The Friction Coefficient of Fractal Aggregates in the Continuum and Transition Regimes

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Abstract: A methodology is introduced for friction-coefficient calculations of fractal-like aggregates that relates the friction coefficient to a solution of the diffusion equation. Synthetic fractal aggregates were created with a cluster-cluster aggregation algorithm. Their fiction coefficients were obtained from gas molecule-aggregate collision rates that were calculated with the COMSOL Multiphysics software. Results were compared and validated with literature values. The effect of aggregate structure on dynamical properties of the aggregate, in particular mobility, was also studied. Both the fractal dimension and the fractal prefactor are required to characterize fully an aggregate.

Keywords: Friction coefficient, fractal aggregate, collision rate, mobility radius.

1. Introduction

Aerosol and colloidal particles form complex structures that are fractal-like, their geometry obeying the fractal scaling law

$$N = k_f \left(\frac{R_g}{R_1}\right)^{d_f} \qquad (1)$$

where N is the number of primary particles (monomers, taken to be spheres), k_f the fractal prefactor, d_f the fractal dimension, R_g the radius of gyration, and R_1 the monomer radius. The study of their dynamical properties is very important as aerosol particles contribute to climate change (via the so-called direct and indirect effects) and as they have been associated with adverse health effects (Giechaskiel et al., 2009). In this study, we calculate the friction coefficient of fractal aggregates by relating it to a gas molecule-aggregate collision rate (Isella and Drossinos, 2011). Accordingly, friction coefficients are obtained from the solution of the Laplace equation instead of the solution of the Stokes equations. Moreover, we investigate the effect of morphology (structure) of a fractal aggregate on its dynamical properties, namely the aggregate friction coefficient, and the hydrodynamic and mobility radii.

2. Theory

In the momentum-transfer continuum regime (where the Knudsen number, defined as the ratio of the gas mean free path λ to the monomer radius R_1, $Kn = \lambda/R_1$, is zero) the Stokes friction coefficient of a N-monomer aggregate is

$$f_N = \frac{1}{B_N} = 6\pi\mu R_h \qquad (2)$$

where B_N is the mechanical mobility, μ the gas dynamic viscosity, and R_h the hydrodynamic radius. Experimental studies (Keller et al., 2001) found that the product of the collision rate between gas molecules and an aggregate (K_N) times the aggregate mechanical mobility is approximately constant, independent of particle material, shape and size. Hence, using the Stokes friction expression, Eq. (2), the ratio of the collision rate to the aggregate friction coefficient is constant. Therefore, the ratio of the friction coefficients of two aggregates (herein taken to be a monomer and an aggregate composed of N monomers) can be related to the corresponding ratio of collision rates,

$$\frac{f_N(0)}{f_1(0)} = \frac{K_N(0)}{K_1(0)} \qquad (3)$$

From Eq. (3) we infer that the friction coefficient in the continuum regime can be calculated by solving the steady-state diffusion equation (with appropriate boundary conditions), since the molecule-aggregate collision rate is

$$K_N = \int_s \boldsymbol{J} \cdot \hat{\boldsymbol{s}}\, dS \qquad (4)$$

where $\boldsymbol{J}$ is the molecular diffusive flux towards the aggregate, $\hat{\boldsymbol{s}}$ the outwards pointing unit vector perpendicular to S, and dS the surface element. The molecular diffusive flux is obtained from the diffusion equation (since it is related to the gradient of the molecular density $\boldsymbol{J} = -D\boldsymbol{\nabla}\rho$, with *D* the molecular diffusion coefficient). The boundary conditions appropriate for collision rate calculations are Dirichlet conditions: dimensionless gas density (ρ) on the aggregate surface zero (absorbing boundary, perfect sticking upon collision, neglect of multiple scattering events) and far away from it unity.

This methodology can be extended to the momentum-transfer transition regime for small Knudsen numbers, in particular to the slip-flow regime where the Knudsen number is $0<\mathrm{Kn}\leq 0.2$). As in the case of other transfer processes in slip flow, the boundary condition on the aggregate surface becomes a Robin condition (radiation boundary condition). In this case, the ratio of the aggregate friction coefficient in the continuum regime to the friction coefficient at a fixed (small) Knudsen number is related to the Cunningham correction factor. Thus, the Cunningham factor equals the corresponding ratio of collision rates

$$C(N, Kn) = \frac{f_N(0)}{f_N(Kn)} = \frac{K_N(0)}{K_N(Kn)} \qquad (5)$$

The appropriate boundary conditions for the solution of the diffusion equation in slip flow are that the (dimensionless) gas density far away the aggregate remains 1 but on the aggregate surface it becomes

$$\rho(R_1) = \alpha(Kn)\frac{d\rho}{dr} \qquad (6)$$

The analytical solution of the steady-state diffusion equation with these boundary condition is

$$\rho(r) = \rho_\infty \left[1 - \frac{1}{1+\frac{\alpha}{R_1}}\frac{R_1}{r}\right] \qquad (7)$$

and, thus,

$$C(1, Kn) = \frac{f_1(0)}{f_1(Kn)} = \frac{K_1(0)}{K_1(Kn)} = 1 + \frac{\alpha}{R_1} \qquad (8)$$

Given a Knudsen number, Eq. (8) is used to determine the unknown proportionality constant in the boundary condition Eq. (6). For the calculation of the Cunningham factor we use the formula proposed by Davies (1945)

$$\mathrm{C}(1, \mathrm{Kn}) = 1 + \mathrm{Kn}(1.257 + 0.4\exp(-\frac{1.1}{\mathrm{Kn}}) \qquad (9)$$

3. Use of COMSOL Multiphysics

We created the fractal aggregates, which were subsequently used in our numerical simulations, by the cluster-cluster aggregation (CCA) algorithm proposed by Thouy and Jullien (1996). The synthetic fractal aggregates generated by the CCA algorithm have prescribed fractal dimension and fractal prefactor. Moreover, the spherical monomers do not overlap. We implemented the algorithm using the software Matlab. The structures we generated had a maximum of 64 spheres, with different d_f, k_f. We inserted the generated structures into COMSOL with the tool Livelink.

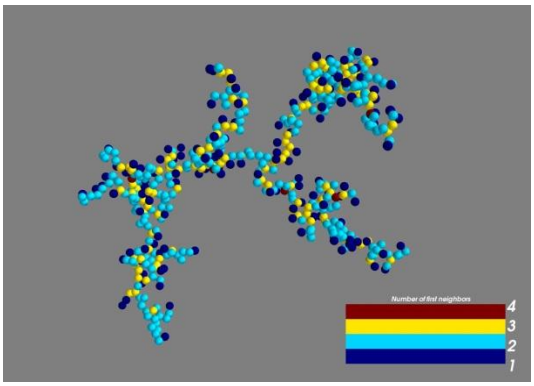

Figure 1. A typical fractal aggregate generated with the CCA algorithm (N=512, df=1.8, kf=1.3).

Our simulations start by creating an external sphere that includes the structure under study. Its radius is at least 10^2 larger than the radius of gyration of the fractal. The collision rate K_N is determined by calculating the steady-state molecular diffusive flux over the aggregate surface. Accordingly, we solve the (3d) partial differential equation

$$-c\nabla^2\rho = 0 \qquad (10)$$

with COMSOL Multiphysics. Care was exercised to use the necessary mesh to limit numerical errors. We choose Dirichlet boundary conditions (continuum regime). We define the gas density as 0 on the primary spheres of the aggregate and 1 on the outside sphere. After solving the PDE, Eq. (10), we perform a surface integration of the diffusive flux over the aggregate surface to determine the molecule-aggregate collision rate, cf. Eq. (4).

Figures 2 & 3 show typical outputs of a COMSOL Multiphysics simulation. Figure 2 presents how the gas concentration changes around an 8-monomer structure, while Figure 3 shows the corresponding diffusive flux. Note that the diffusive flux towards the outer spheres is larger, since these spheres are more exposed to gas molecules, whereas the flux towards inner monomers is smaller as they are more shielded.

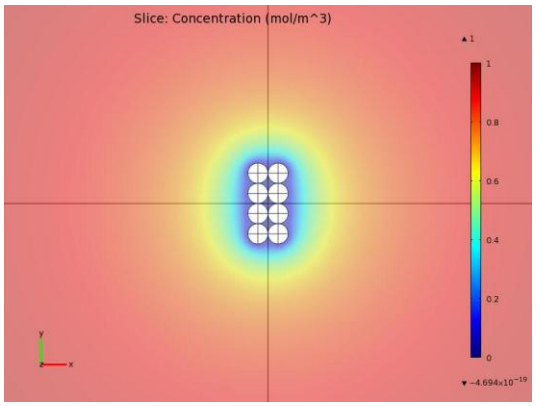

Figure 2. Gas concentration around an 8-monomer cluster (COMSOL Multiphysics).

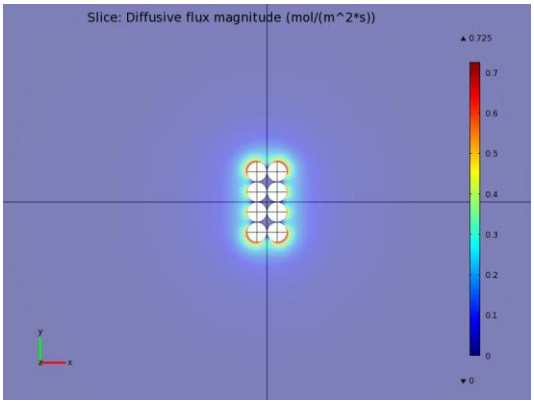

Figure 3. Diffusive flux around an 8-monomer cluster (COMSOL Multiphysics).

We follow the same procedure for the friction coefficient calculations in the slip-flow regime, except that the boundary condition on the aggregate surface changes. In these calculations, we use a flux/source condition and we define α, see Eqs. (6) and (8), in the boundary absorption/impedance term.

4. Results

The proposed methodology was validated in both the continuum and slip flow regimes. In the continuum regime, we compare two different 8-monomer structures to the analytical results presented by Filippov (2000), Table 1. These structures were a cube and a rectangle (Figs. 2 and 3). The quantities presented in Table 1 are the ratios of the aggregate friction coefficient to the product of the monomer friction coefficient times the number of monomers (8 in this case), a measure of average monomer shielding within an aggregate. The agreement is very good.

Table 1: Friction-coefficient ratios determined by collision rate calculations and analytically (Filippov, 2000).

Structure (8 particles)	Filippov (2000)	Collision rate
Cube	0.290	0.293
Rectangle	0.361	0.366

The method was applied to fractal aggregates composed of up to 64 monomers. We calculated the ratio of the mobility radius, i.e., the radius of a sphere that has the same mobility as the aggregate under identical conditions, to the

radius of gyration. We found that both (d_f, k_f) and the number of monomers influence the ratio

$$\frac{R_m}{R_g} = g(N, k_f, d_f) \qquad (11)$$

Figure 4 shows this ratio calculated for various fractals, parametrized by the fractal dimension and prefactor, plotted against the number of monomers.

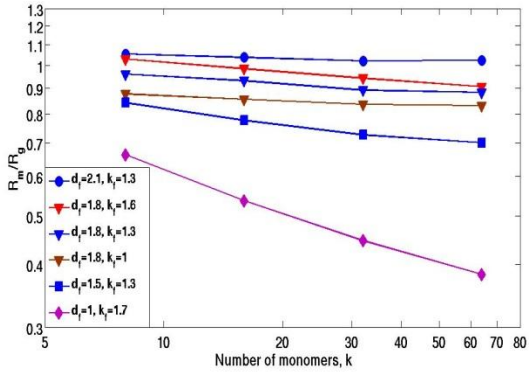

Figure 4. R_m/R_g for different fractal-like structures.

In the slip-flow regime we compare the Cunningham factor of straight chains to numerical fits (Dahneke, 1982) (Fig. 5). The Knudsen numbers were 0.1, 0.2 and 0.5. In both continuum and slip-flow regime the agreement between collision-rate predictions and numerical fits (interpolations) is very good.

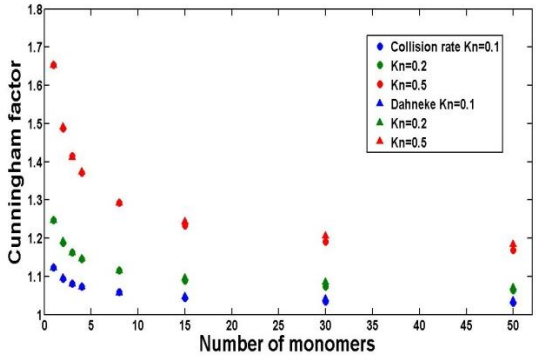

Figure 5. A comparison of the Cunningham factor of straight chains calculated via collision rate simulations and numerical fits (Dahneke, (1982).

5. Conclusions

A methodology was introduced to calculate the friction coefficient of fractal aggregates in the momentum-transfer continuum and slip-flow regimes by solving the Laplace equation (a diffusion equation). The required calculations of the collision rate between a gas molecule and an aggregate were performed with the finite-element software COMSOL Multiphysics. We found an excellent agreement of our results for the fractal friction coefficient with previously-published literature values. In addition to the friction coefficient, the aggregate mobility radius, namely the radius of a sphere with the same mobility as the aggregate under identical conditions, was calculated. This radius was related to the aggregate morphology. Both the fractal dimension and the fractal prefactor were found to influence the dynamic behavior (mobility) of the synthetic, fractal-like aggregates used in this work.

6. References

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