification and Implications

Randy L. Vander Wal*

and

Aaron J. Tomasek

Gordon M. Berger

Kenneth Street

David R. Hull

William K. Thompson

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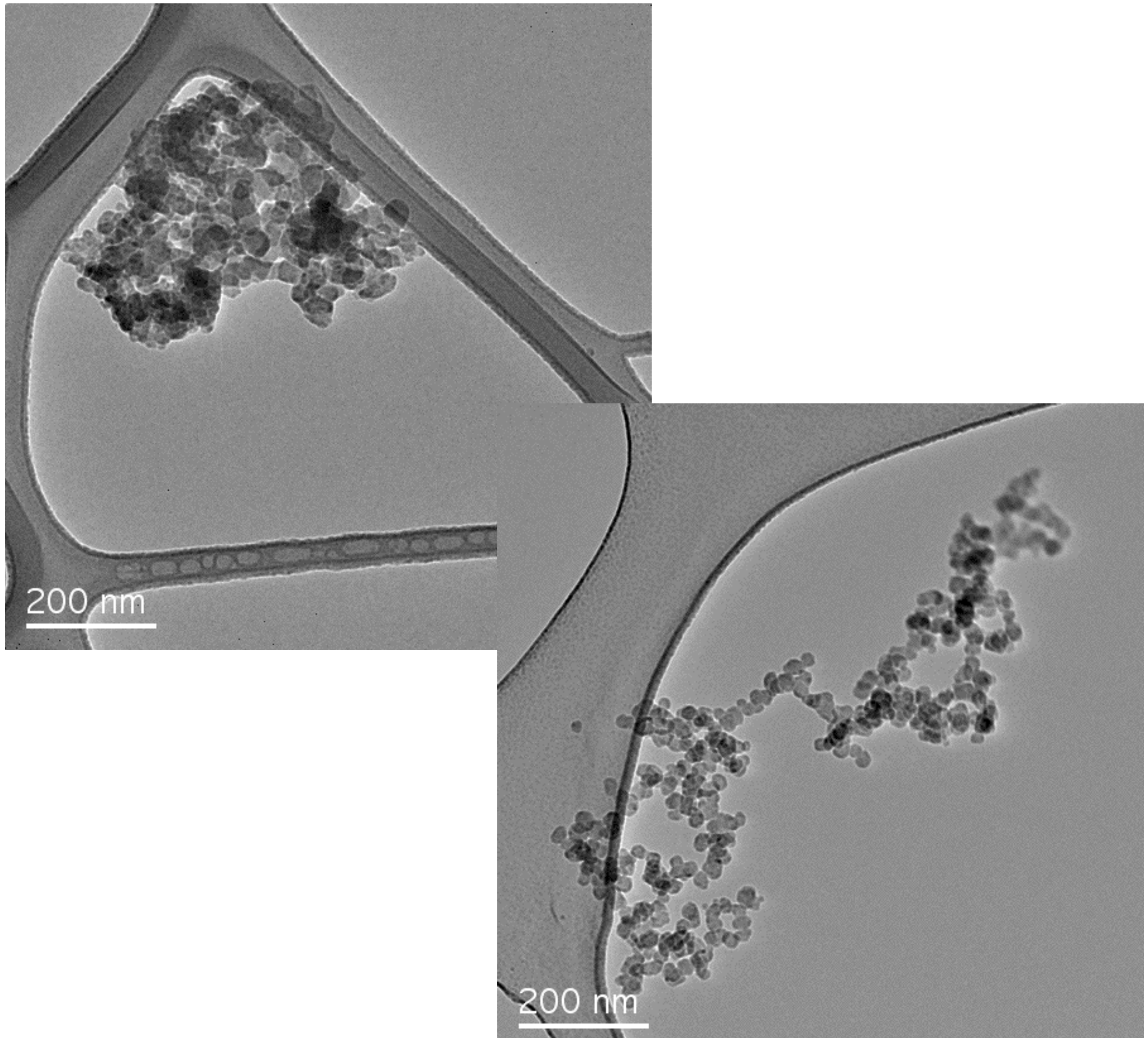
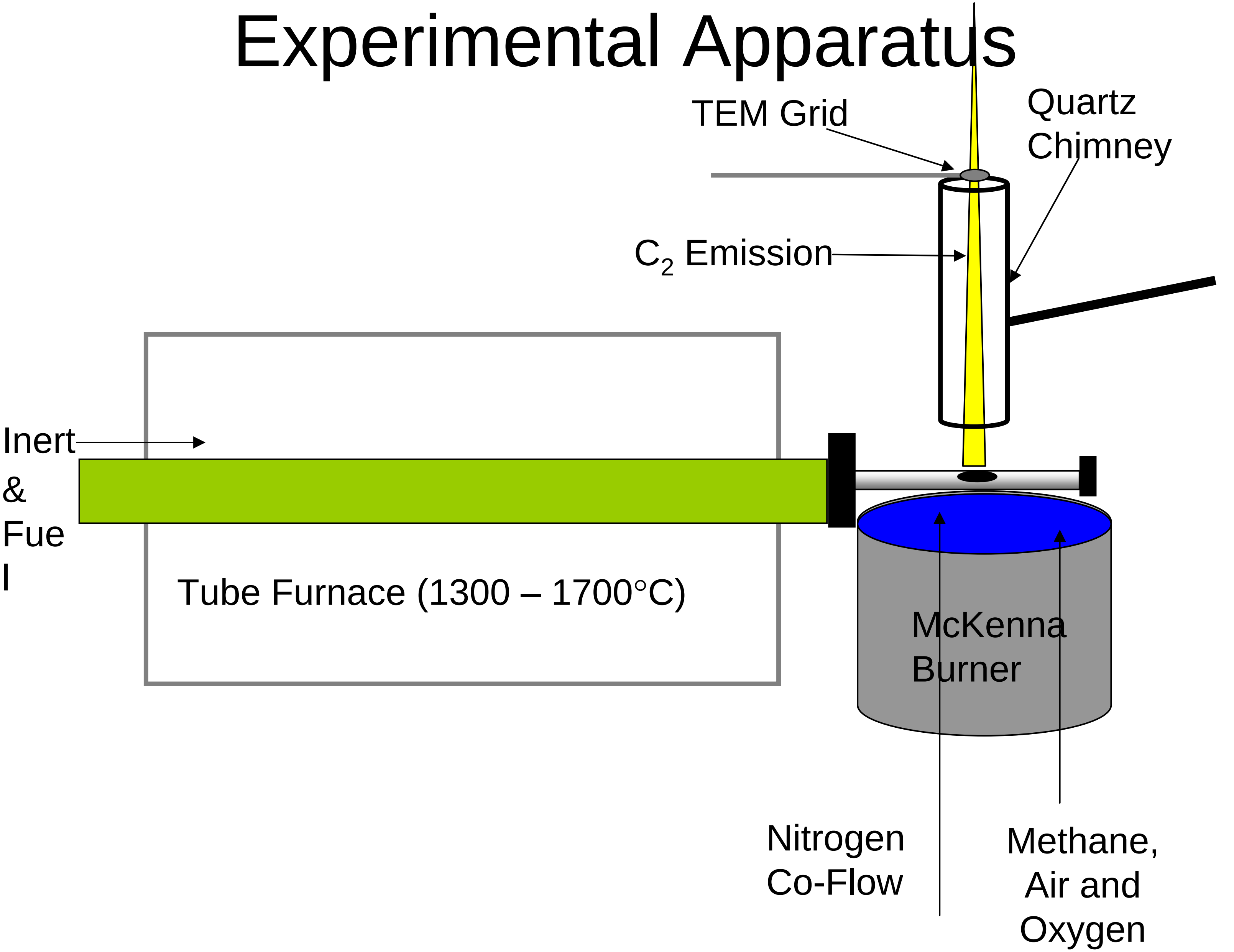
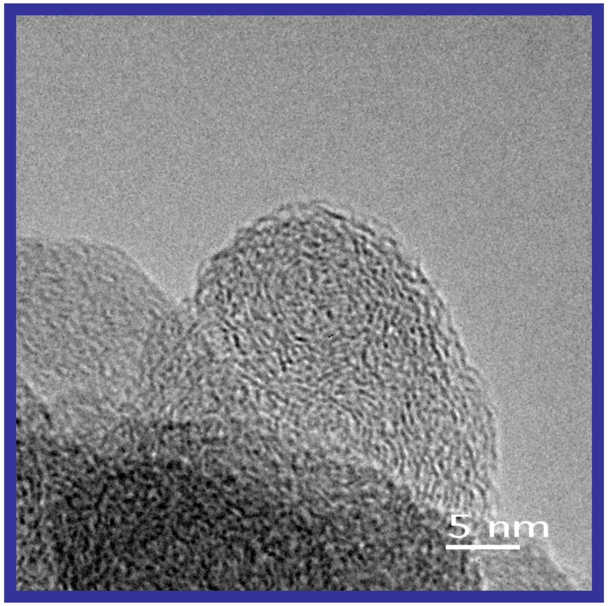
Soot Macrostructure: Aggregate Size and Morphology



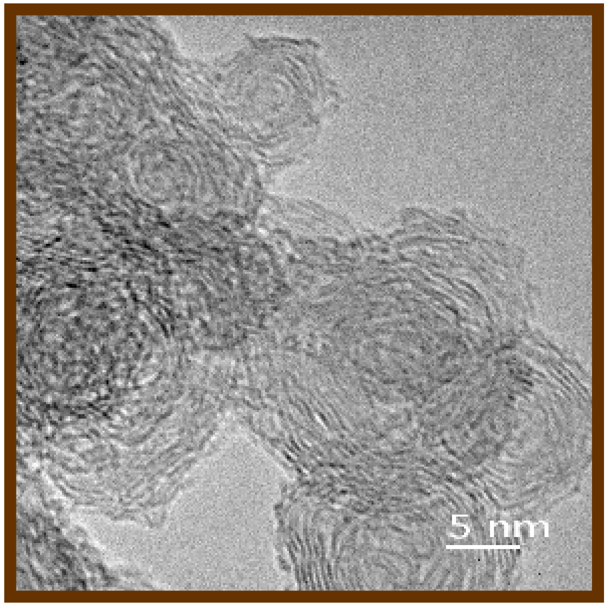
Soot Nanostructure: Experimental Apparatus



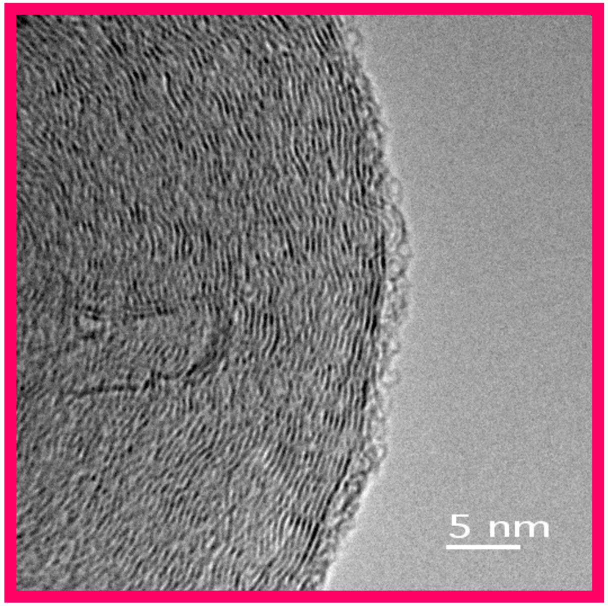
Soot Nanostructure: Definition using HRTEM Images of Primary Particle (Internal) Structure



Amorphous (Benzene)

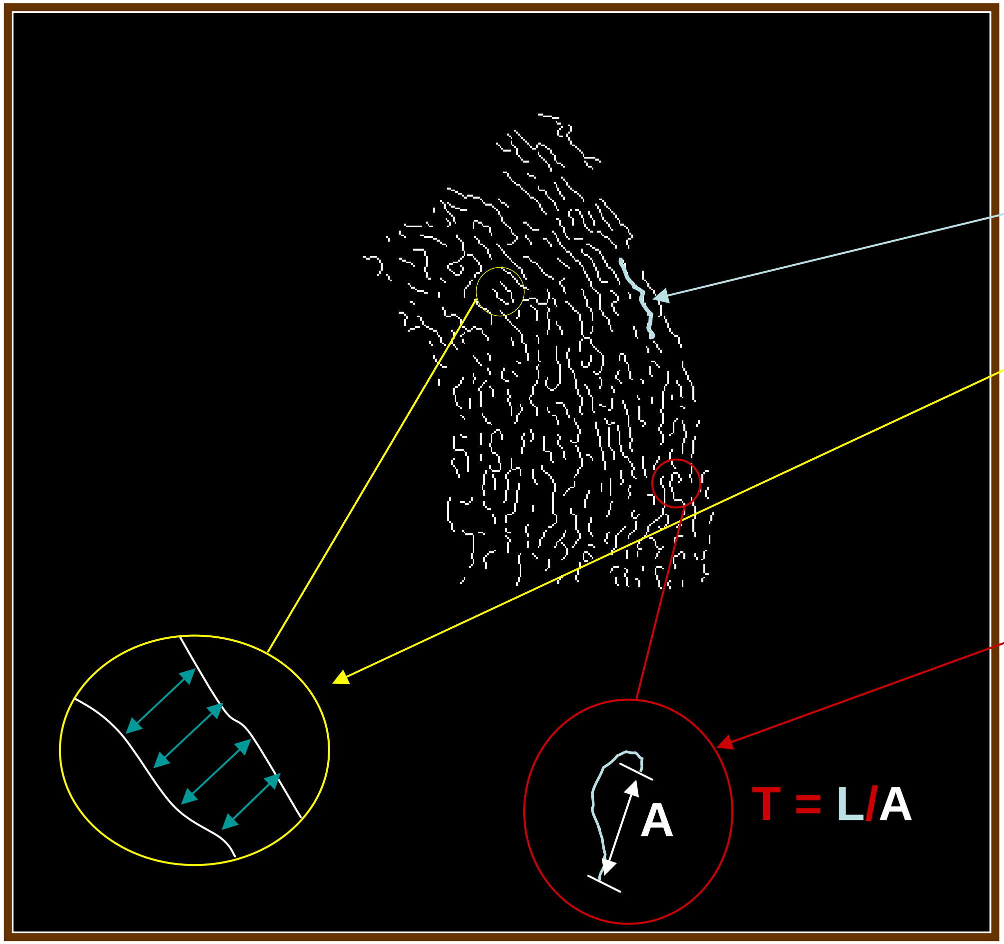


Fullerenic (Ethanol)



Graphitic (Acetylene)

Statistical Properties Extracted from HRTEM Images (of soot nanostructure)



Position

Length

Fringe Separation

Orientation

Tortuosity

Fringe Density

Soot Nanostructure: Quantification via Fringe Analysis

Algorithm-Optimas® Version 6.5

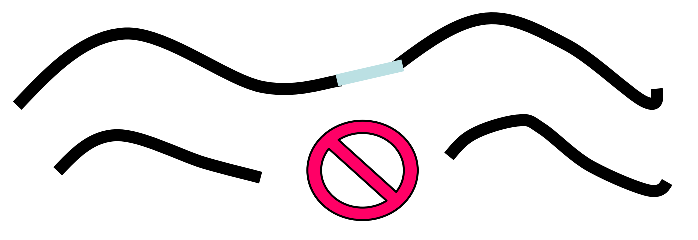
Operations:

1. **Switches 256 grayscale image to binary**
2. **Removes all pixels not above threshold**
3. **Removes remaining pixels and groups of pixels that do not form extended lines**
4. **Uses position of pixels within lines to determine length, curvature, etc. of fringes**

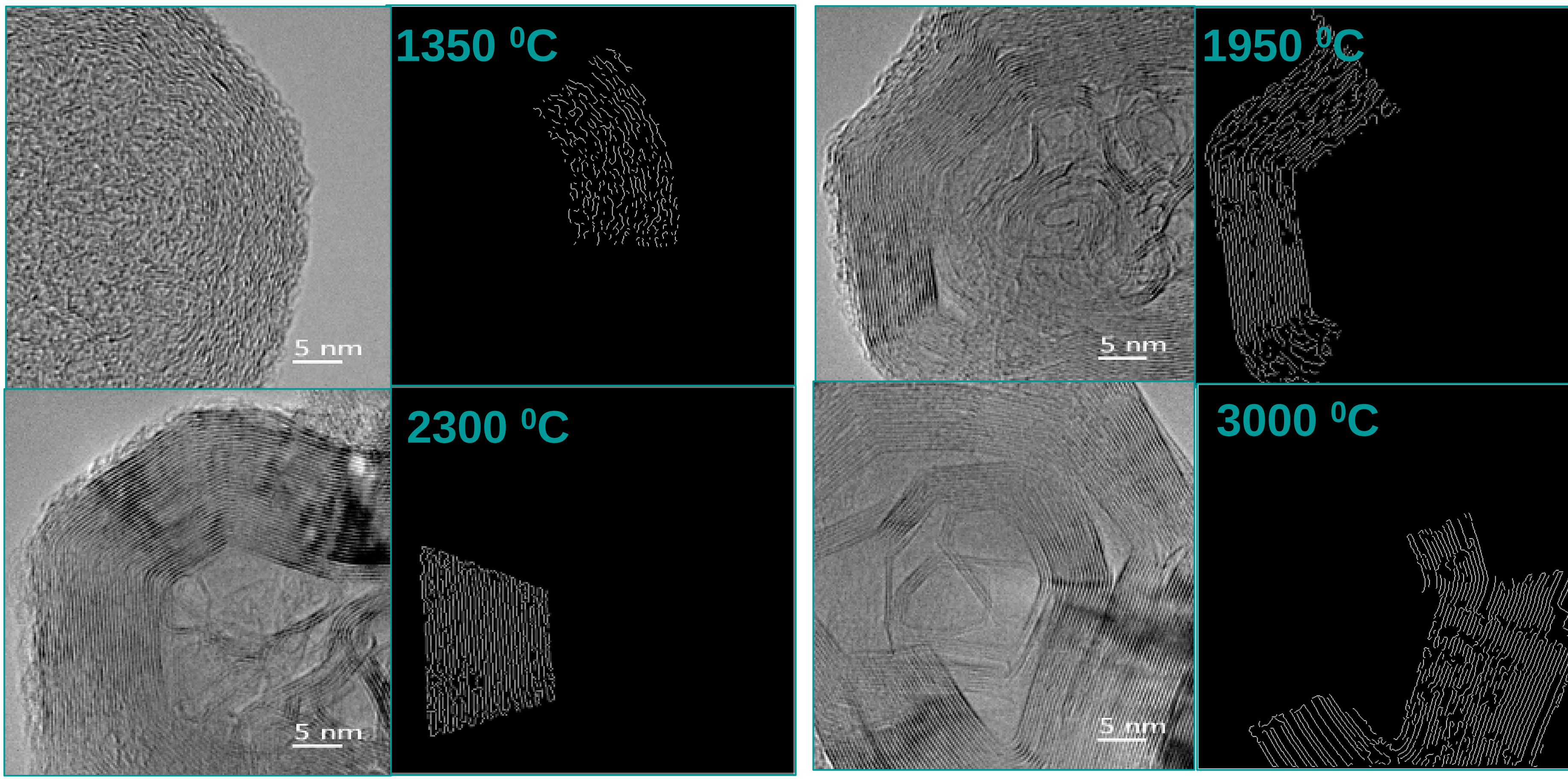
Soot Nanostructure: Quantification via Fringe Analysis

* **Image refinements - To overcome HRTEM image limitations**
  - **Region of interest**
  - **Spatial filtering**
  - **Binary thresholding**

* **Other inputs**
  - **Maximum join distance**
  - **Minimum fringe length**

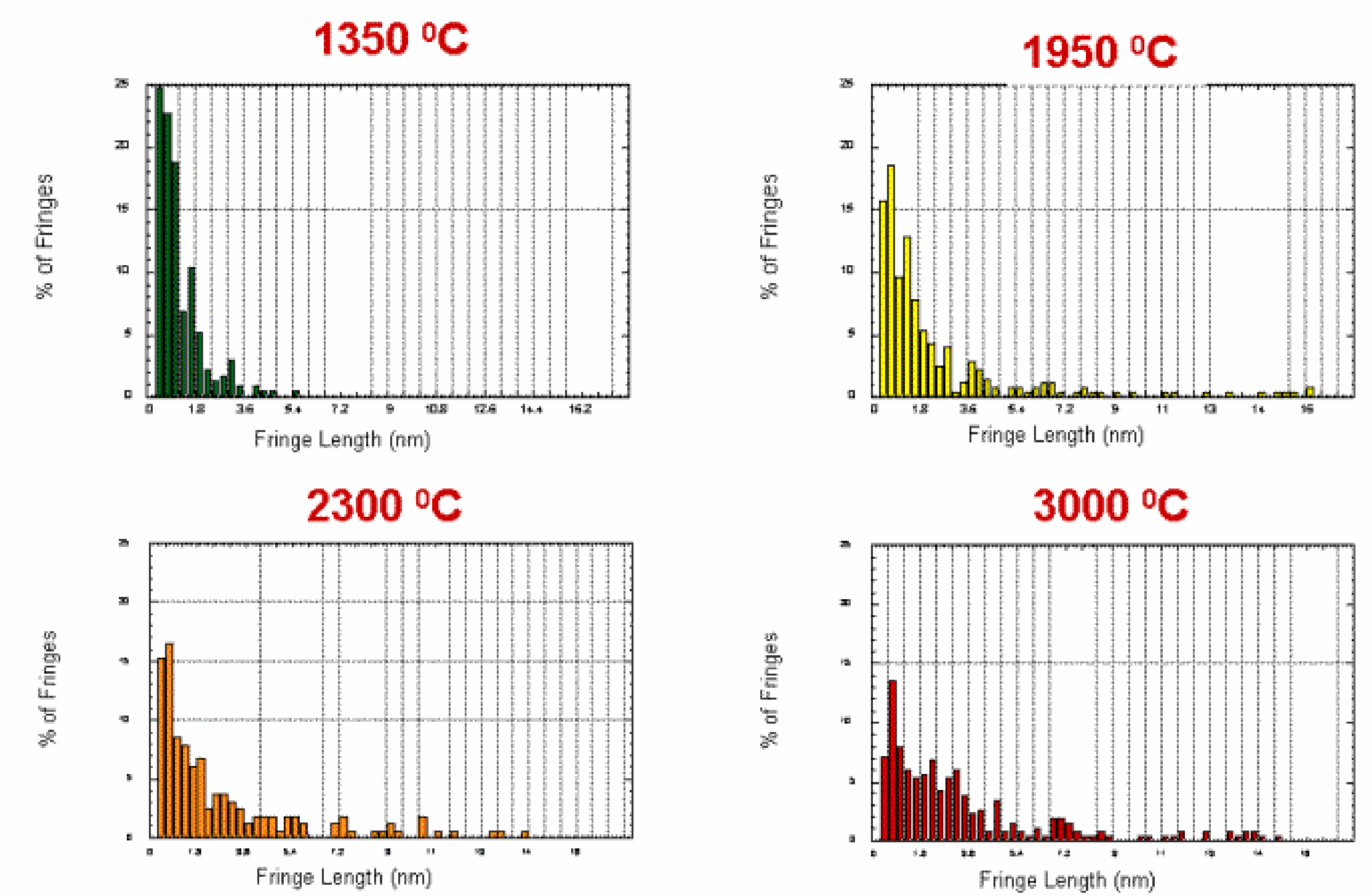


Comparison of Input (HRTEM) and Output (Binary-Fringe) Images



Selected samples of heat-treated carbon black

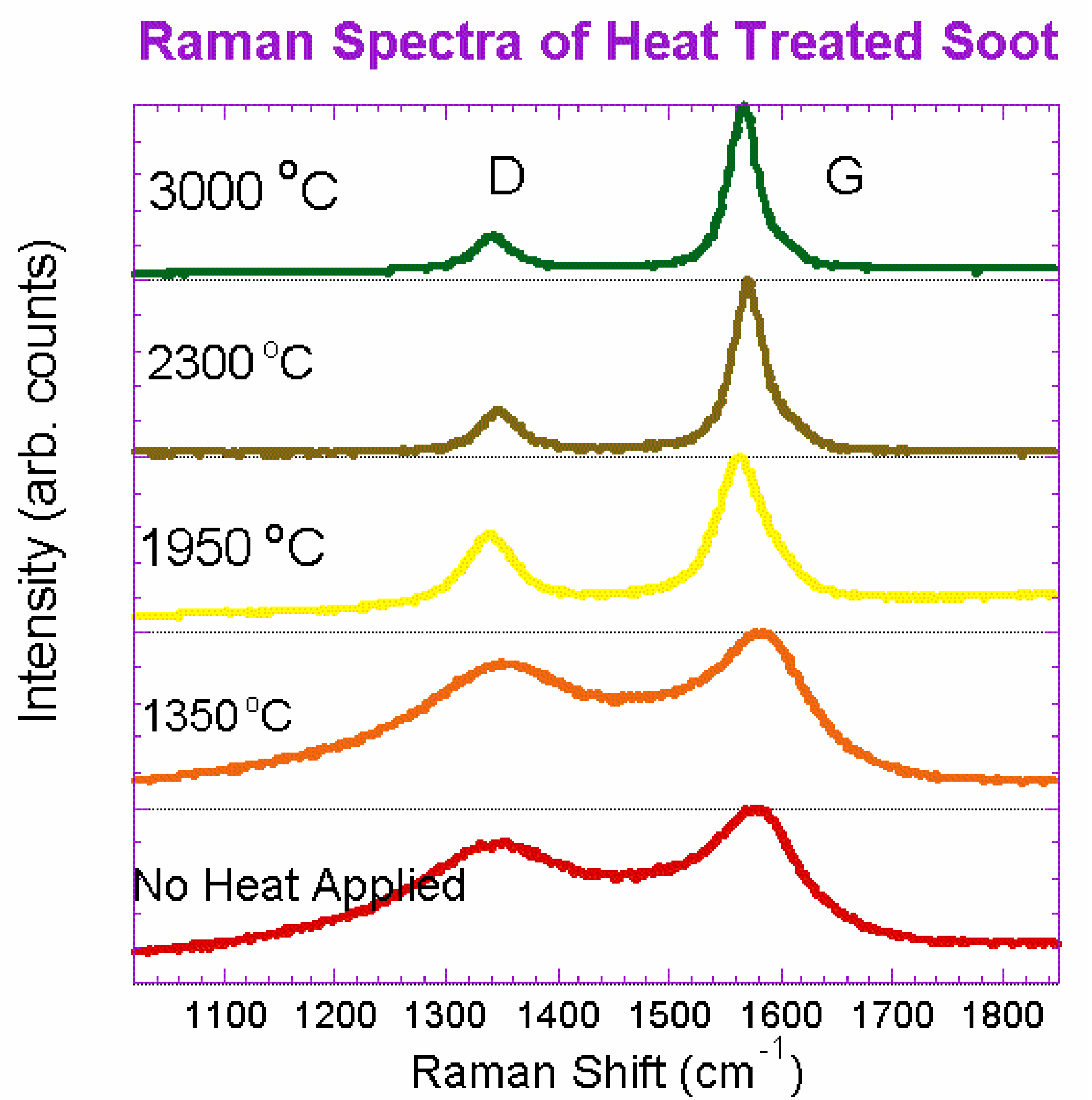
Fringe Analysis Output Data - Fringe Length Histograms



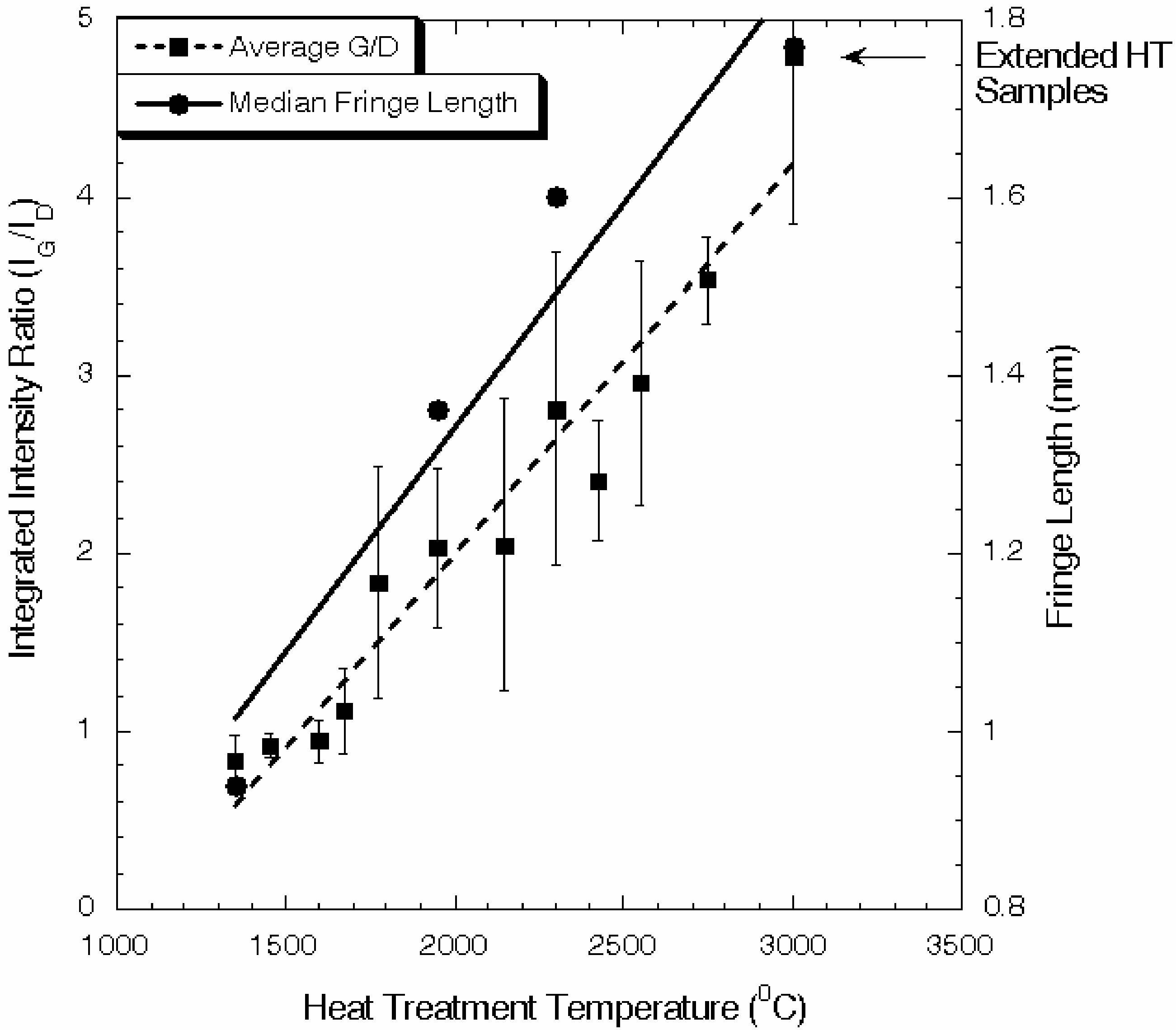
Comparison to Benchmark Methods

E_{2g} or (G) peak at 1580 cm^{-1} (Graphitic)

- A_{1g} or (D) peak at 1360 cm^{-1} (Disordered)
- Intensity ratio have been used to measure in-plane dimensions

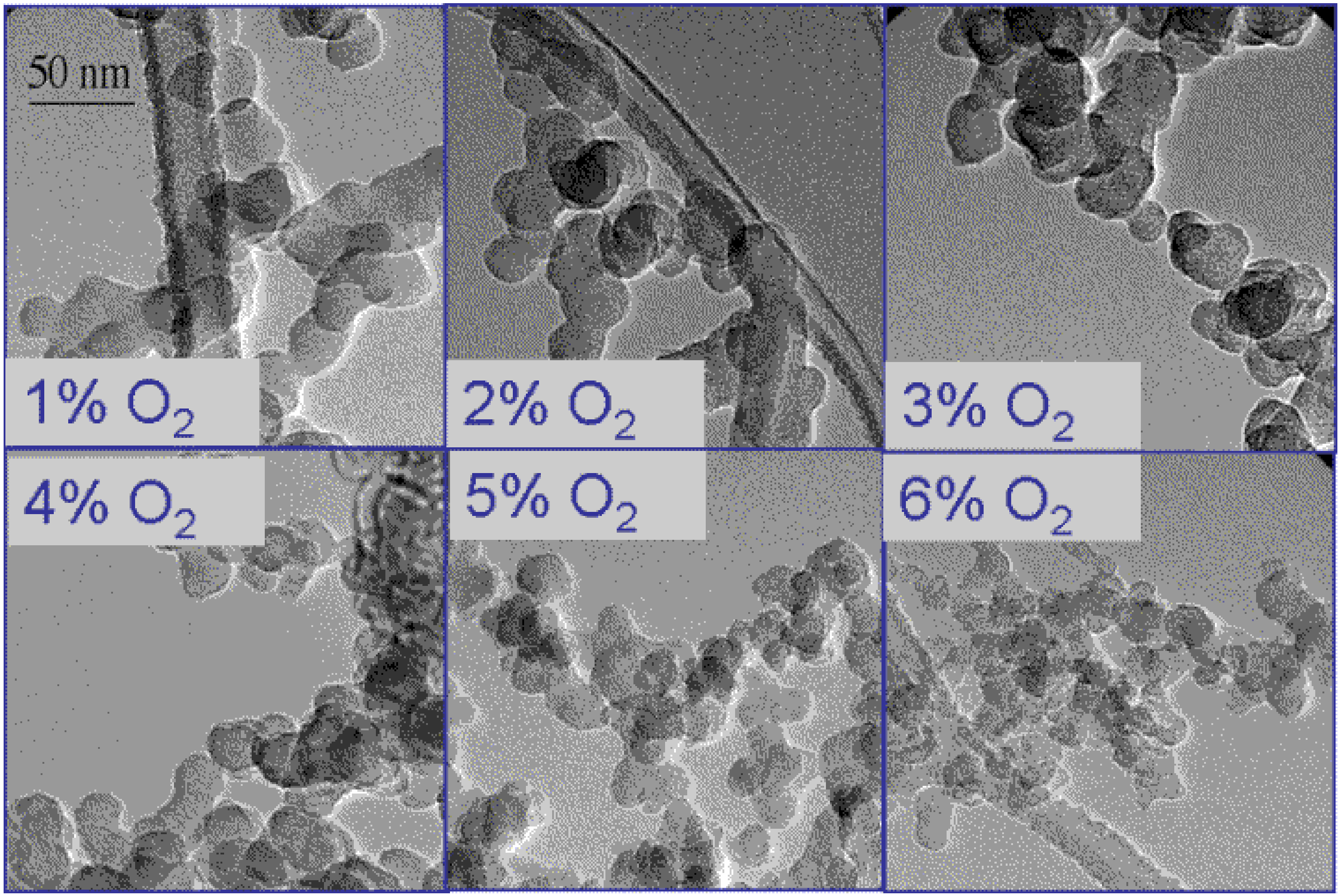


Ratios of Integrated Raman Intensities for Heat-Treated Carbon Soots



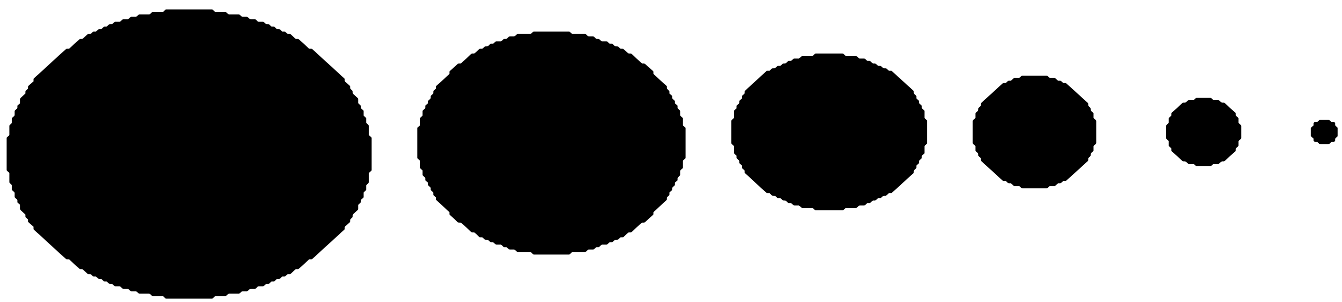
Soot Nanostructure and Implications: Reactivity

TEM images of partially oxidized benzene soot



Oxidation Analysis

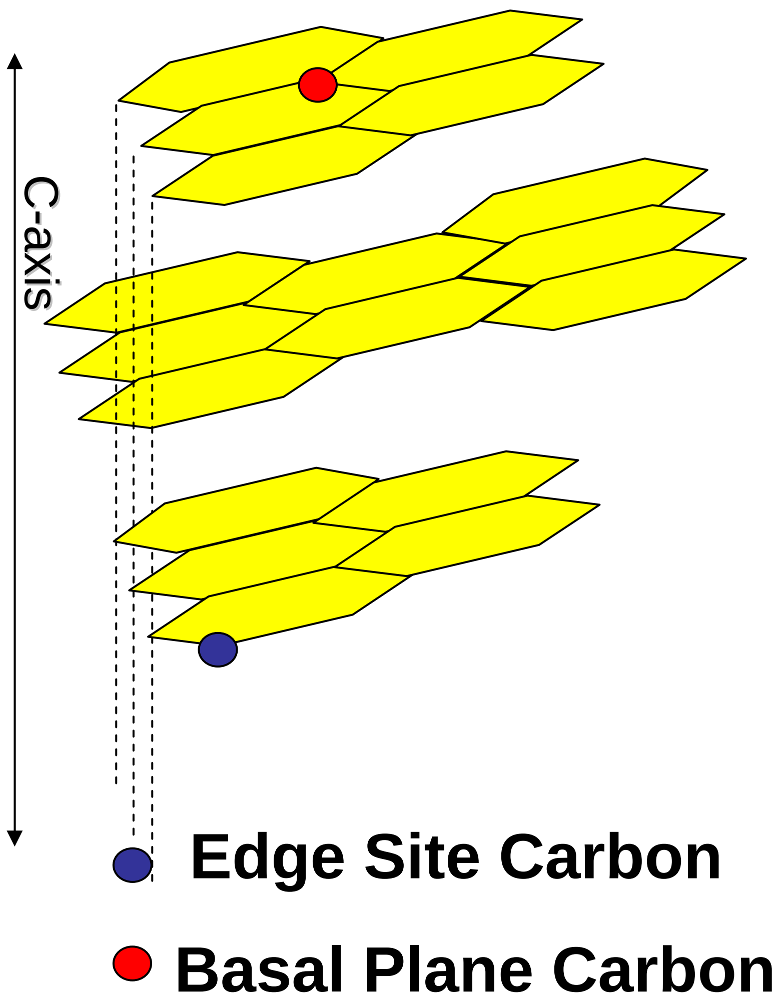
- **Shrinking Spheres Model**



- **Soot Burnout Rate Expression**

$$\omega(kg/m^2 s) = \frac{\rho r_0}{t}\left(1 - \frac{r_t}{r_0}\right) = \frac{1}{A}\frac{dm}{dt}$$

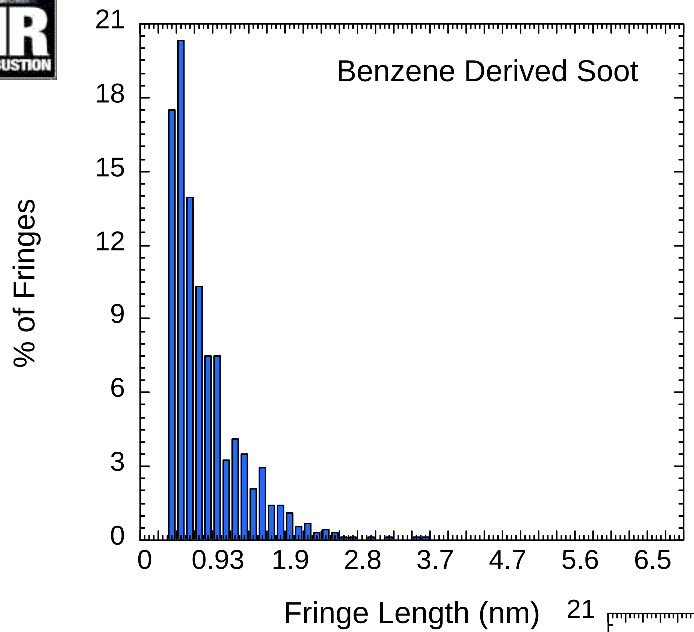
Carbon Soot Nanostructure

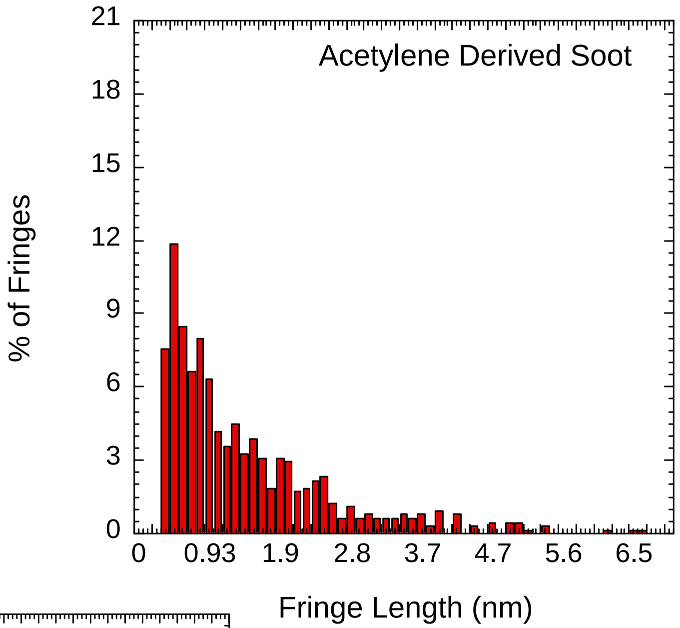


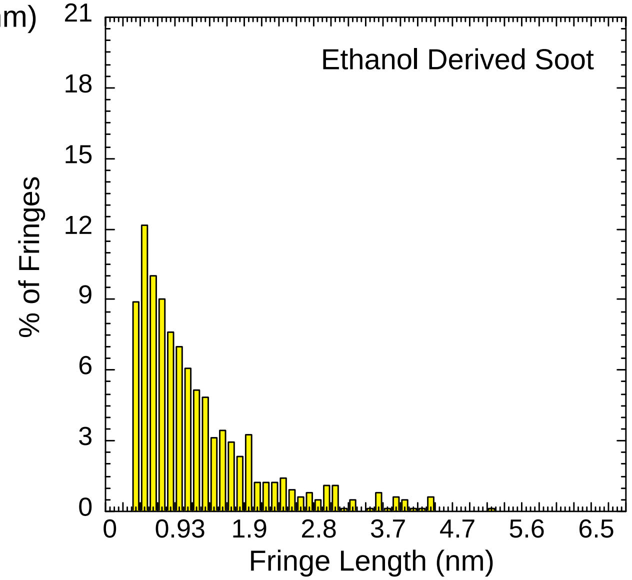
- **Soot oxidation rates are different. What is the cause?**
- **Previous studies ignored nanostructure.**
- **Graphitic carbons are less reactive than amorphous carbons.**
- **Is it just fringe length?**

Nanostructure and Implications: Reactivity

Fringe Length Histograms



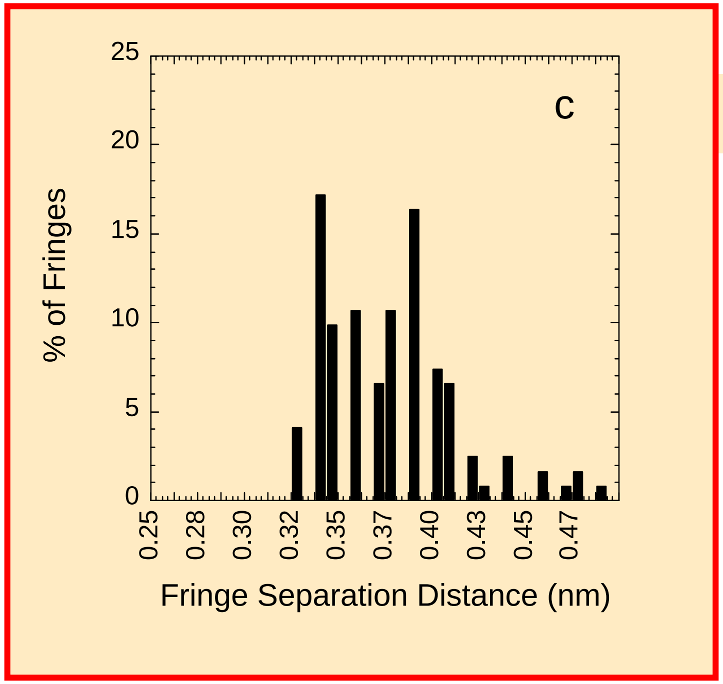




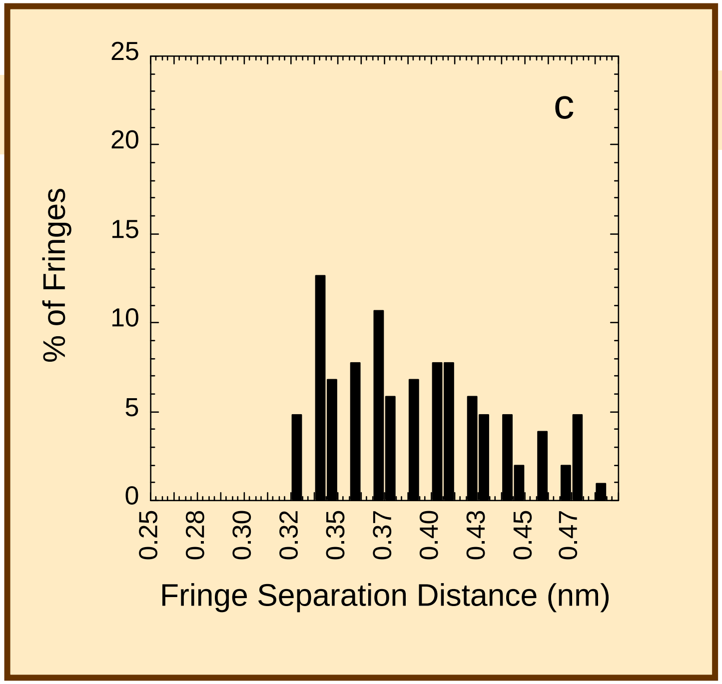
Nanostructure and Implications: Reactivity

Fringe Separation Histograms

Acetylene



Ethanol

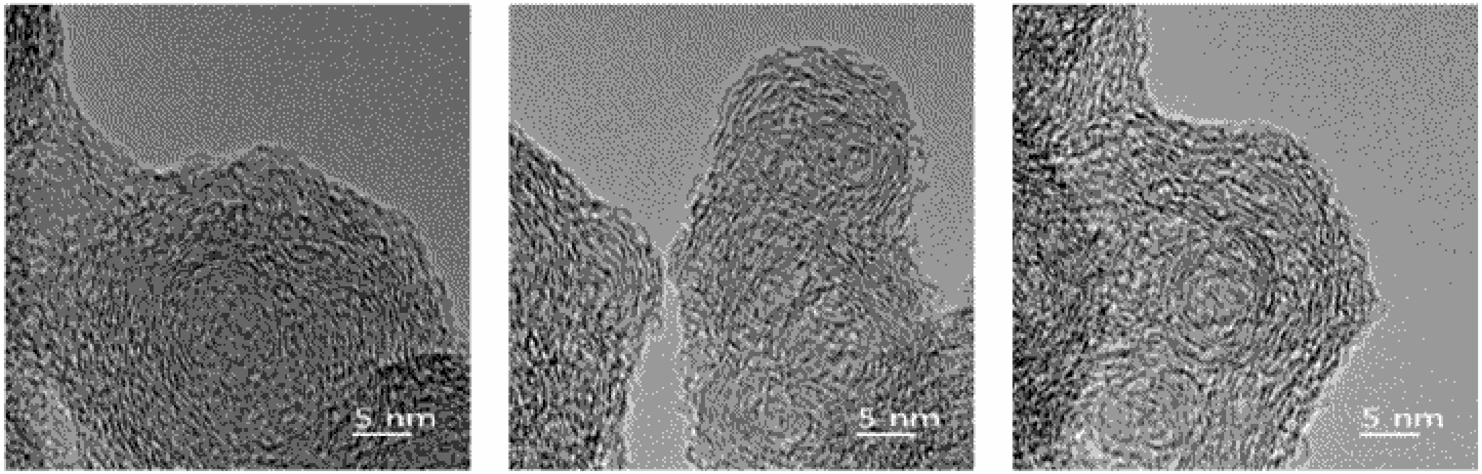


Average $[\omega/\omega_{NSC}]$
17.9 Ethanol
6.7 Acetylene

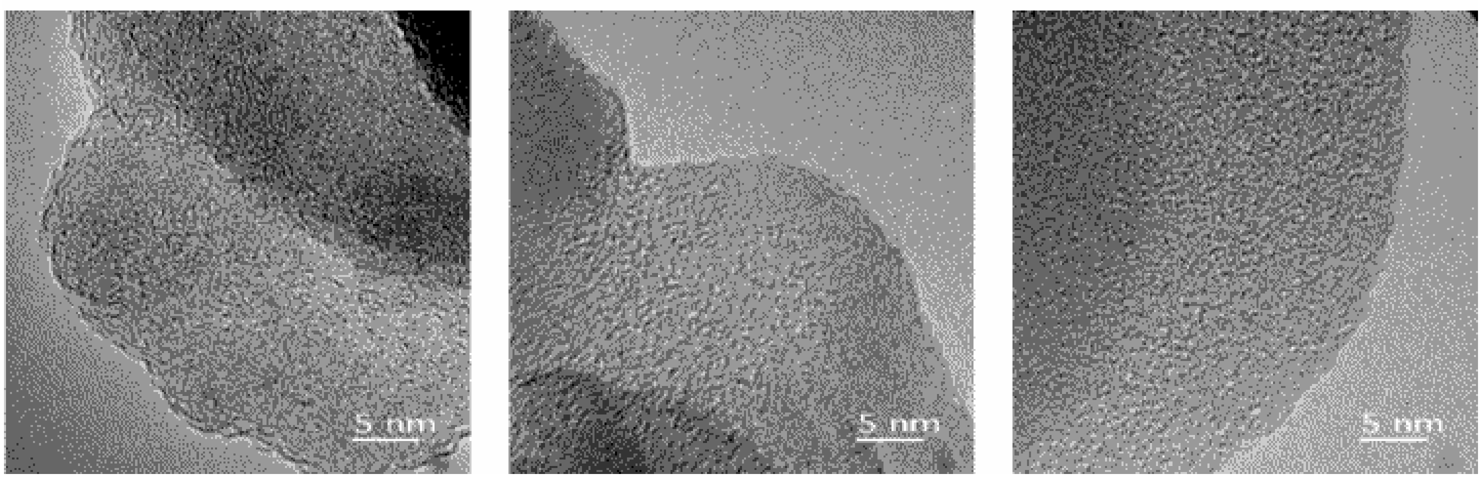
Diesel Engine Soots

(courtesy Sandia Nat. Labs)

Reference Fuel - n-hexadecane + heptamethylnonane
(CN 45-020926B)



Diethylene glycol diethyl ether (DGE)



Conclusions

Soot Nanostructure: (Definition)

*** Soot Nanostructure refers to carbon lamella (layer plane) length, orientation, separation and tortuosity.**

*** Nanostructure is variable, dependent upon temperature, residence time and fuel identity.**

Fringe Analysis Algorithm: (Quantification)

*** Lattice fringe analysis can be used to analyze HRTEM image data and quantify carbon nanostructure through statistical analysis.**

Oxidation Rates: (Implications)

*** Oxidation rates are dependent upon nanostructure - suggests using nanostructure to control (accelerate) oxidation.**

*** Source apportionment via analysis of nanostructure?**

*** Health consequences related to nanostructure?**

*** Environmental impact dependent upon nanostructure?**