SCEJ 51st Autumn Meeting

Sept. 25, 2020

Structure of dried colloidal films controlled by the morphology of aggregated particles

微粒子分散液の凝集状態による乾燥粒子膜の構造制御

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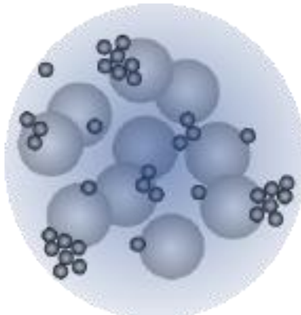
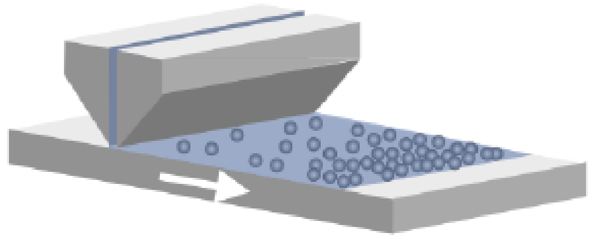
辻 佳子　（東大環安セ/東大院工）

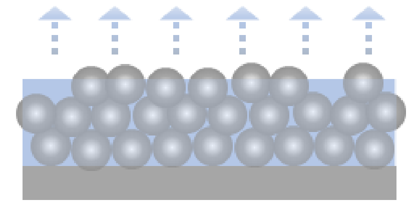
Material Fabrication from Colloidal Suspensions



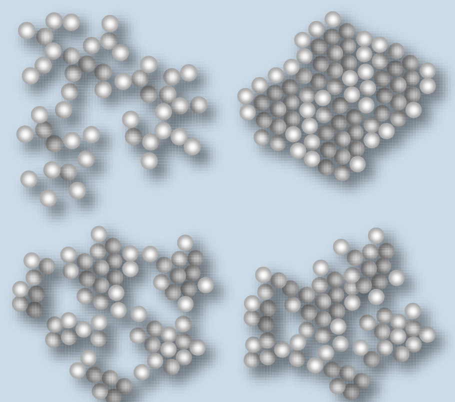
Dispersing



Coating



Drying



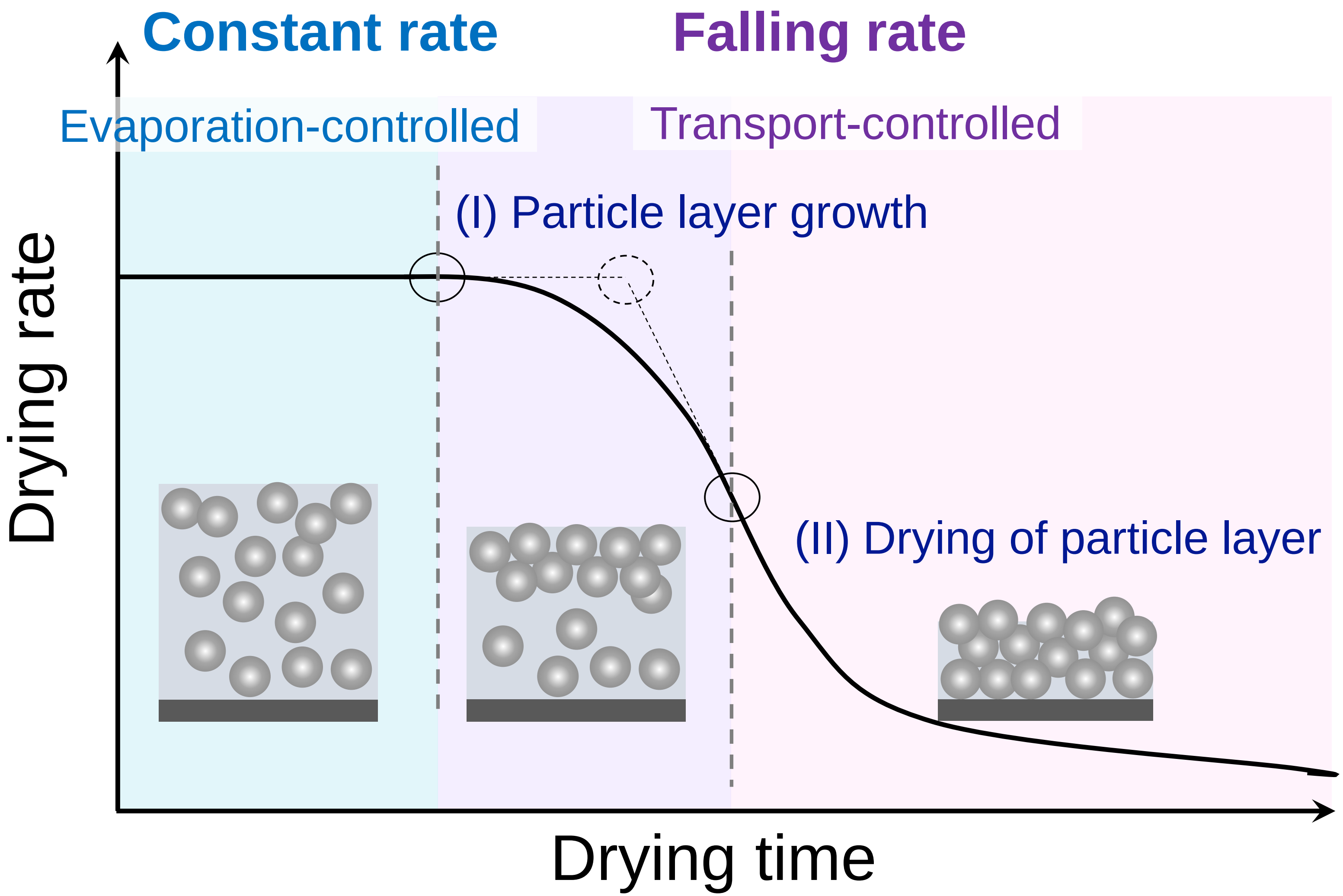
Functions

- Strength
- Permeability
- Electrical/Thermal conductivity
- Optical property

Structures

- Porosity
- Contact network

Drying Curve of Colloidal Suspensions

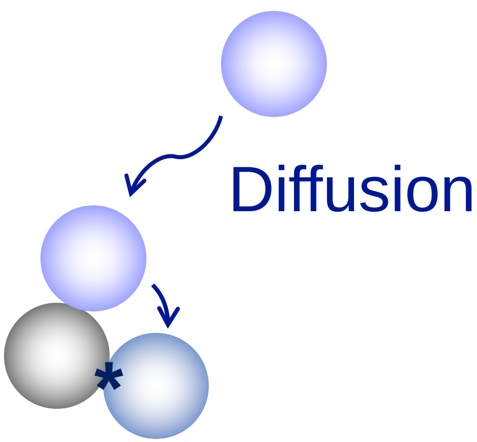


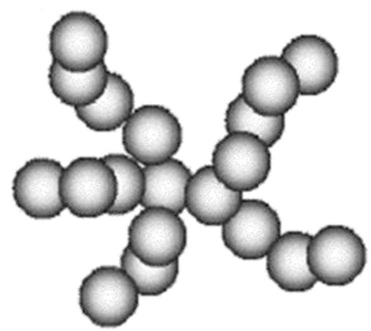
Drying rate vs. Structure

Objective

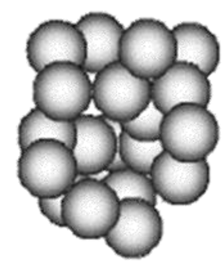
- Modeling of adhesion to describe the various morphologies of aggregated particles

- Effects of adhesion on structure formation during drying





Diffusion-limited



Reaction-limited

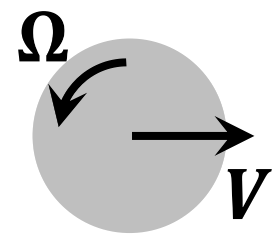
Adhesion: Fixation of contact points (Reaction)

Equations of Particles' Motion

Langevin equation

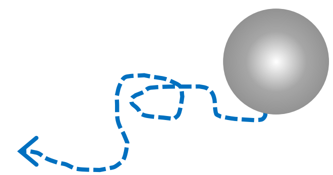
$$M\dot{\boldsymbol{V}} = -\xi\boldsymbol{V} + \boldsymbol{F}^{\mathrm{R}} + \boldsymbol{F}^{\mathrm{cnt}} + \boldsymbol{F}^{\mathrm{DLVO}} + \boldsymbol{F}^{\mathrm{cpl}}$$

$$I\dot{\boldsymbol{\Omega}} = -\zeta\boldsymbol{\Omega} + \boldsymbol{N}^{\mathrm{R}} + \boldsymbol{N}^{\mathrm{cnt}}$$

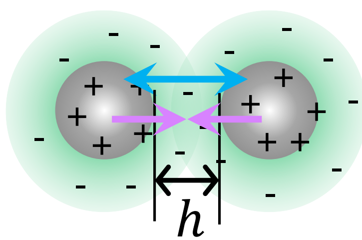


- **Hydrodynamic force/torque**

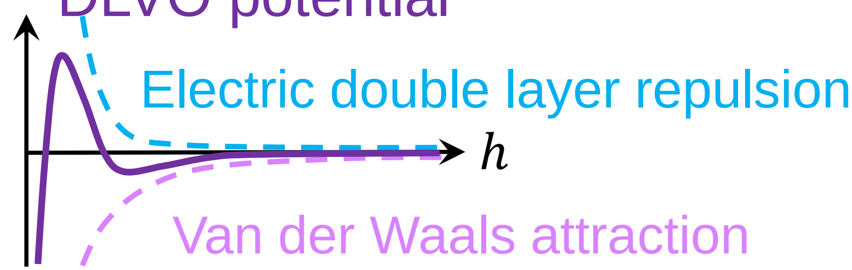
Drag + Fluctuations → Brownian motion



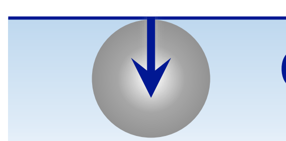
- **DLVO force**



DLVO potential



- **Capillary force**



Contact angle $\alpha = 0$

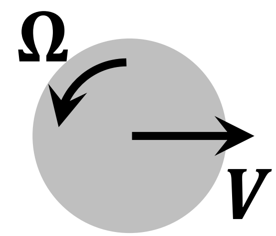
Vertical push into liquid

Modeling of Adhesion

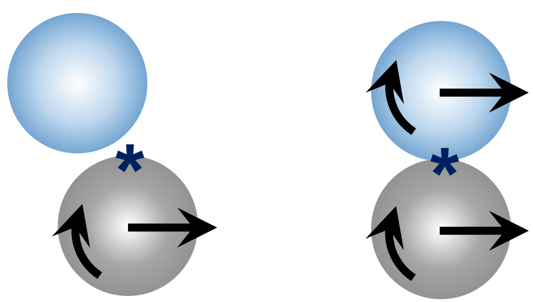
Langevin equation

$$M\dot{\boldsymbol{V}} = -\xi \boldsymbol{V} + \boldsymbol{F}^{\mathrm{R}} + \boldsymbol{F}^{\mathrm{cnt}} + \boldsymbol{F}^{\mathrm{DLVO}} + \boldsymbol{F}^{\mathrm{cpl}}$$

$$I\dot{\boldsymbol{\Omega}} = -\zeta \boldsymbol{\Omega} + \boldsymbol{N}^{\mathrm{R}} + \boldsymbol{N}^{\mathrm{cnt}}$$

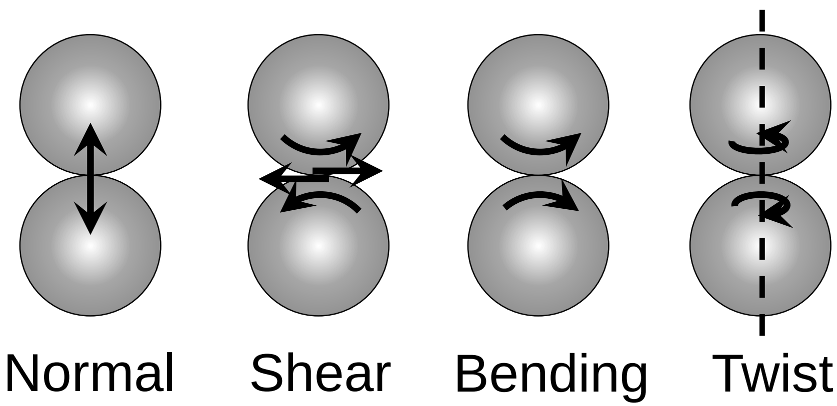


- **Contact force/torque**



Slip **Stick**

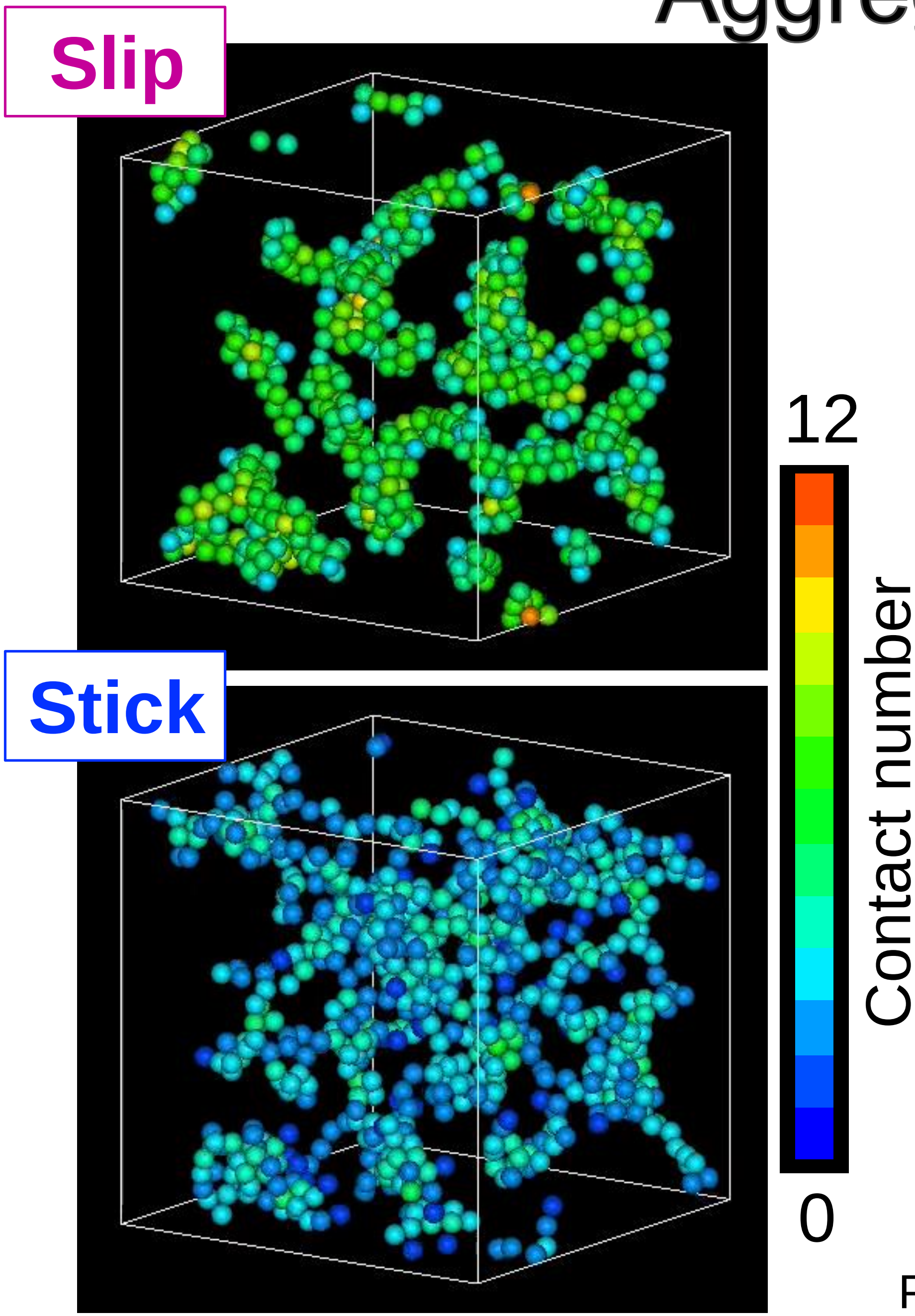
Adhesion

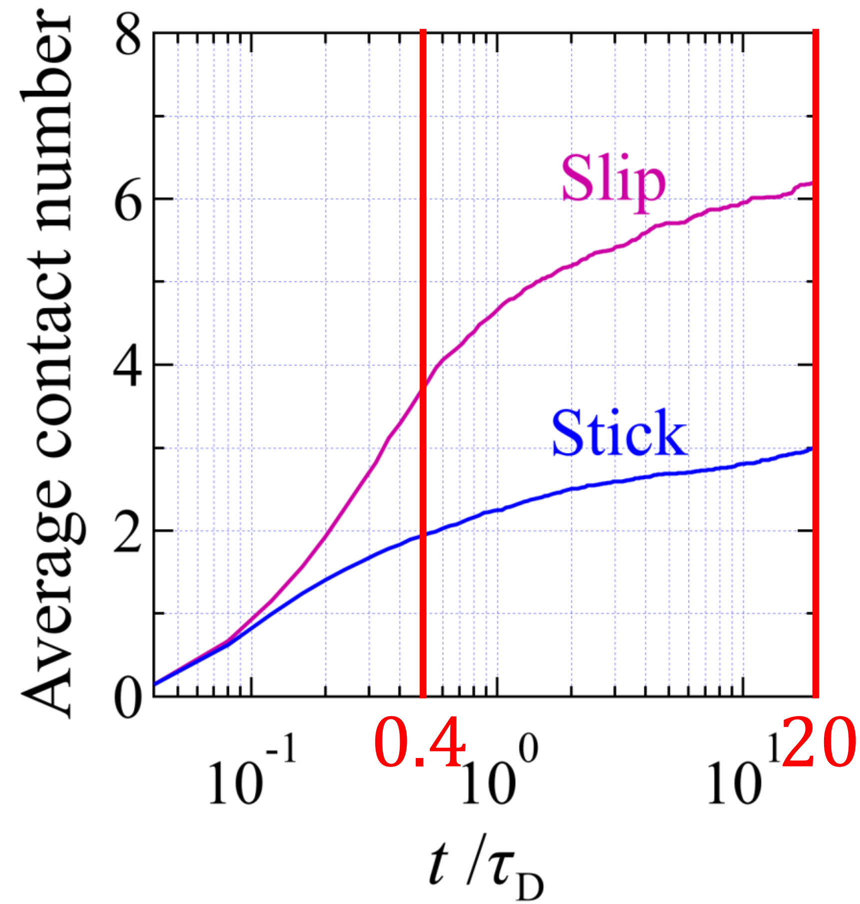


Constraint on the relative motions

Aggregation

Particle diameter: $d = 10\ \mathrm{nm}$
Zeta potential: $0\ \mathrm{mV}$
Volume fraction: $5\ \mathrm{vol\%}$



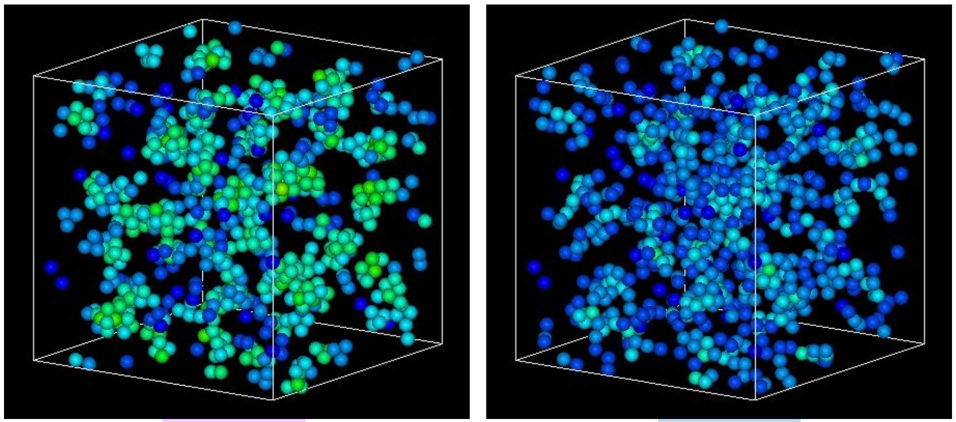


Characteristic time of particle diffusion: $\tau_{\mathrm{D}} = d^2/D$

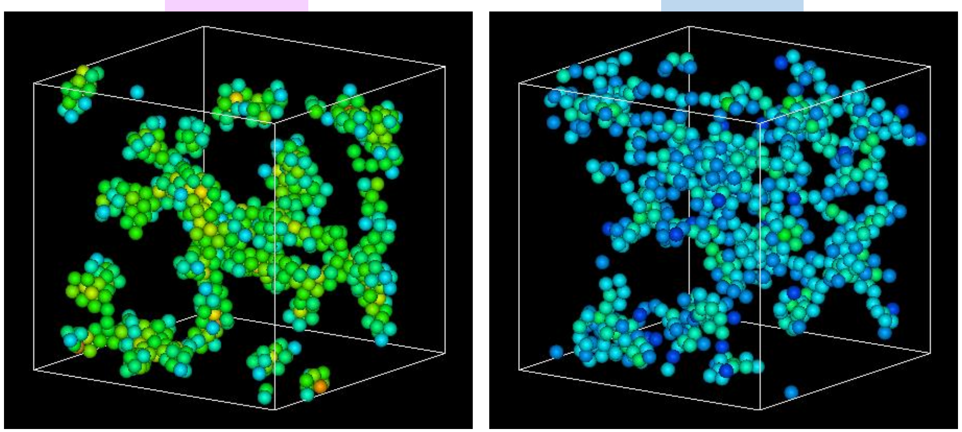
Particle diameter: d Diffusion coefficient: D

Aggregation

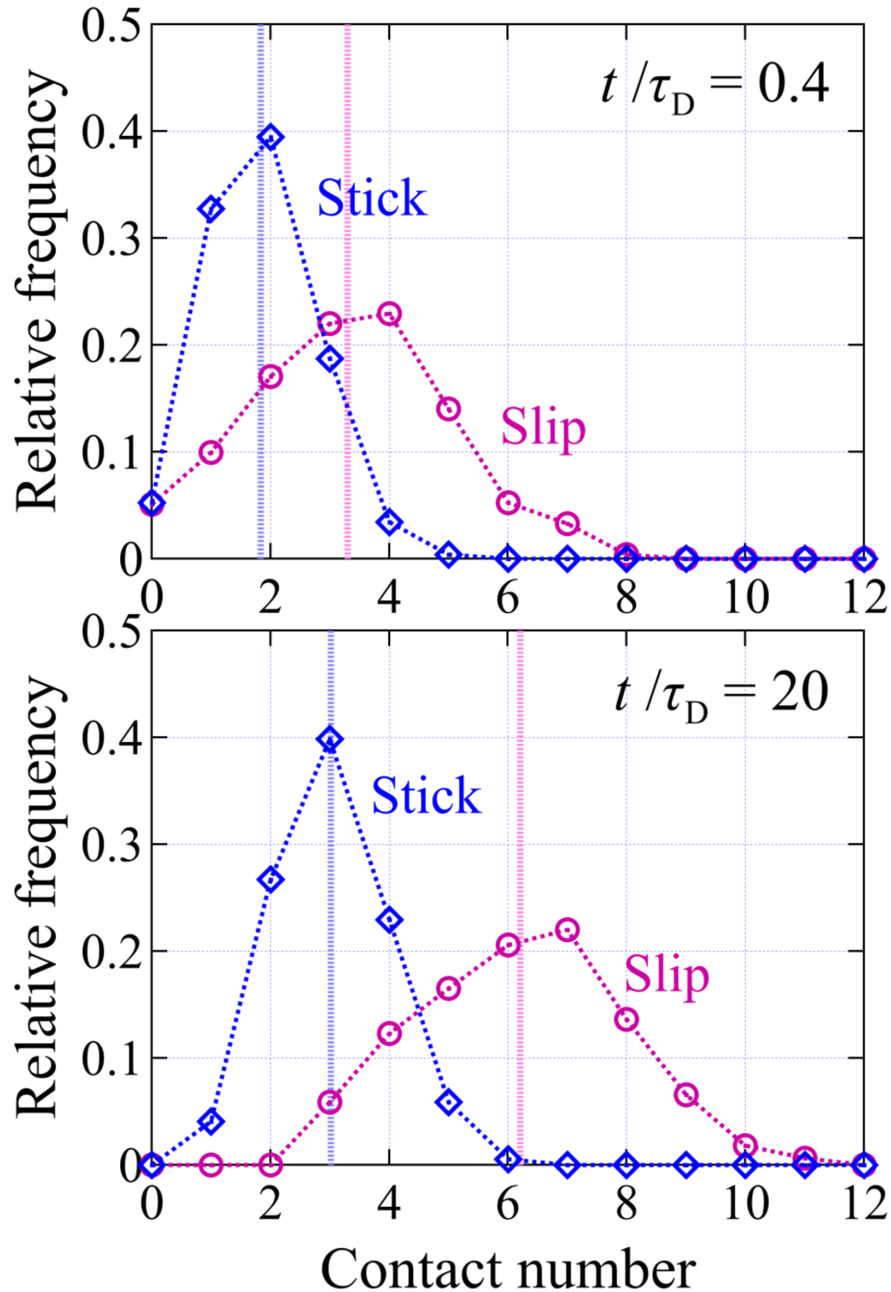
Slip **Stick**



$t/\tau_D = 0.4$



$t/\tau_D = 20$



Simulation Conditions

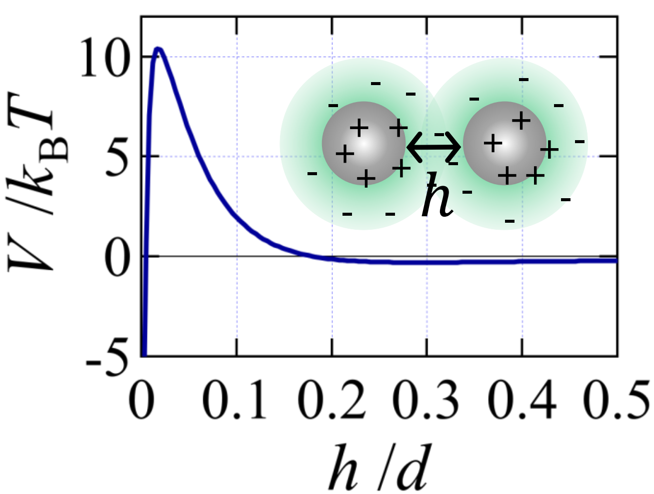
Particles

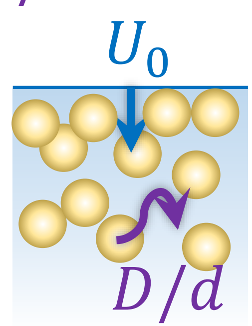
- Diameter $d = 20\ \mathrm{nm}$
- Initial volume fraction $10\ \mathrm{vol\%}$
- Zeta potential $-50\ \mathrm{mV}$
- Contact **Slip** / **Stick**

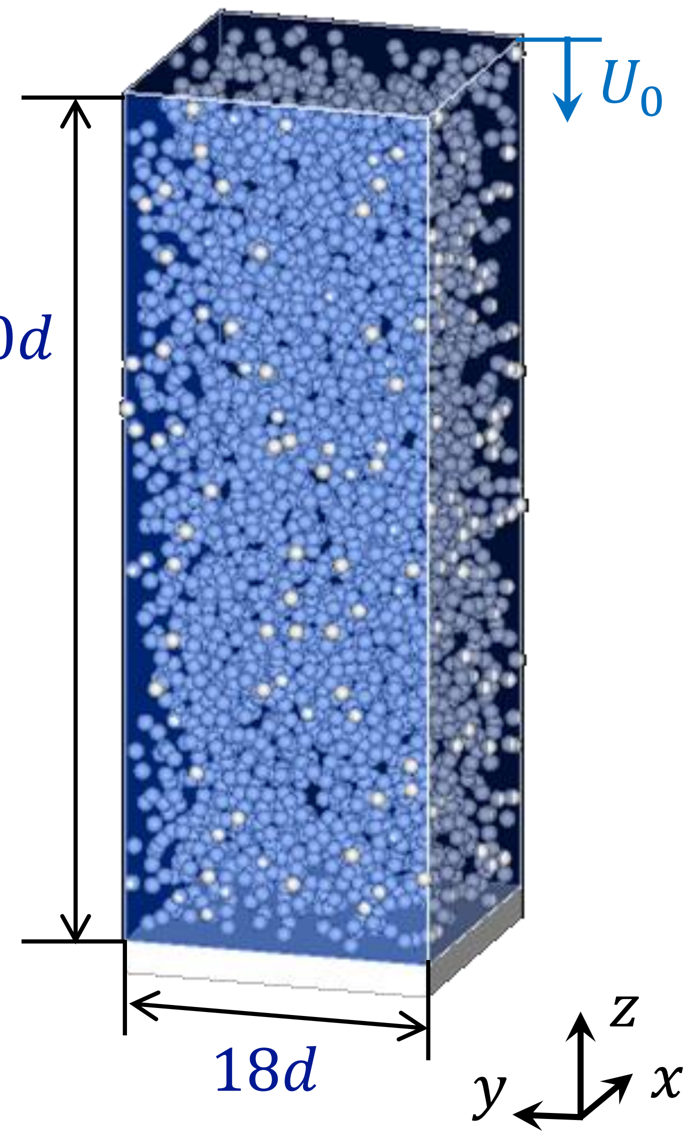
Liquid: water

- Initial particle drying Péclet number

$$\mathrm{Pe}_0 = \frac{(\text{Drying rate})}{(\text{Diffusion rate})} = \frac{U_0}{D/d} = 400$$







Periodic boundaries x, y

Modeling of Falling Drying Rate

Drying rate: $$\frac{U}{U_0} \simeq \frac{R_0}{R_0 + R_\mathrm{p}}$$

Resistance of evaporation: R_0

Resistance of particle layer: R_p

↑

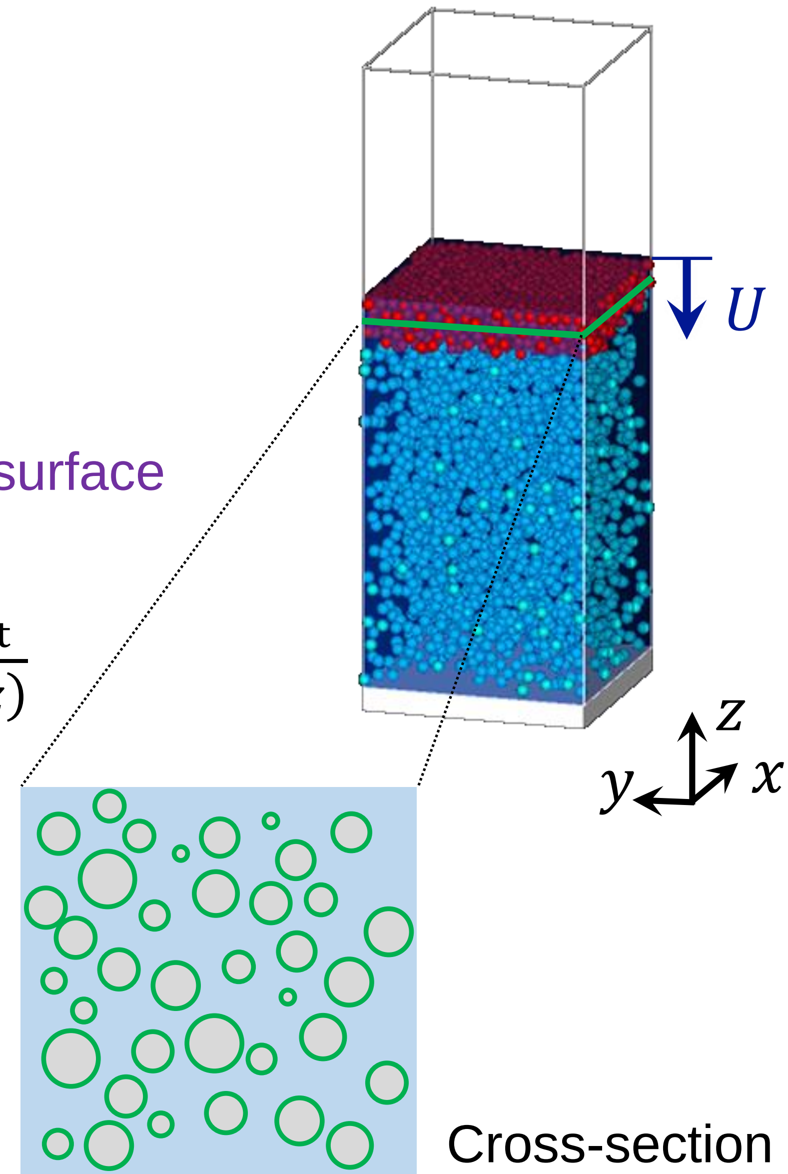
Aggregated particles moving with free surface

$$R_\mathrm{p} = \int_P r(z)\mathrm{d}z \qquad r(z) = \frac{80}{[D_\mathrm{H}(z)]^2}\frac{S_\mathrm{tot}}{S_\mathrm{f}(z)}$$

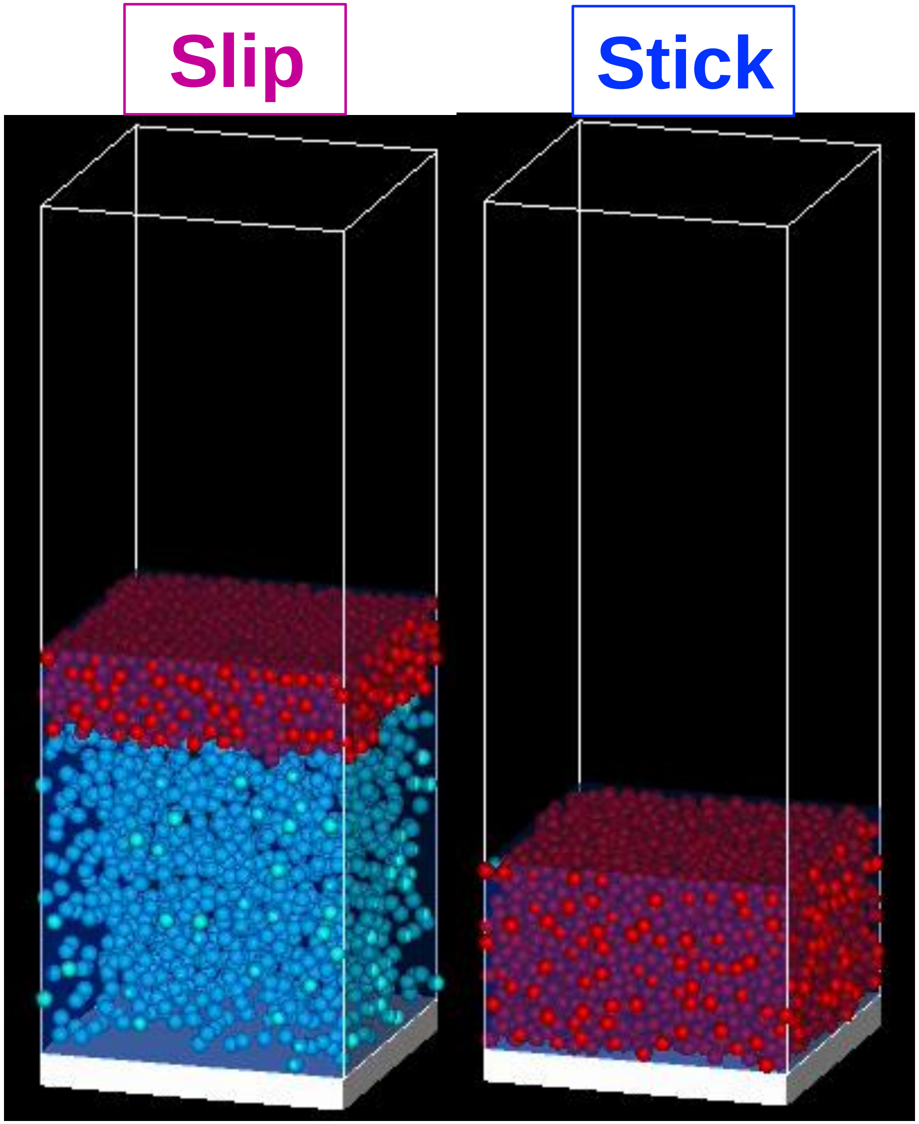
Hydraulic diameter: $D_\mathrm{H} = \dfrac{4S_\mathrm{f}}{L_\mathrm{f}}$

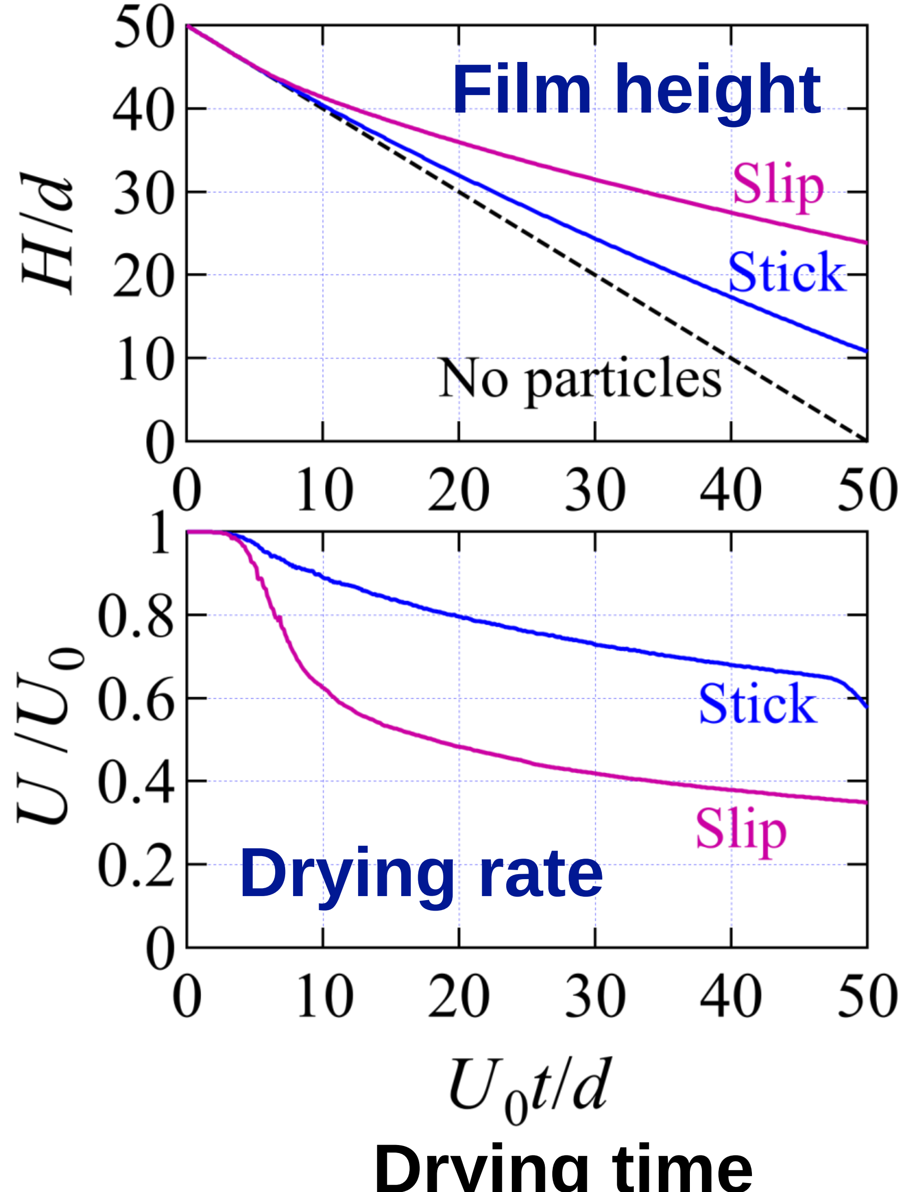
Cross-sectional area of the flow: S_f

Wetted perimeter: L_f

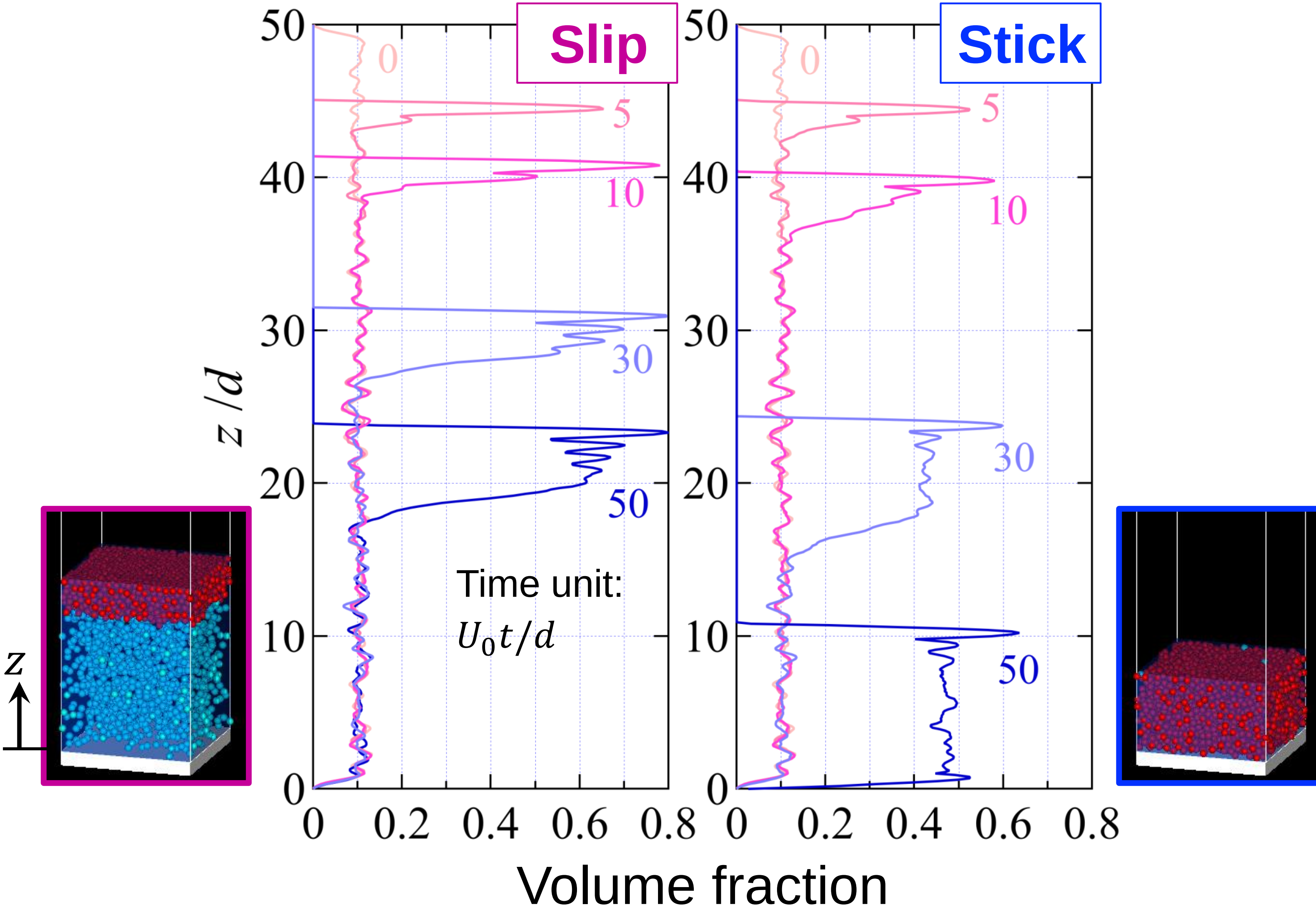


Drying Curves

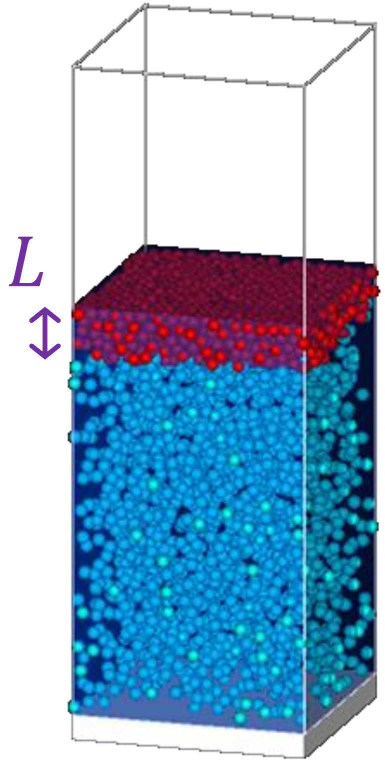


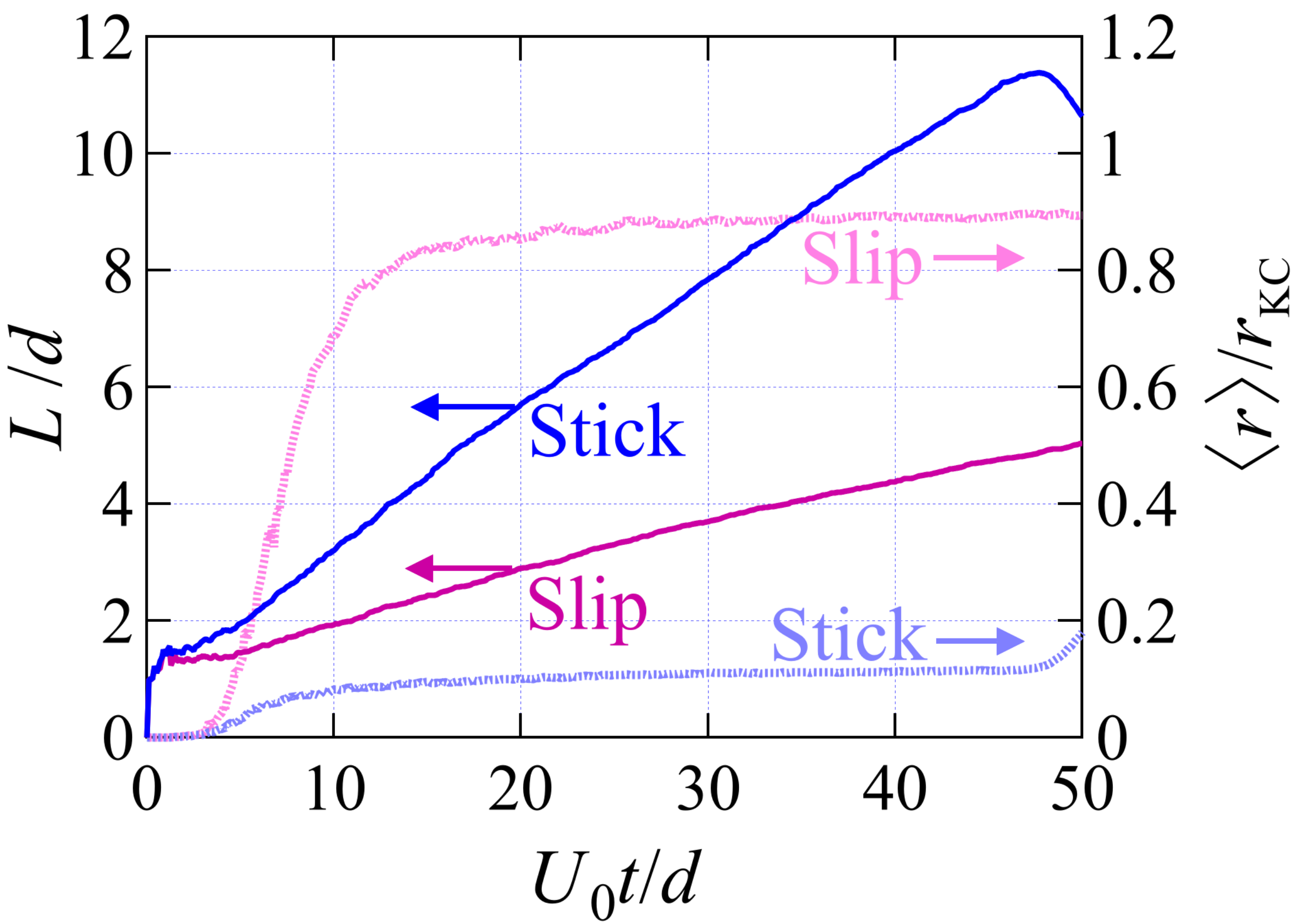


Particle Distribution



Growth of Particle Layer





Summary

- ◆ Modeling of adhesion between particles
  → Morphologies of aggregate

  - Constraint on relative motions between contacting particles
    → Fixation of contact points

  - Possible factor of adhesion in real systems: Binder addition

- ◆ Effects of adhesion on structure formation

  - Tree-like aggregates

  - Structures with high porosity formed during drying
    → High permeability (drying rate)