

Crack Formation during Drying of Colloidal Solutions

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Thin films fabricated from colloidal solutions by coating and drying processes have been used for fabricating various products. When the drying rate is increased, cracks can appear on the surface of films. Since the cracks during drying may degrade physical properties of thin films, the mechanism of crack formation has been intensively studied ^{(1),(2),(3)}. It is revealed that the onset of cracking is due to the transition to the falling drying rate period.

The reason of crack formation is to enhance the drying rate by air penetration into a concentrated particle layer (CPL) which is formed during a constant drying period. The crucial point to elucidate the cracking mechanism is to understand the fluid flow rate through CPL formed near the free surface. As CPL grows during drying, the evaporation rate reaches a critical point where the falling rate drying period starts due to the decrease of flow rate through CPL. At the same instant, the free surface is deformed for increasing the evaporation by introducing a capillary force which appeared on the free surface of CPL. The capillary force increases the drying rate. It is well known that the capillary force plays a crucial role to deform the free surface of colloidal solutions.

The evaporation of the solvent concentrates the particles into CPL. In order to study the CPL growth, we experimented pseudo-two dimensional drying of colloidal solutions sandwiched by two plates. The evolution of CPL such as menisci between two plates is measured as a function of the drying rate. The deformation of menisci builds up negative pressure just inside the vicinity of free surface. The pressure-driven flow transports particles to CPL during drying.

We also studied the evolution of CPL due to pressure-driven flow by using a numerical simulator, SNAP ⁽⁴⁾ (Structure formation of Nano-Particles). SNAP ^{(5),(6)} has been developed for simulating the dynamics of meso-scaled structure formation under still standing, coating flow and drying for colloidal solutions. We set up the simulation conditions as shown in Figure 1. Then, we succeeded to predict the concentration of particles in terms of friction coefficient and drying rate, as shown in Figure 2. CPL is realized by a numerical simulation under a given flow rate. Our simulation results are shown in Figure 3. The maximum density of CPL obtained here is roughly 55 % under zero friction. The maximum random packing is said to be 63% for mono-disperse spheres. With increasing friction coefficient, The particle density of CPL gradually decreases as shown in Figure 3.

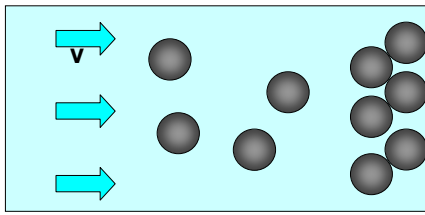


Figure 1. Simulation configuration,
The right end only allows water to go out.
The particles are trapped at the right end.

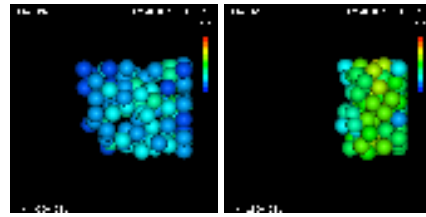


Figure 2. Simulation results. The friction coefficients are 1.0 (left), and 0.1, respectively. The friction coefficient is defined between particles.

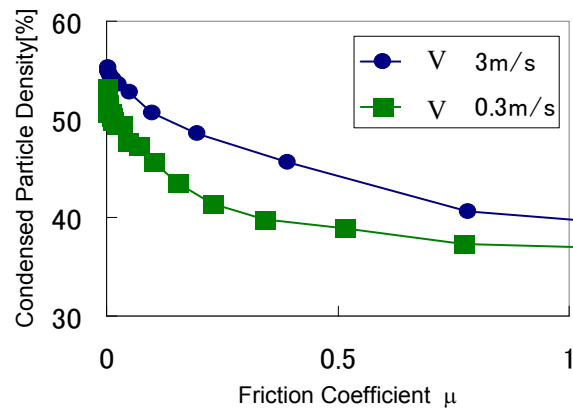


Figure 3. Particle density of CPL as functions of flow rate and friction coefficient.

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.

We elucidate the mechanism of cracking during drying. First, concentrated particle layer (CPL) appears under constant drying rate period. Second, the onset of cracking is just on the start point where falling drying rate period starts. We could predict those two critical points if physical properties such as diffusion coefficient, viscosity, friction coefficient between particles are estimated. It is concluded that the cracks take place under wet drying state.

References

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