Nano Carbon Particles in the Atmosphere: Formation and Transformation

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The University of Utah has assembled an interdisciplinary team specializing in combustion, computational chemistry, aerosol dynamics and analytical chemistry to address the important problem associated with the emission of carbonaceous nanoparticles from diesels. The project integrates experimental studies and theoretical simulation, in multi-level time and particle size scales, which will increase the overall understanding of nanoparticle formation and transformation, specifically addressing the following needs:

- Development of theoretical and experimental methodologies to describe the formation of heavier hydrocarbons and particle inception;
- modeling and measuring the particle size distribution of nanoparticles formed in combustion and in diesel exhaust; and,
- modeling of optical properties and free electrons of soot precursors and soot as it ages.

In addition to the modeling and experimental work, which is underway at the University of Utah, the investigators have collaborations with Argonne National Laboratory (Winans) and the University of Naples (D'Alessio and D'Anna).

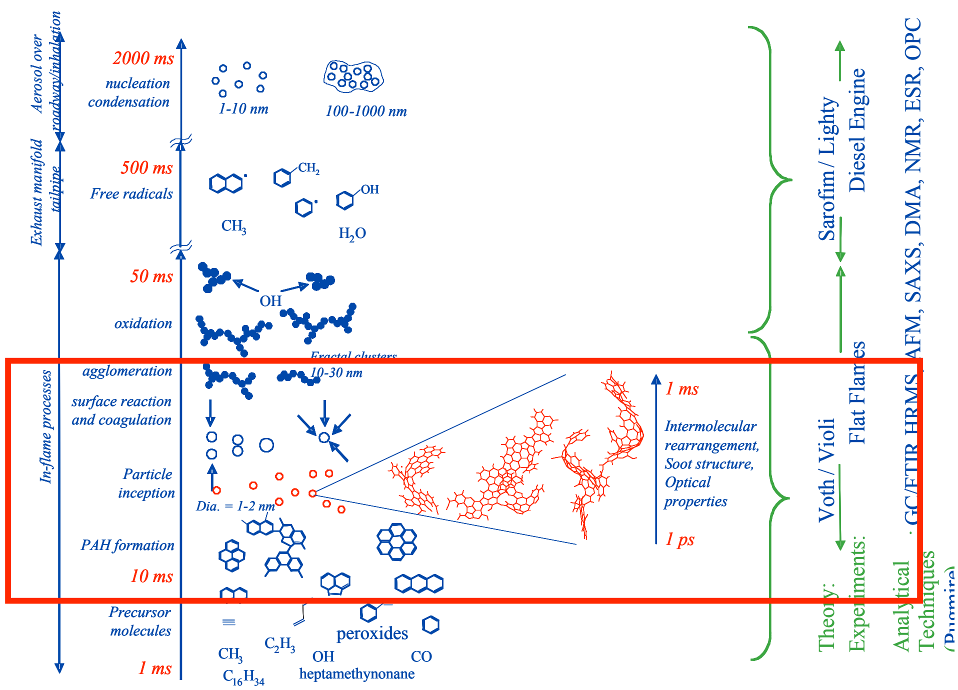


Fig. 1 (left) shows the "roadmap" for the project and illustrates the work which has been accomplished in Year 1 (boxed area). As seen in this figure, the work has focused on particle inception and agglomeration, optical properties, and experimental work on young soot.

The internal structure of soot is important for its optical properties, free radical stabilization, etc., and requires a description of the intramolecular reactions and mesoscale rearrangements that occur during the early stages of soot particle formation. As a first step, the nanoparticles inception is predicted with the atomistic combined Kinetic Monte Carlo/Molecular Dynamics code (1, 2). These nanoparticles are mapped onto coarse-grained particles while preserving their unique topology (see below) by a multi-scale course-graining methodology. These coarse-grained units then interact and aggregate into much larger structures.

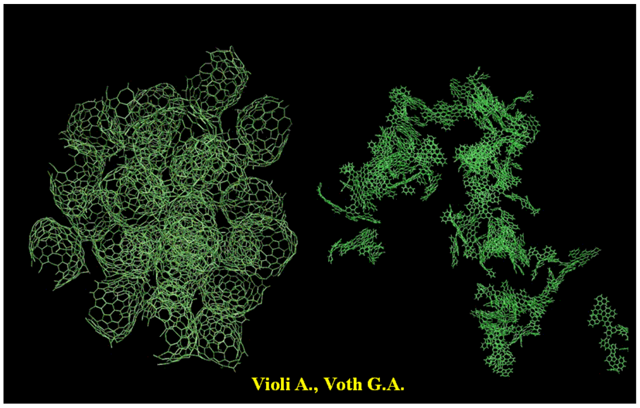
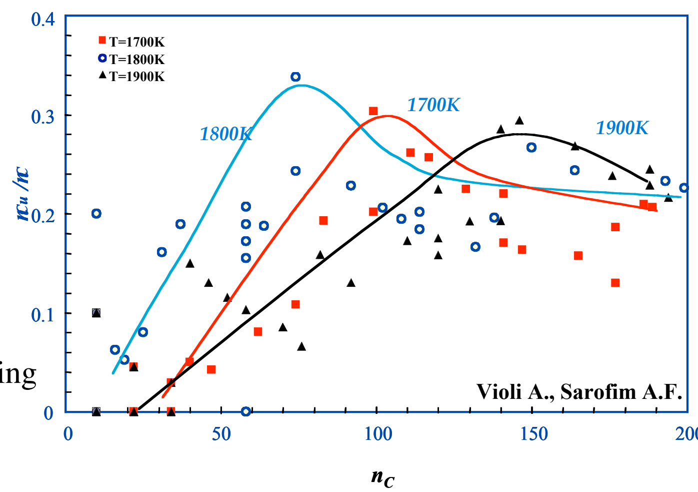
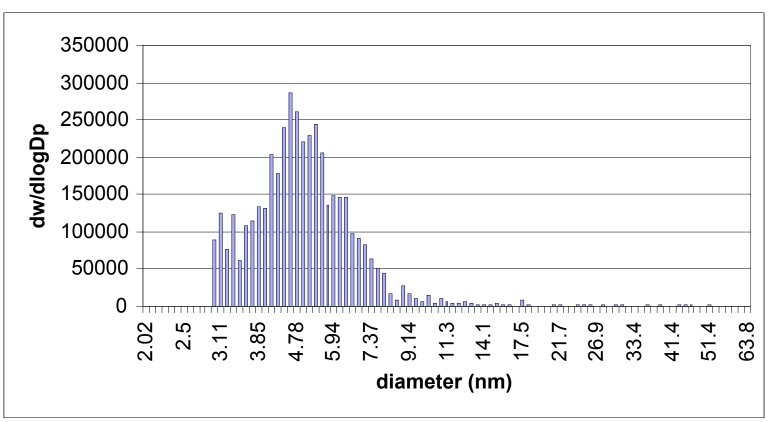


Fig. 2 (left) shows an example of the simulation, for two different agglomeration geometries. Round particles (left) tend to cluster, while "flaky" particles behave differently and tend to subcluster. These simulations, combined with experimental results, will begin to help validate the agglomeration kinetics of soot particles.

Additional modeling work, using an Atomistic Model for Particle Inception, is investigating the presence of free electrons in soot particles. The data shown in Fig. 3 (right) illustrate the number of unpaired carbons per total carbons versus the number of carbons in a cluster. As seen in these simulation results (the lines have been added for clarity), given a constant number of carbons in a cluster, the fraction of unpaired carbons decreases with increasing temperature. These data will be compared to experimental data obtained by Pugmire over the next year.

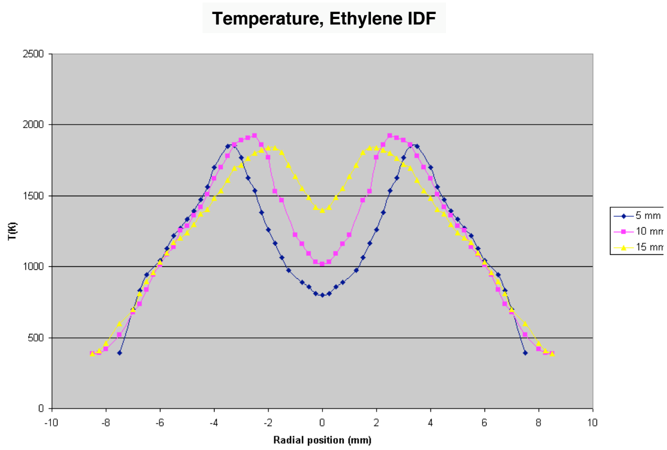


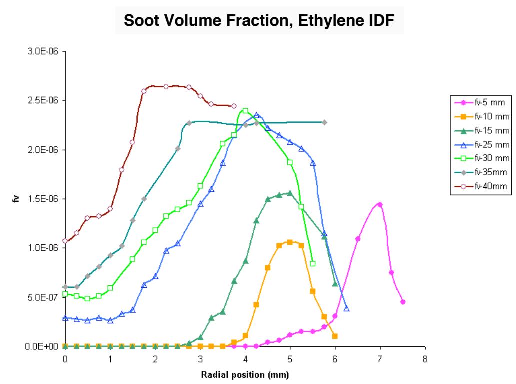
The modeling and experimental work are underway to further understand the role of free electrons and their persistence as soot ages.



In terms of experimental results, an inverse diffusion flame (IDF) to investigate "young" soot (4) has been built. In the IDF, air is fed in the middle of the burner, surrounded by fuel, and a nitrogen shroud. As soot is formed, it passes through nitrogen into the cooler region, quenching reactions. This differs from the traditional, premixed burner, where soot is formed and then diffuses through an oxygen region where the soot further reacts. The resulting particle size distribution is below 10 nm from the IDF flame, as seen in Fig. 4 (left), and as compared to the premixed flame which yields particle size distributions around 30-55 nm. These particle size distributions were obtained from a nano-Scanning Mobility Particle Sizer (nano-SMPS), purchased with the funds from this grant. The nano-SMPS allows us to obtain particle size distributions down to 3 nanometers.

The resulting temperature and soot distributions, as measured by thermocouples and thermophoresis, are shown below, Fig. 5.





As we begin our collaborations with ANL, we will switch to a diffusion burner and further explore through Small Angle X-Ray Scattering (SAXS) the particle geometry and size. In addition, we will begin data exchange with Naples, where both the experimental and modeling PIs have now visited. Naples will give us information regarding particle geometry, size, and optical properties.

References

[1] For further information about this project email **jlighty@coe.utah.edu**

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