Computational modeling of aerosol formation and evolution using OpenFOAM®

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In the process of single-species homogeneous vapor condensation into aerosol, surface is created between liquid and vapor. The energy of formation of such surface limits the condensation of vapor from a supersaturated state. Nucleation, the mechanism of generation of embryo sites, or nuclei, on which vapor is able to condense, is an important process. Developing an understanding of this process is important, as it may lead to a considerable contribution to many engineering problems, as well as atmospheric and environmental science. In this work, we present an OpenFOAM®-based numerical simulation tool, which is capable of predicting aerosol formation using a two-moment representation of the aerosol and classical nucleation theory. We aim at developing a flexible utility, which enables researchers interested in various aerosol production-related applications to quickly study concepts like aerosol nucleation, condensation, diffusion and transport.

The general convection-diffusion-reaction equations for species vapor mass fraction Y, aerosol droplet number concentration N (first moment) and aerosol mass fraction Z (second moment) in an Eulerian framework are given with:

$$\partial_t(\rho Y) + \partial_j(u_j \rho Y) = \partial_j(D\partial_j(\rho Y)) - S_{v\to l} \quad (1a)$$
$$\partial_t(\rho Z) + \partial_j(u_j \rho Z) = S_{v\to l} \quad (1b)$$
$$\partial_t N + \partial_j(u_j N) = J_N - J_C \quad (1c)$$

with carrier fluid density ρ, velocity field u_j, vapor diffusivity D and droplet coalescence rate J_C. The source term $S_{v\to l}$ associated with the transfer from vapor to liquid is governed by two contributions: that of nucleation, S_{nuc}, and that of evaporation or condensation, S_{e-c}. The nucleation mass flow rate is specified as the mass of a critical cluster, multiplied by the nucleation rate J_N. A critical cluster is defined as a kernel of condensed molecules with a probability for growing into a droplet equal to the probability for dispersing into individual molecules again. The nucleation rate for a vapor is given by classical nucleation theory, and strongly depends on the saturation. Transport equations (1) contain multi-scale physics. The typical time scales of diffusion and convection are orders of magnitude larger than those of nucleation or condensation. The formation of critical clusters is relatively fast. It is clear that the formation of aerosol droplets should numerically be resolved at the smallest time scale, in order to yield an accurate result.

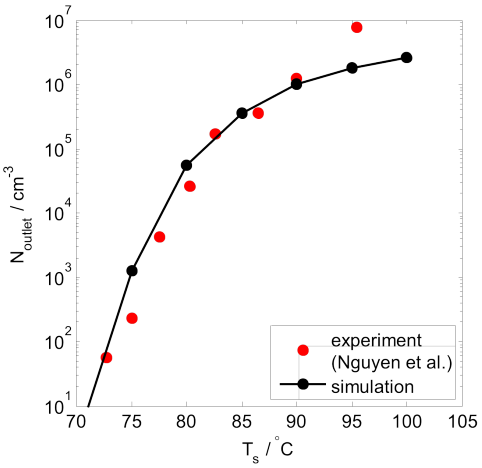

Figure 1: Aerosol droplet number density at pipe outlet as a function of saturator temperature T_s.

We compare our numerical approach with the results of physical experiments, each using a laminar flow diffusion chamber (LFDC), (Nguyen *et al*, 1987; Lihavainen and Viisanen, 2001; Hämeri and Kulmala, 1996) with different species and/or carrier gasses. Figure 1 shows a good agreement between experimental and numerical results for the aerosol droplet number density N as a function of saturator temperature T_s, with dibutylphthalate (DBP) as saturated vapor.

The development of the OpenFOAM®-based simulation tool is a stepping stone towards a more comprehensive multi-species model. We aim at developing a robust numerical approach which is capable of determining the composition and size of the critical cluster and its subsequent evolution. Moreover, we will extend the presented two-moment approach with a third moment, taking into account the width of the droplet size distribution of the aerosol.

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