A detailed multivariate model for soot growth and coagulation based on particle volume, surface area, reactivity, and H/C ratio

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While the majority of existing soot models describes the particles with a single parameter, usually mass or volume, particles with the same volume can differ greatly with respect to morphology, composition and chemical properties. To account for non-constant surface reactivity as well as the increasing surface-to-volume ratio of large soot aggregates compared to spherical particles, Blanquart and Pitsch (2009) formulated a trivariate model based on particle volume, surface area, and number of hydrogenated surface sites (VSH model).

In this work, the VSH model is extended by a fourth parameter, the particles' H/C ratio, which has been identified as an important quantity to account for the chemical diversity of the particles, as it carries particle properties and information about their history. This model is integrated into a Monte Carlo simulation code, which allows a detailed analysis of the soot Number Density Function (NDF), and is here validated for a series of laminar premixed flames. With this multivariate characterization of the soot particles, in addition to the soot volume fraction and number density, important physico-chemical properties of the particles can be tracked during their formation and growth process.

The purpose of including the H/C ratio into the model is twofold. First, the evaluation of the H/C ratio is used to reduce the uncertainty in the soot growth rates. Second, an improved coagulation model is formulated based on the H/C ratio.

Reduction of the Uncertainty of Soot Growth Rates

It is well accepted that the mass growth of soot particles can be categorized into acetylene-based growth according to the HACA mechanism, which adds mainly carbon to the particle, and PAH-based growth, which adds both carbon and hydrogen. However, large uncertainties exist for the rates of both processes. By evaluating the H/C ratio, it can be assessed whether the relative importance of these processes is well captured. Extending the sensitivity study by Wick and Pitsch (2015), different rates suggested in the literature are compared for laminar premixed methane, ethylene, and benzene flames experimentally studied by Russo *et al* (2015). A combined evaluation of the soot yield and the H/C ratio in these flames leads to a reduction in the uncertainty of the growth rates.

Coagulation Model based on the H/C Ratio

The two limiting cases for coagulation are coalescence, which results in spherical particles, and aggregation, which leads to fractal aggregates composed of smaller primary particles. To capture the transition between these coagulation regimes, in the VSH model by Blanquart and Pitsch (2009), a collision model has been introduced, which is based on the size ratio of the colliding particles. However, the transition from coalescence to aggregation depends more on the rigidity of the particles, which is expressed by the H/C ratio. Particles with a large H/C ratio are more deformable and tend to coalesce, while particles with a low H/C ratio are more rigid and form aggregates. Figure 1 compares the predicted NDF in a premixed ethylene flame with experimental data by Camacho *et al* (2015). Using the collision model based on the diameter, aggregation is overpredicted. With the novel formulation of the collision model based on the H/C ratio, the NDF is well predicted at all heights above burner (HAB). Compared to the collision model based on the volume, improvements are obtained regarding the onset of aggregation and maximum particle diameter close to the burner and the shape of the trough of the bimodal NDFs further downstream.

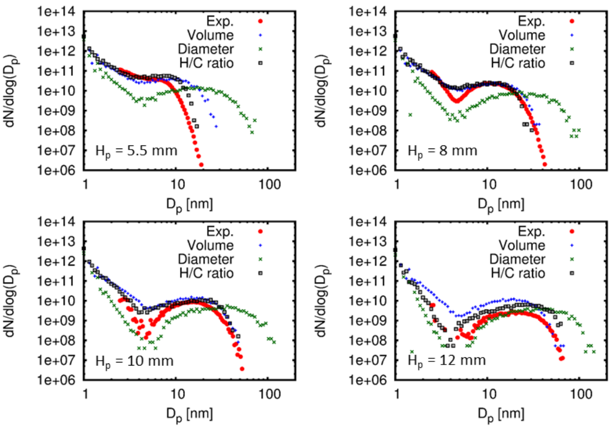

Figure 1. Soot NDFs in a premixed ethylene flame. Collision models based on particle volume, diameter, and H/C ratio. Exp. data by Camacho *et al* (2015).

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