Differentiating between sintered and non-sintered aggregates

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Through light-scattering simulation we investigate whether measurement of the extinction spectrum can be used to characterize the morphology of sintered aggregates and whether we can distinguish between sintered and non-sintered aggregates.

INTRODUCTION

Methods for the quantitative assessment of the sintering morphology of aggregates are of recent interest in aerosol science. If aggregates are sintered, mechanical stability is improved. Additionally with conducting aggregates electrical conductivity is improved.

There are various experimental methods to characterize the state of sintering of aggregates that are based on impaction experiments [1]. If an aggregate hits a plane it will break into a number of subcomponents depending on whether the primary particles are sintered together or not. In this way one can distinguish soft and hard aggregates. An optical method for characterization would be of advantage because it is noninstrusive and could be used in process control.

Optical methods for the characterization of aggregates of nanoparticles commonly involve scattering measurements at multiple wavelengths. Using light-scattering simulation we investigate whether spectrally resolved scattering can be used to differentiate between sintered and non-sintered titania (TiO_2) aggregates.

SINTERING

In order to generate aggregates of spheres on the computer, a Diffusion Limited Aggregation (DLA) algorithm has been used [2]. The algorithm starts with a fixed particle in the origin of the coordinate system. Additional particles are then positioned individually on a starting radius and then start a so-called random walk to model Brownian motion. If a particle hits the first particle or the growing cluster it will stick and a new particle will start is walk. In this way realistic aggregates consisting of 24 primary particles are produced.

Next some sintering method is needed to produce sintered aggregates on the computer. The morphological transformation of an aggregate during sintering is driven by minimization of the free energy of the aggregate by surface reduction. To model this process we use a

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phenomenological method. This phenomenological method is based on the Metaball algorithm [3]. In the Metaball algorithm all primary particles are replaced by a potential function positioned at the centre of the every particle. The strength of the potential field decreases with the distance to the centre of the particles. The potential functions of neighboring primary particles overlap. Next a threshold is introduced in such a way that volume will be generated if the total potential field is higher than this threshold. The exact value of the threshold is chosen such that the generated boundary resembles a sintered aggregate. Additionally some small amount of shrinkage is introduced such that the total volume of the sintered aggregate is kept constant. To characterize the degree of sintering by a suitable parameter the ratio of the surface area of the sintered to the surface area of the non-sintered aggregate is used A/A_0.

For the sample computation we use the following parameters, radius of primary particles r = 25 nm, fractal dimension D_f = 1.8, number of primary particles N = 24. Fig. 1a gives the shape of a non-sintered DLA aggregate. An exemplary figure of the three dimensional shape of a sintered aggregate is shown in Fig. 1b. This aggregate has an intermediate sinter parameter A/A_0 = 0.86.

Figure 1. a) Shape of DLA aggregate, r = 25 nm, D_f = 1.8, N = 24. **b)** Shape of sintered aggregate with sinter parameter A/A_0 = 0.86.

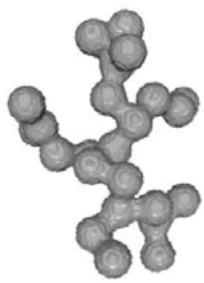

Figure 2. DDSCAT shape data of sintered aggregate of Fig. 1b.

LIGHT SCATTERING COMPUTATION

Next we present some computational results of the extinction spectra of sintered and non-sintered aggregates. For the aggregate material we use titania. The refractive index of titania used for the scattering computations is plotted in Fig. 3.

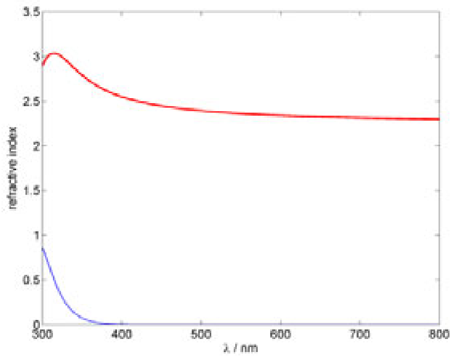

Figure 3. Refractive index of titania. Real part: red, imaginary part: blue.

To compute light scattering, DDSCAT has been applied. For the discretization a grid spacing of 3nm has been used. The input DDSCAT shape data used for the sintered aggregate is visualized in Fig 2. DDSCAT has been validated by comparing to results computed via the NFM-DS [4] for an aggregate of titania spheres. In Fig. 4 the orientationally averaged scattering efficiency of the sample aggregate with two degrees of sintering is plotted. One can see that depending on the degree of sintering the maximum is shifted to shorter wavelength.

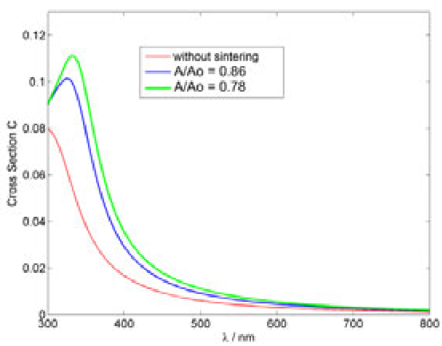

Figure 4. Computed extinction cross section for a non-sintered and a sintered aggregate with sinter parameters $A/A_0 = 0.86$ and 0.78.

In Figure 5 the position of this maximum is plotted versus the sintering parameter A/A_0. One can see that there is an almost linear relation between the position of this maximum and the sintering parameter.

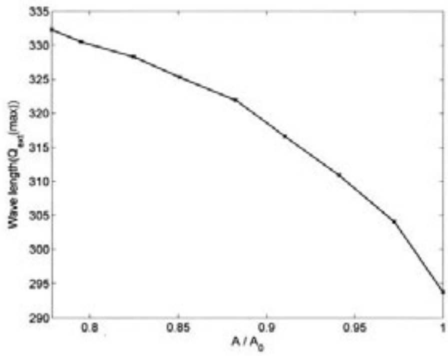

Figure 5. Position of maximum in computed extinction cross section versus sintering parameter.

CONCLUSION

Using light-scattering simulation the spectral extinction of sintered and non-sintered aggregates of titania has been investigated. There is a shift in the extinction curves such that we hope to be able to develop a spectral method for characterization of the state of sintering of aggregates.

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