Modelling aerosol agglomeration using molecular dynamics methodology

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In this study, we investigate the dynamic behaviour and morphologies of the aerosol aggregates in the free molecular ($Kn \gg 1$) and transition ($Kn \approx 1$) regimes. We model the formation and growth of the aerosol aggregates using the molecular dynamics (MD) simulation software ESPRESSO (Arnold, 2013). During the simulations, the binding agents are created between the collided particles in order to irreversibly adhere the collided particles and construct aggregates such that the particles remain fixed at the first contact point and do not rearrange their positions. The random motion of the Brownian particles is simulated by solving the Langevin equations of motion. We use the Langevin thermostat in ESPRESSO (for details please see (Arnold, 2013)). At each time step, all particles are subjected to a random and a frictional force. These two forces satisfy the fluctuation-dissipation theorem and balance each other. The Langevin equation for the i-th particle is then given by

$$m\dot{u}_i(t) = -\gamma u_i(t) F_C + W_i(t), \qquad (1)$$

where F_C are conservative forces, γ is the friction coefficient between the primary particles and surrounding fluid, u_i is the velocity of the i-th particle and $W_i(t)$ is Einstein's white noise term, which is a Gaussian random source.

The exact modeling of the friction coefficient depends on the Knudsen number, $Kn = 2l_{fluid}/d_p$. In the continuum regime, the well known Stokes law applies. However, in the free molecular regime, a slip velocity between the particle and the surrounding fluid exists. The friction coefficient will be lower than the Stokes law would predict, and the so-called Cunningham correction, C_C needs to be introduced (Chen, 1984),

In this work, the simulations are carried out with a system consisting of 1250 spherical particles suspended in motionless air. Particles are randomly placed in a cubic box with periodic boundary conditions for all sides. The particle interactions are taken to be a Lennard-Jones intermolecular potential assuming all primary particles have the same (known) diameter. For the purpose of illustration, we consider 3 different cases; (i) d_p=100nm and T_{air}=300K, (ii) d_p=50nm and T_{air}=300K, and (iii) d_p=50nm and T_{air}=1200K.

The morphology of the aggregates is quantified by using fractal dimensions, D_f. The agglomeration methodology prevents restructuring during the simulation and fractal aggregates are obtained. Moreover, it is found that the fractal dimensions of the aggregates with more than 10 particles obey the power law.

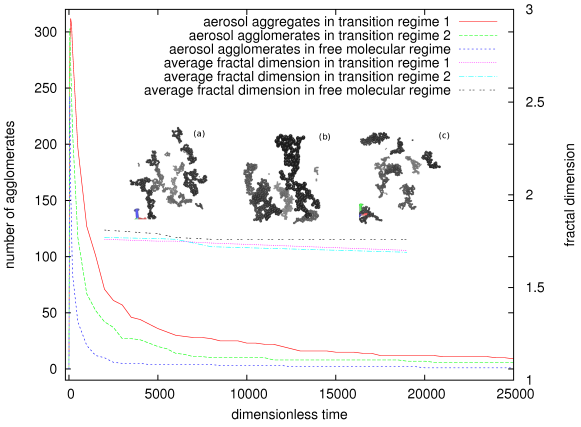

Figure 1: Evolution of aerosol aggregates, change of fractal dimension and aggregates (a) in the transition regime Kn=1, (b) in the transition regime Kn=2, and (c) in the free molecular regime.

Figure 1 shows that the fractal dimensions of the aggregates decrease as the aggregates grow in both transition and free molecular regimes. In the free molecular regime, a regime encountered for example at high temperatures, particles move fast, therefore the collision probability is high. Aggregate growth is more rapid in the free molecular regime than in the transition regime, however, aggregates that are formed in the free molecular regime are more compact. The average fractal dimensions of the aerosol aggregates at different times but possesing similar aggregate distributions are $D_f = 1.69$ and $D_f = 1.7$ in the transition regime ($Kn = 1$, $Kn = 2$) and $D_f = 1.81$ in the free molecular regime. These results agree well with other numerical and experimental studies, and the typical aggregates as shown in the inlet of Fig. 1 stronlgy resemble in shape and structure real aggregates of e.g. soot (Flagan, 1993). Further studies will investigate the aggregate dynamics in different flow fields ranging from simple shear flows to fully turbulent flows.

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