Total simulation of meso-scale formation in coating process for colloidal solutions

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1. Introduction

Thin films of nanoparticles fabricated by coating and drying processes of colloidal solutions [(1)] have been studied because of their potential applications such as electronic, optical and magnetic devices. We have developed a total simulator named SNAP (Structure formation of NAno-Particles) which can simulate the dynamics of meso-scaled structure formation under still standing, coating flow and drying for colloidal solutions. SNAP based on a governing equation set consisting of both particle and fluid momentum equations can predict three-dimensional spatio-temporal structures under both equilibrium and non-equilibrium conditions. In other words, the meso-scaled structures can exhibit self-organized structure formation under specific conditions due to particle-particle and particle-fluid interactions for condensed colloidal solutions.

There are a lot of problems relating to condensed colloidal solutions such as aggregation, gelation, fouling, shear-thinning and shear-thickening. Besides such process problems, we want to predict the final structures after drying because the desired functions strongly depend on the structure. It is concluded that the design of meso-scaled structures of nanoparticles are indispensable for creating sophisticated functional materials and devises.

2. Total simulation

We have developed a total simulator, SNAP, to understand the mechanisms of meso-scale structures of nanoparticles for fabricating functional materials and devices. A lot of problems appear in coating processes. For instance, dispersion-aggregation of colloidal soltutions, gelation in filtering of condensed colloidal solutions and final structures of nanoparticles in drying should be well understood. It is desired to develop a general and fundamental simulator which can be applied to each different process. SNAP-L [(2),(3)] (Liquid) and SNAP-F[(4)] (Fluid) are separately developed and merged soon. SNAP-L basically handles free surfaces appearing in spray drying, ink jet and drying after coating. In the case of drying, a capillary force due to the deformation of free surfaces because of the interaction between particles and free surface plays a crucial role for meso-scale structure formations. SNAP-L dose not solve the fluid motion. On the other hand, SNAP-F can solve the fluid motion with particle motions for condensed colloidal solutions. SNAP-LF as

shown in Figure 1 can predict the meso-scaled structures appeared in coating processes. Since SNAP focuses on the dynamics of meso-scaled structures, macroscopic flow fields must be solved by a commercial fluid simulator such as FLUENT which cannot predict the meso-scaled structures.

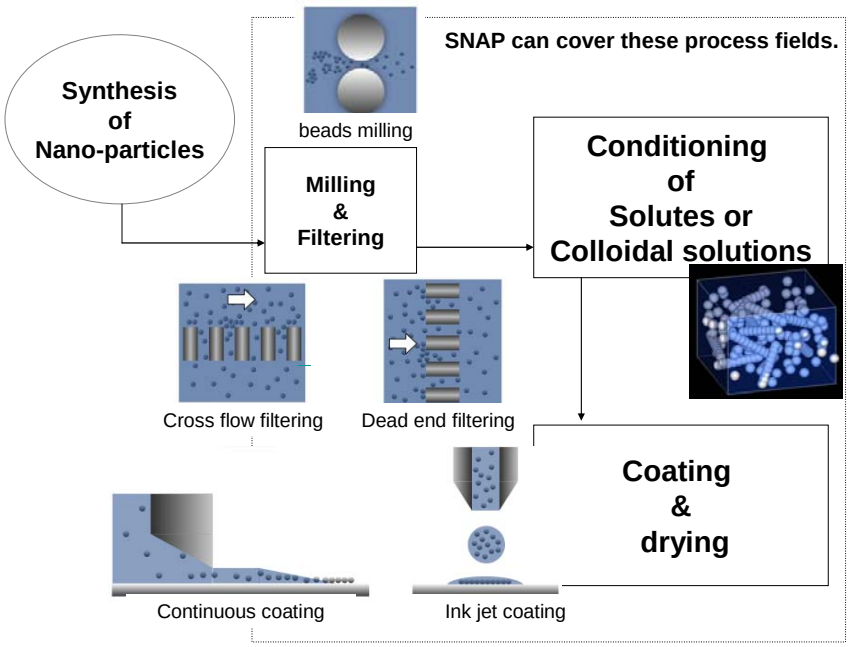

Figure 1　Total model in coating simulated by SNAP

The solutions without nanoparticles are coated and result in meso-structured thin films as schematically shown in Figure 2. The onset of nucleation takes place under super cooling. The crystal size and aggregation strongly depend on the super cooling. In general, higher cooling rate results in smaller crystal size. It is critically important to control both the thickness and characteristics of coated films. SNAP can be applied to the uniform system if a nucleation model is incorporated. However, there are few studies on nucleation in coating because of its rapid phenomena.

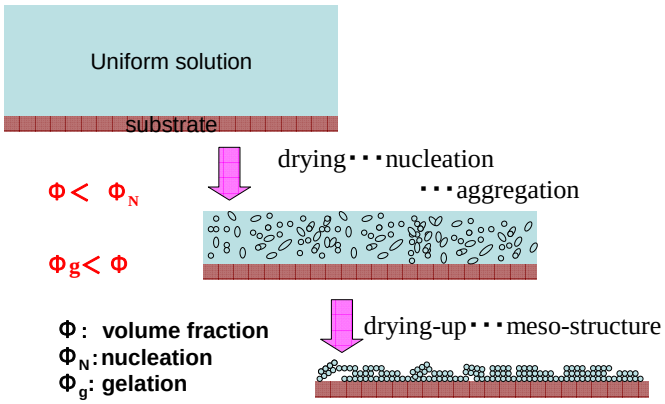

Figure 2　Schematic image of meso-scaled structure formation during drying

3. Results

(1) SNAP-L [2],[3]

Nanoparticles in a colloidal solution are subject to multiscale surface forces such as capillary force, contact force, electrostatic force, van der Waals force and friction drag as well as Brownian force and fluid drag. Both a drop and a uniform film on a substrate result in the final meso-structures shown in (c) and (f) of Figure 3 after drying. The key point to obtain ordered structures is to control the repulsive force such as electrostatic force. Rod-like particles [5] other than spherical particles are also simulated by SNAP-L.

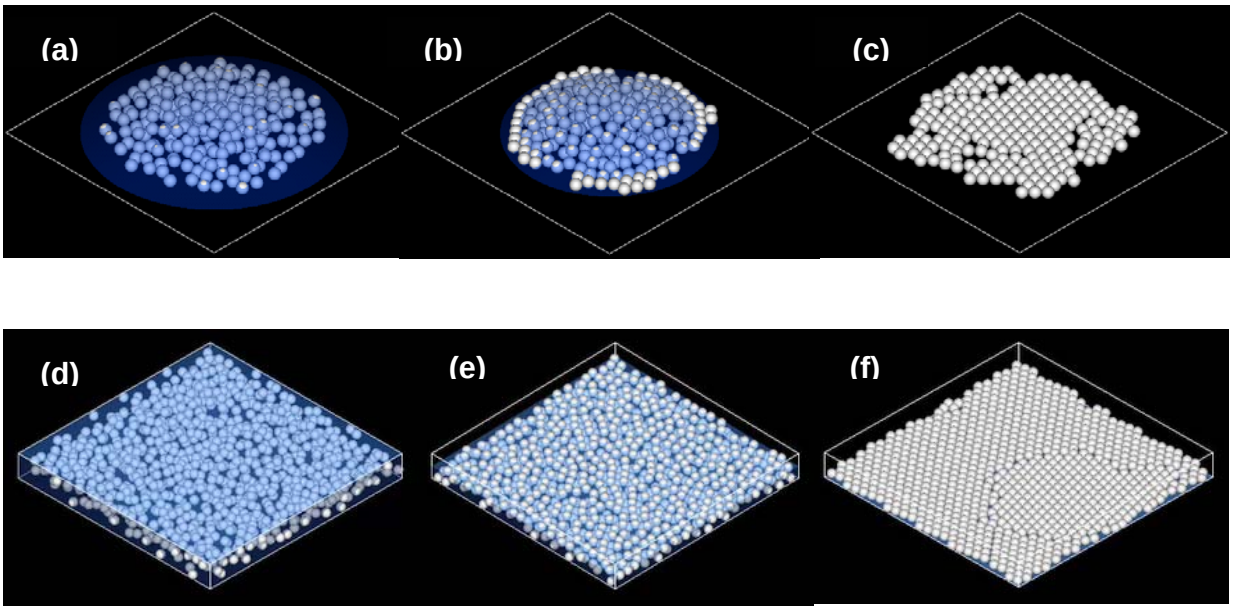

Figure 3 Meso-scaled structure formation during drying for discontinuous coating (a), (b) and (c) and continuous coating (d), (e) and (f) simulated by SNAP-L (3-dimension).

(2) SNAP-F [4]

SNAP-F solves both particle motion and fluid motion in relatively fine meshes compared with a particle size. Then, the aggregation of nanoparticles for condensed colloidal solutions can be simulated under even high shear rates as shown in Figure 4. Since the results presented here are two-dimensional, the effect of aggregation due to fluid forces is overestimated. Rheological characteristics as shear-thinning and shear-thickening with increasing shear rate can be well predicted without experimental results in Figure 5.

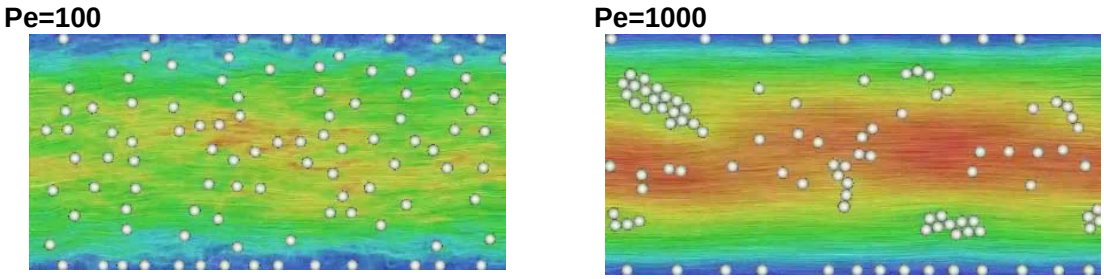

Figure 4 Meso-scaled structure formation during pressure driven flows. The zeta potentials are -10mV. *Pe* numbers are (a) 100 and (b) 1000, respectively.

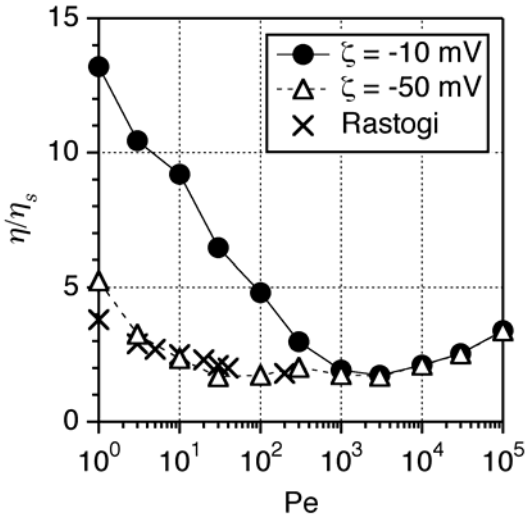

Figure 5　Viscosity changes predicted by SNAP-F (2-dimensional flow).

In Figure4 and Figure 5, *Pe* number is defined as uR^2/LD (u: velocity, R: particle radius, L: gap length, D: particle diffusivity). Apparent viscosity is summarized by initial zeta potentials and *Pe* number.

4. Conclusions

Meso-scaled structures of nanoparticles in a coating process can be predicted by a particle-fluid numerical model, SNAP. The structures of nanoparticles are responsible for the performance and life of functional materials and devices. Physical models to predict the performance of nano-materials from predicted meso-scaled structures are studied with experiments. Aggregation, gelation and nucleation must be studied for establishing nano-processing. SNAP-LF (3-dimensional version) will be developed, which is more powerful for solving the existing problems.

References

(1) S. Maenosono, T. Okubo and Y Yamaguchi, "Overview of Nanoparticle Array Formation by Wet Coating", J. Nanopart. Res. 5, 5-15　(2003).

(2) M. Fujita and Y. Yamaguchi, "Development of three-dimensional structure formation simulator of colloidal nanoparticles during drying", J. Chem. Eng. Jpn. **39**, 83-89　(2006).

(3) M. Fujita and Y. Yamaguchi, "Multiscale simulation method for self-organization of nanoparticles in dense suspension",　J. Comp. Phys. **223** (1), 108-120　(2007).

(4) M. Fujita and Y. Yamaguchi,　"Simulation model of concentrated colloidal nanoparticulate flows", Physical Review E **77** (2), 026706 (2008).

(5) Osamu Koike, Seiichi Ohta, Masahiro Fujita and Yukio Yamaguchi, “Simulation Model of Concentrated Colloidal Rod-like Nanoparticles”, J. of Japanese Applied Physics, accepted.