Mesoscale Modeling of Colloidal Films Dried with Controlling the Morphology of Aggregated Particles

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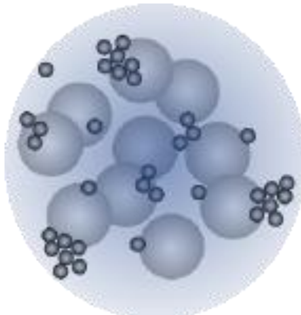
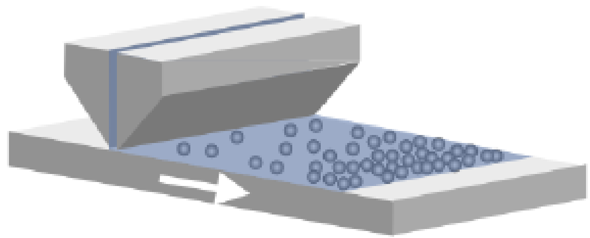
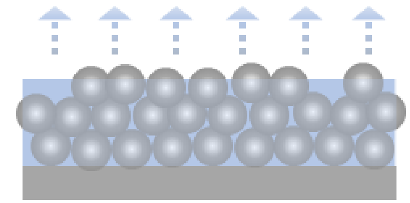
Material Fabrication from Colloidal Suspensions



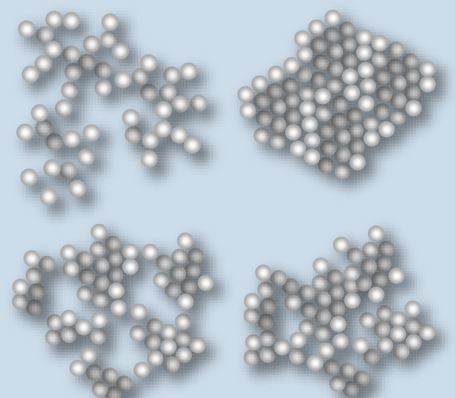
Dispersing



Coating



Drying



Functions

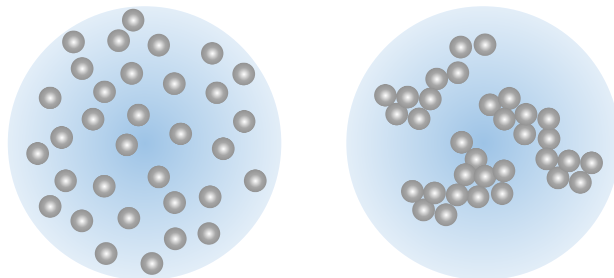
- Strength
- Permeability
- Electrical/Thermal conductivity
- Optical property

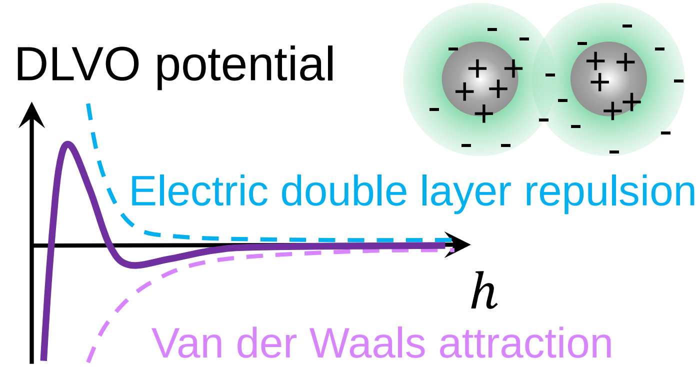
Structures

- Porosity
- Contact network

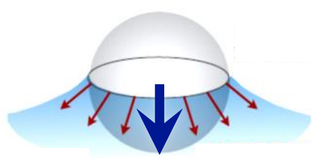
Structure Formation during Drying

Particle dispersion/aggregation

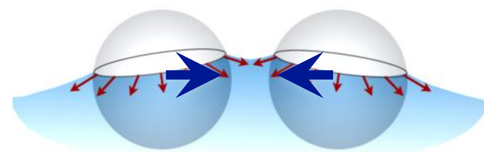




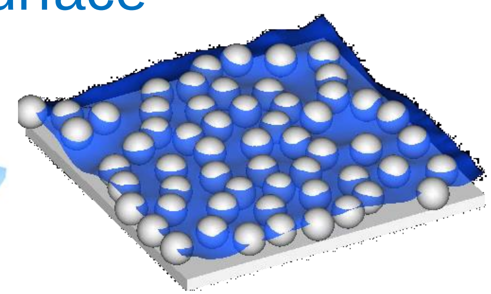
Drying: capillary force induced by free surface



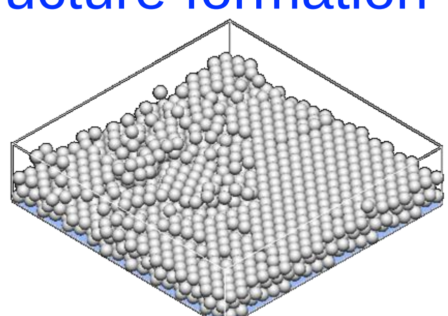
Vertical push into liquid

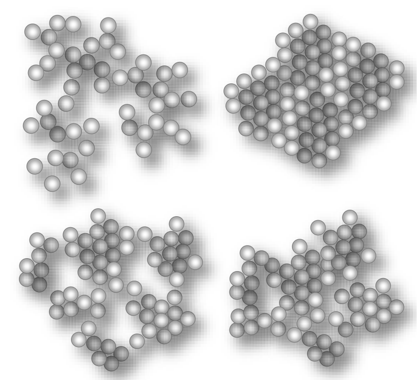


Lateral attraction



Structure formation

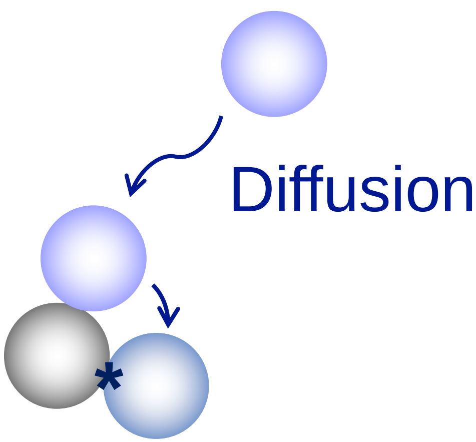




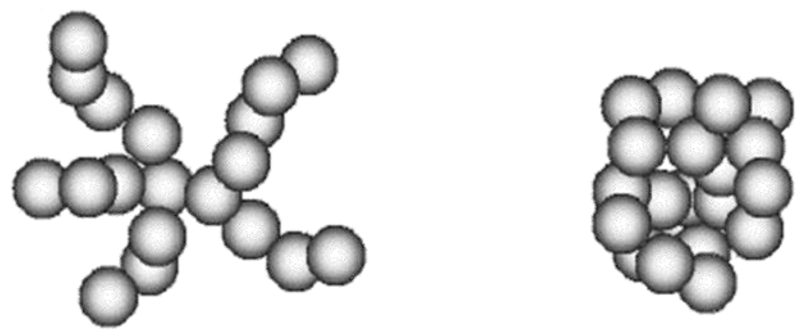
Objective

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

- Effects of adhesion on structure formation during drying



Adhesion: Fixation of contact points (Reaction)

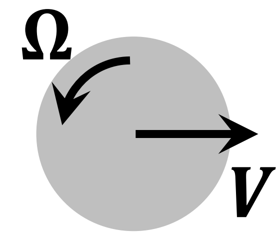


Diffusion-limited Reaction-limited

Equations of Particles' Motion

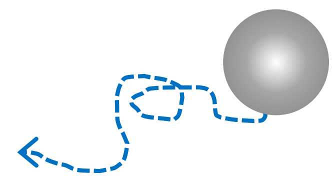
$$M\dot{\boldsymbol{V}} = -\xi\boldsymbol{V} + \boldsymbol{F}^{\mathrm{R}} + \boldsymbol{F}^{\mathrm{cnt}} + \boldsymbol{F}^{\mathrm{DLVO}} + \boldsymbol{F}^{\mathrm{cpl}}$$

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

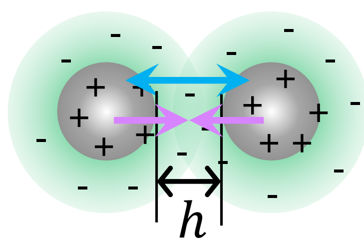


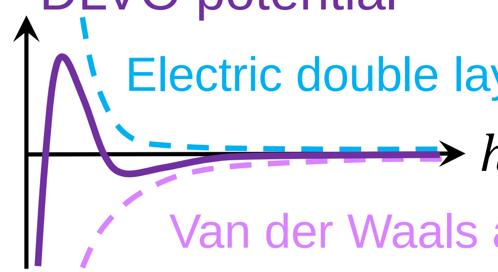
Hydrodynamic force/torque

Drag + Fluctuations → Brownian motion

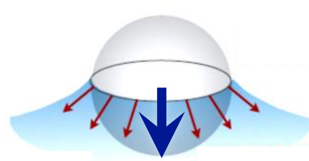


DLVO force

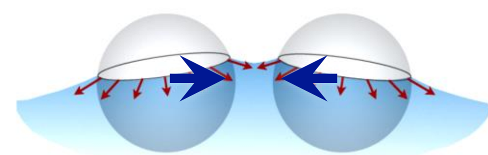




Capillary force



Vertical push into liquid

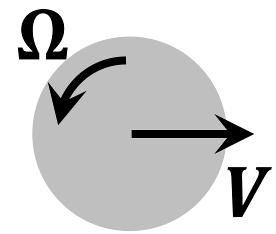


Lateral attraction

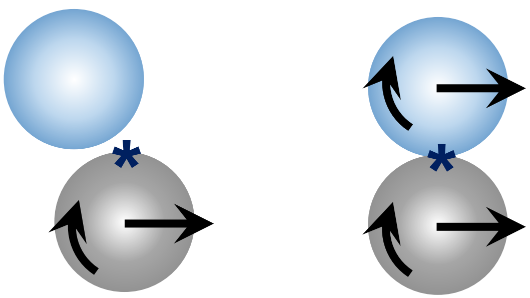
Modeling of Adhesion

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

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



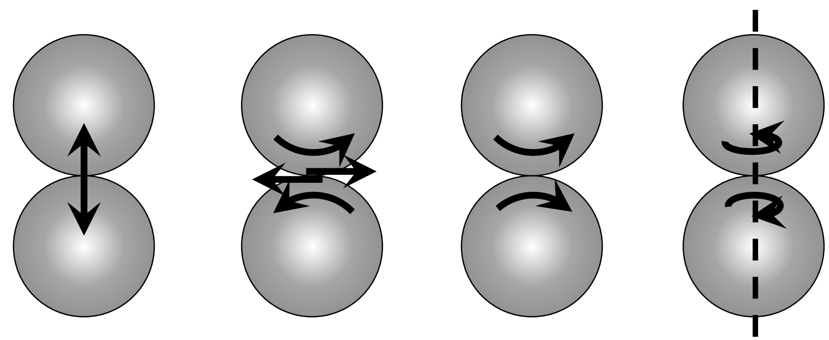
Contact force/torque



Slip

Stick

Adhesion

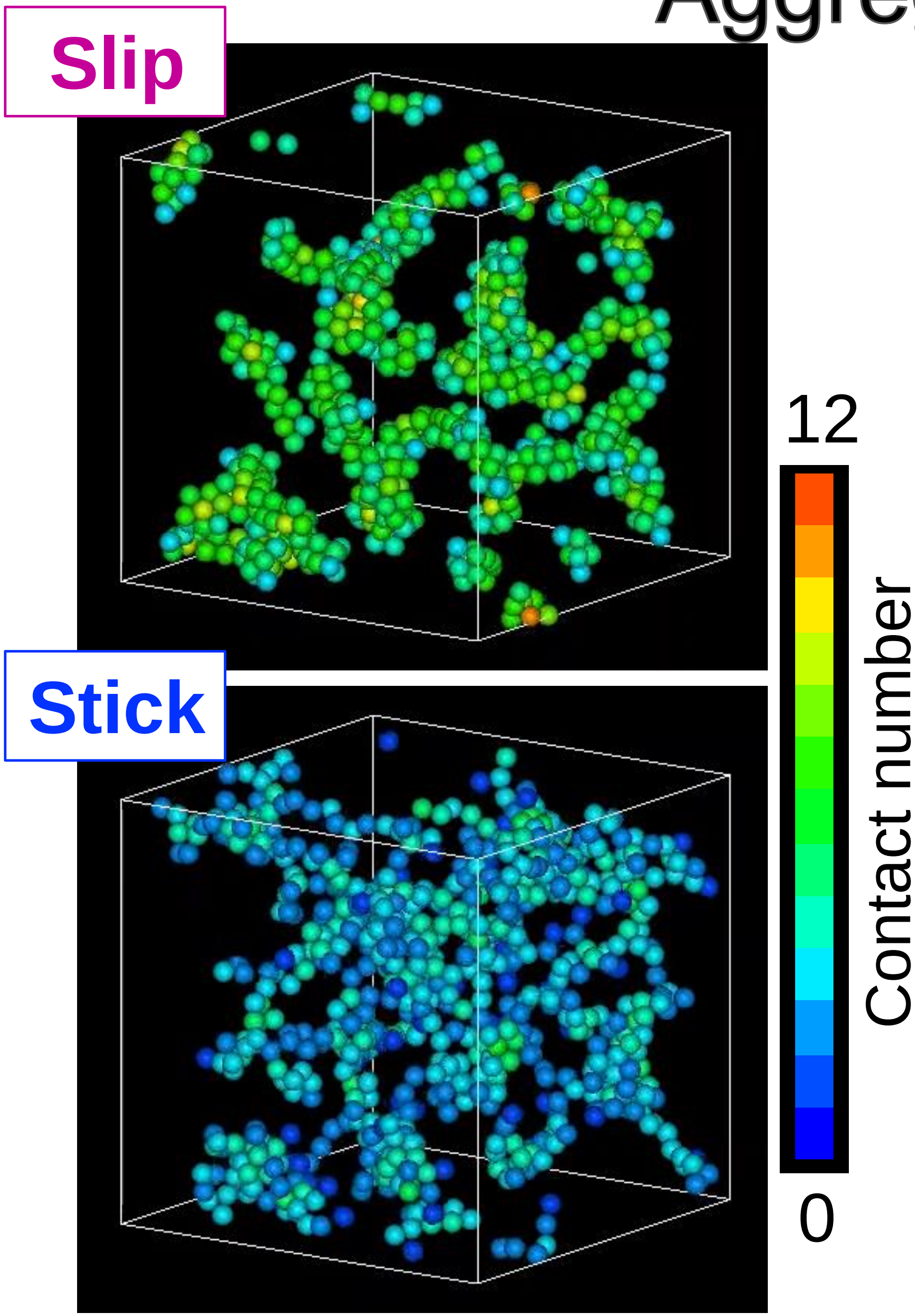


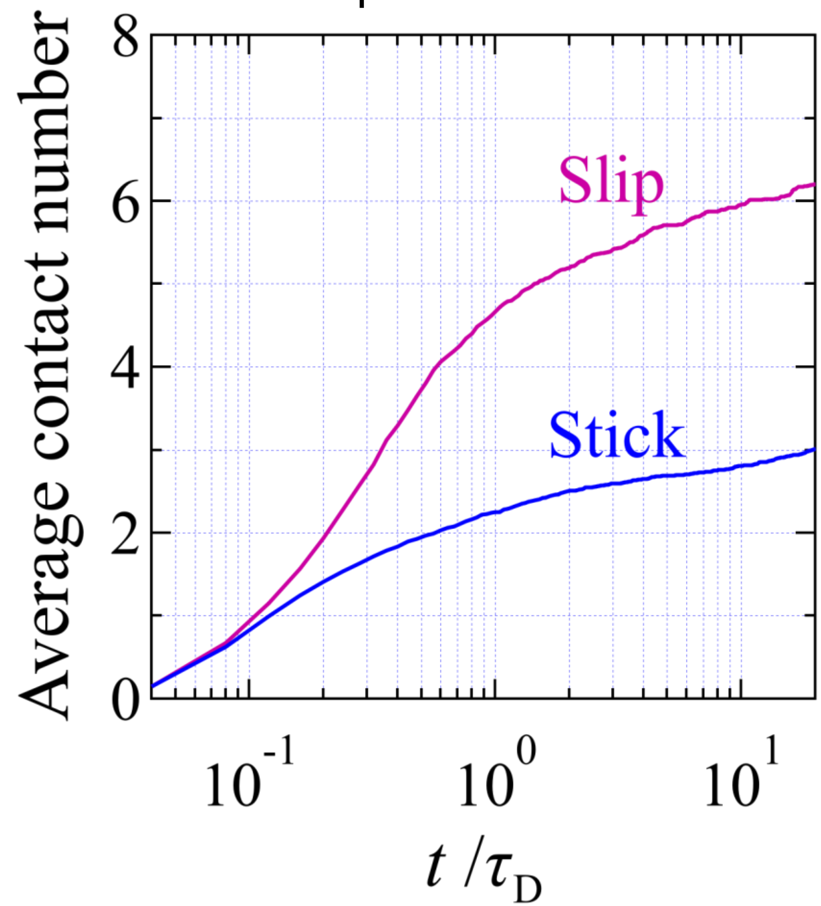
Constraint on the relative motions

How does adhesion affect the structure of particles?

Aggregation

Particle diameter: $d = 10$ nm
Zeta potential: 0 mV

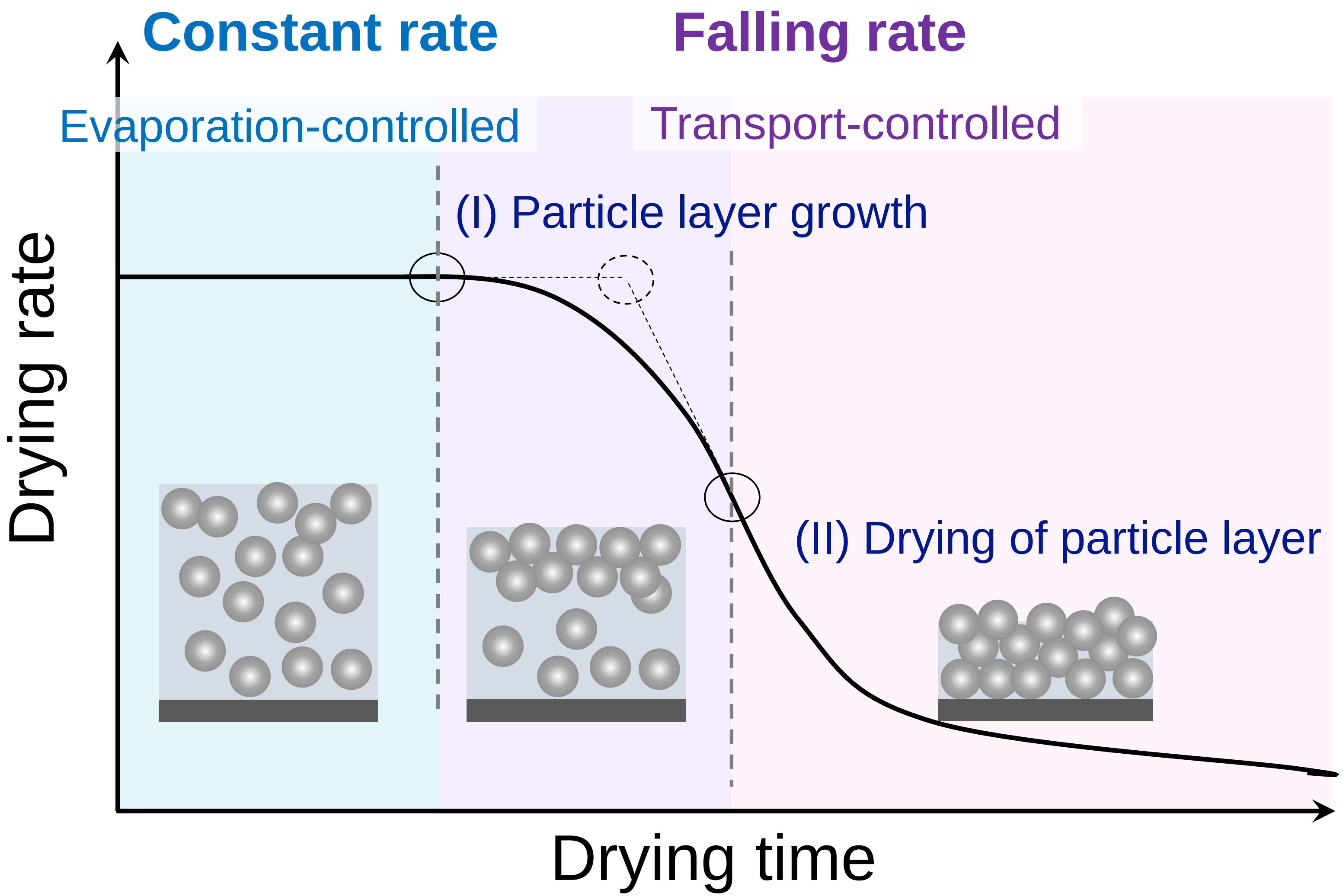




Characteristic time of particle diffusion: $\tau_D = d^2/D$

Particle diameter: d
Diffusion coefficient: D

Drying Curve of Colloidal Suspensions



Drying rate vs. Structure

Modeling of Falling Drying Rate

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

Resistance of evaporation: R_0

Resistance of particle layer: R_p

↑

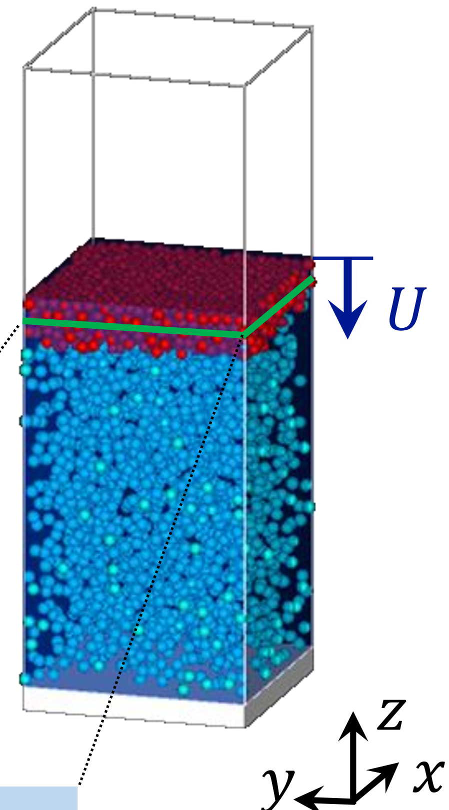
Aggregated particles moving with free surface

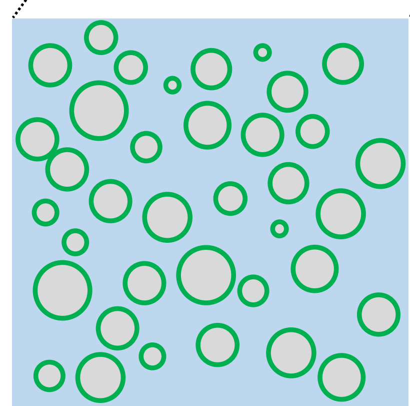
$$R_\mathrm{p} = \int_P r(z)\mathrm{d}z \qquad r(z) = \frac{80}{[D_\mathrm{H}(z)]^2}\frac{S_\mathrm{tot}}{S_\mathrm{f}(z)}$$

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

Cross-sectional area of the flow: S_f

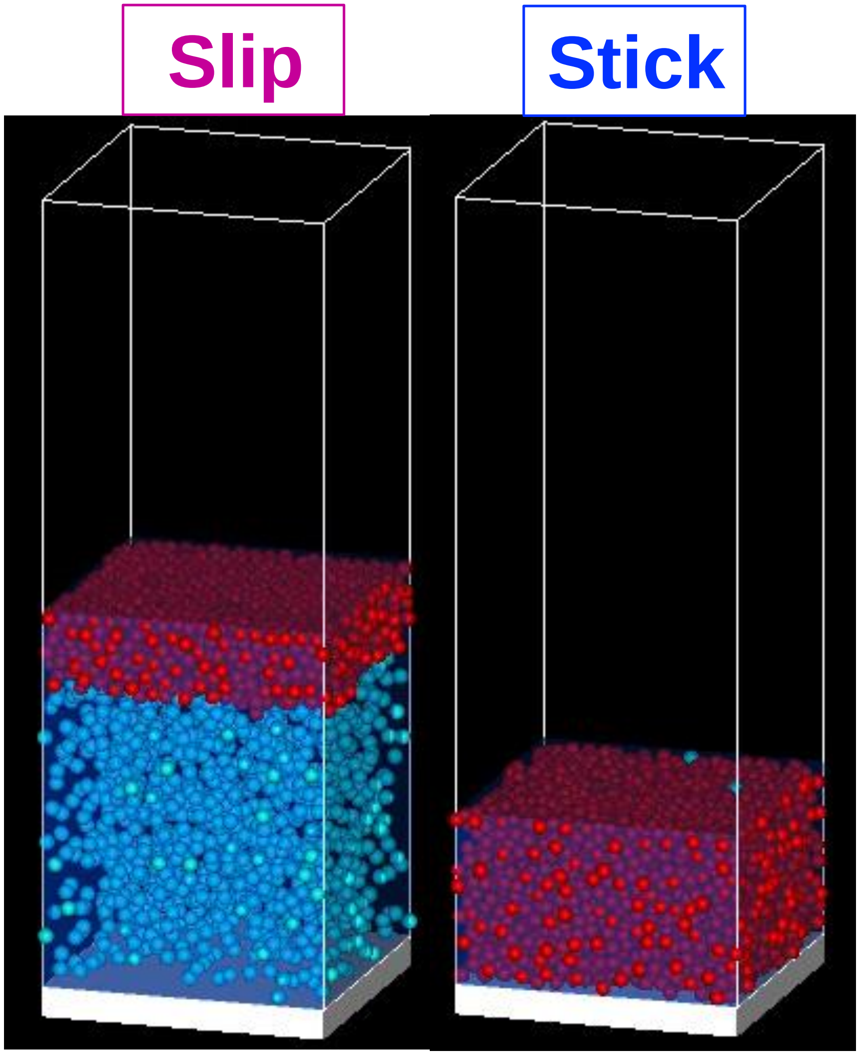
Wetted perimeter: L_f

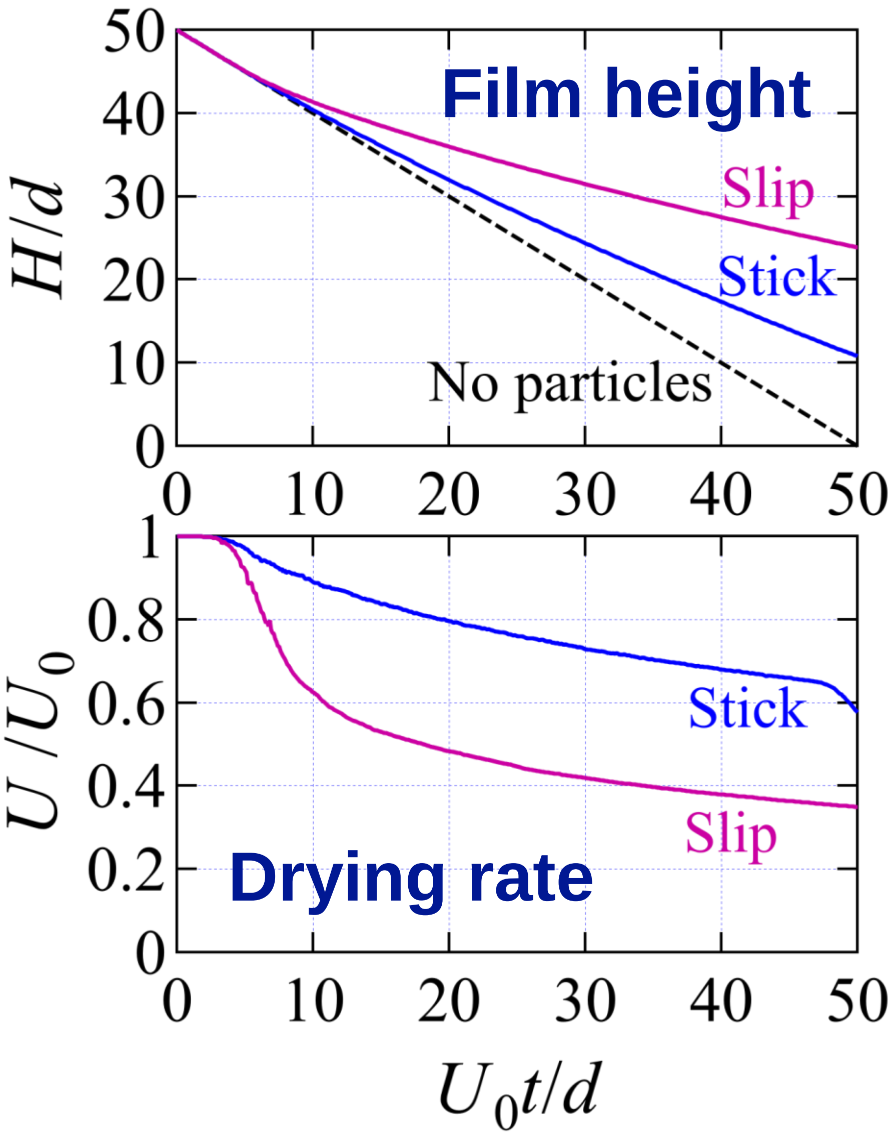




Cross-section

Drying Curves

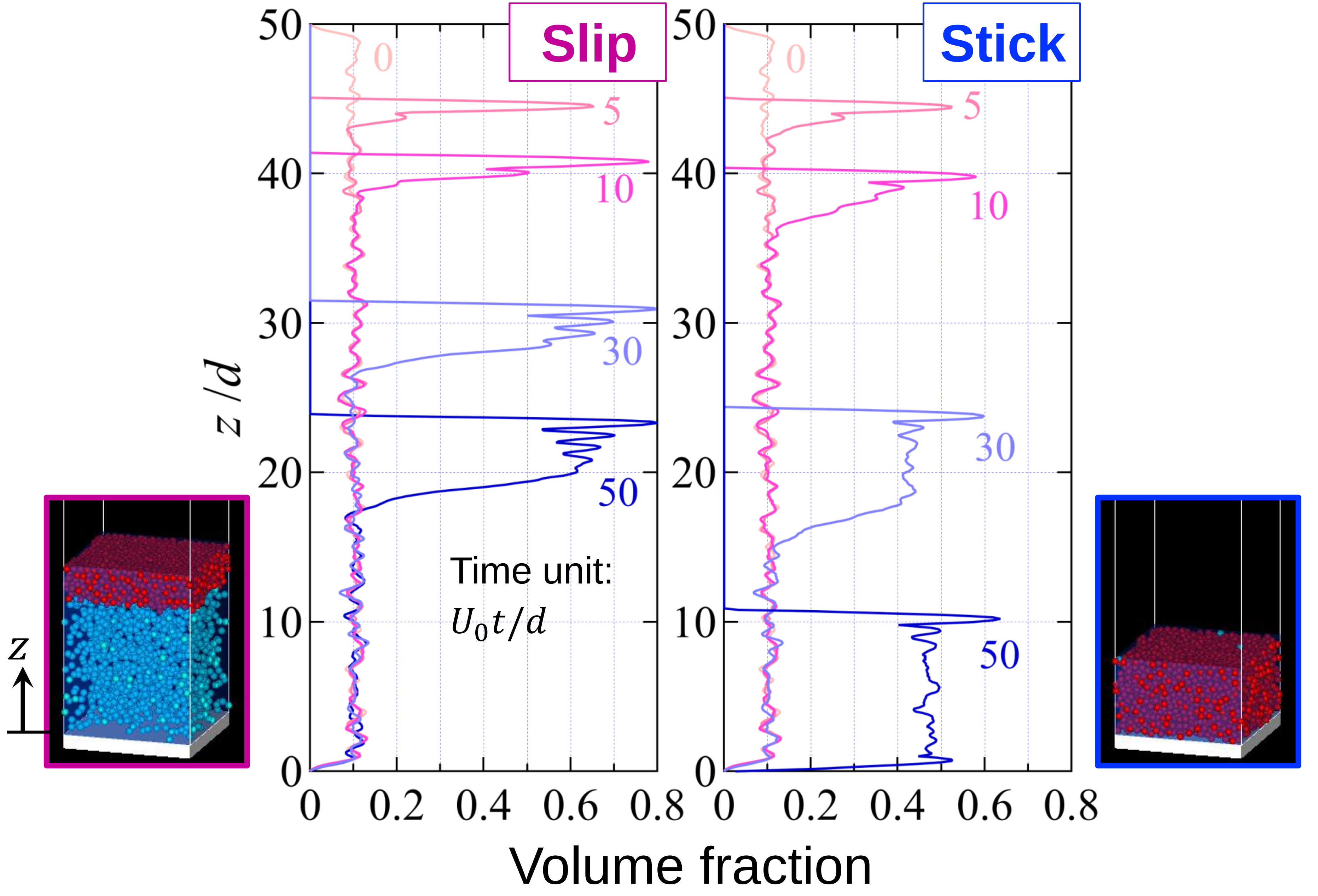




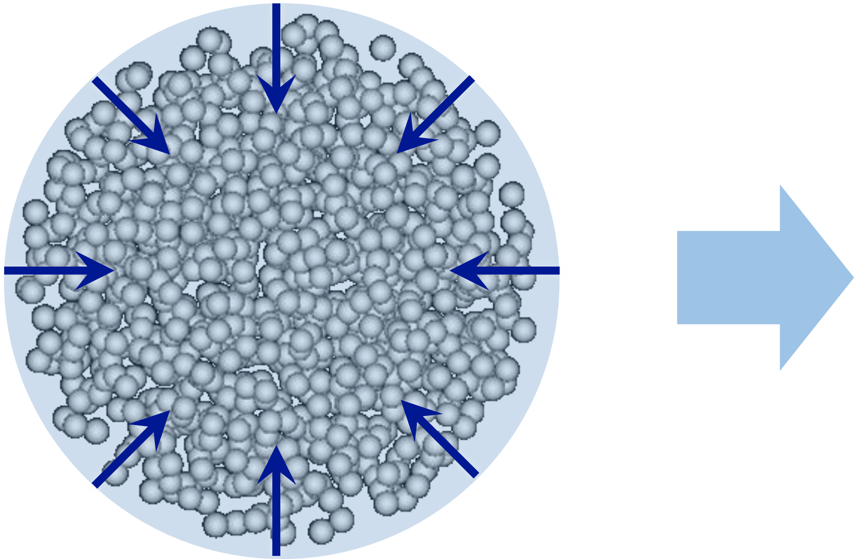
Particle diameter: $d = 20$ nm
Zeta potential: -50 mV

Drying time

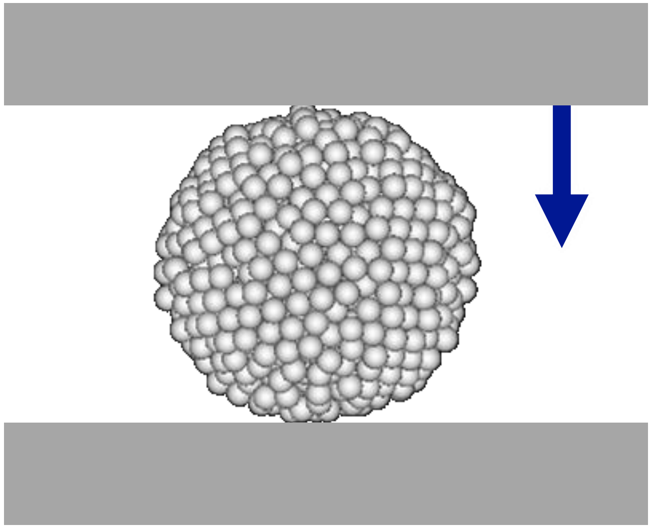
Particle Distribution



Strength of Structure



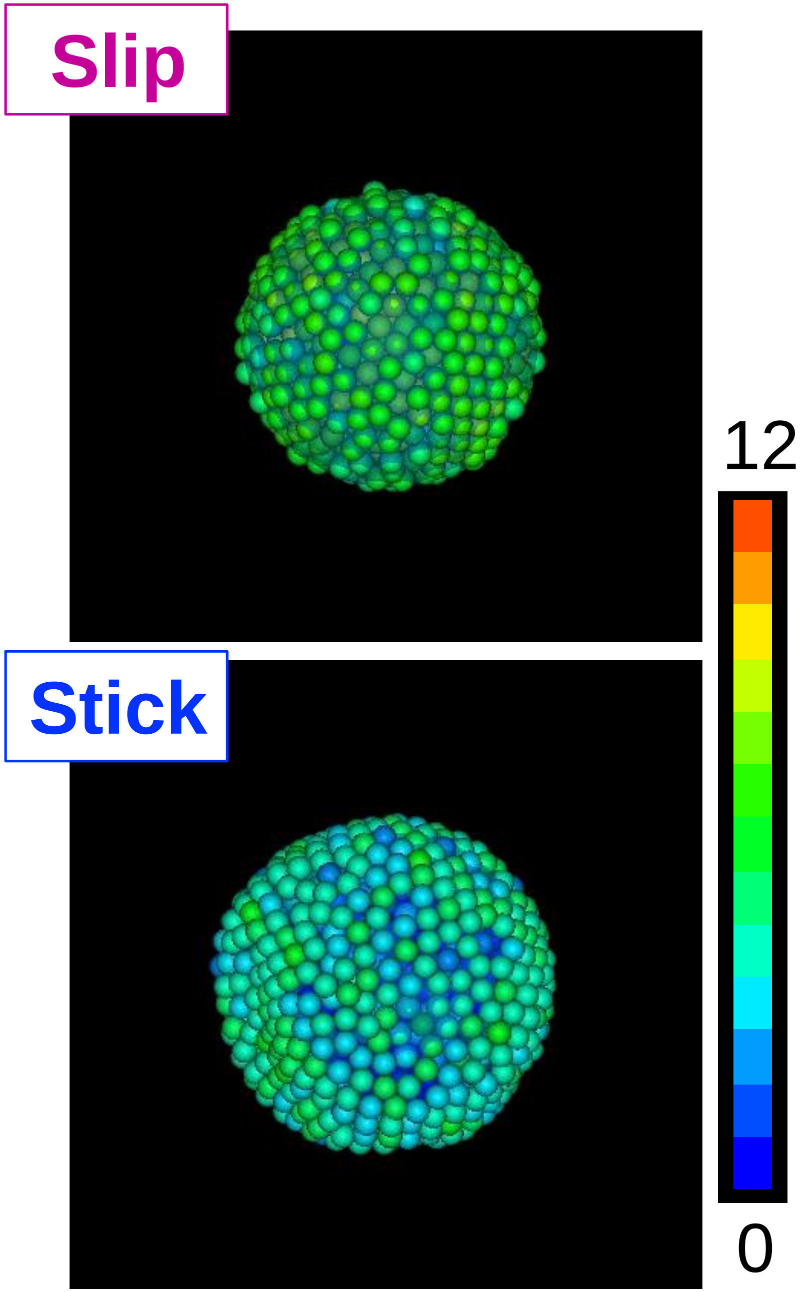
Drying of colloidal droplets



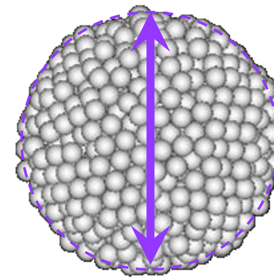
Compression

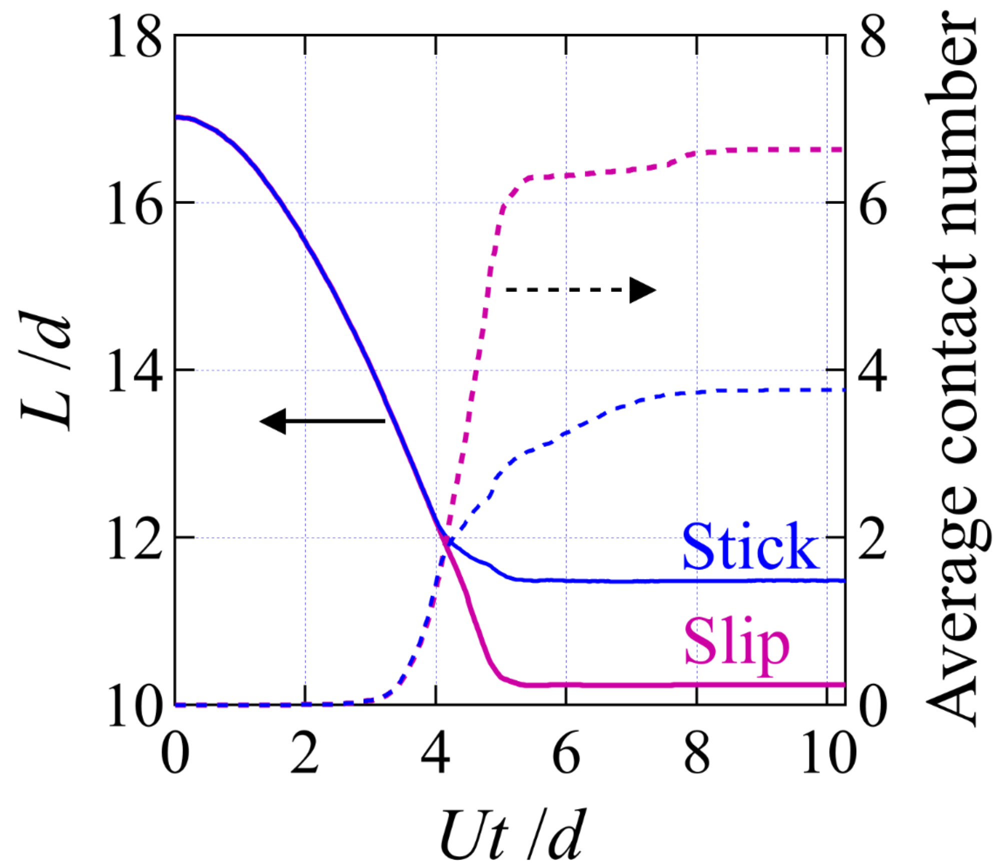
Porosity vs. Strength

Drying



Apparent diameter: L

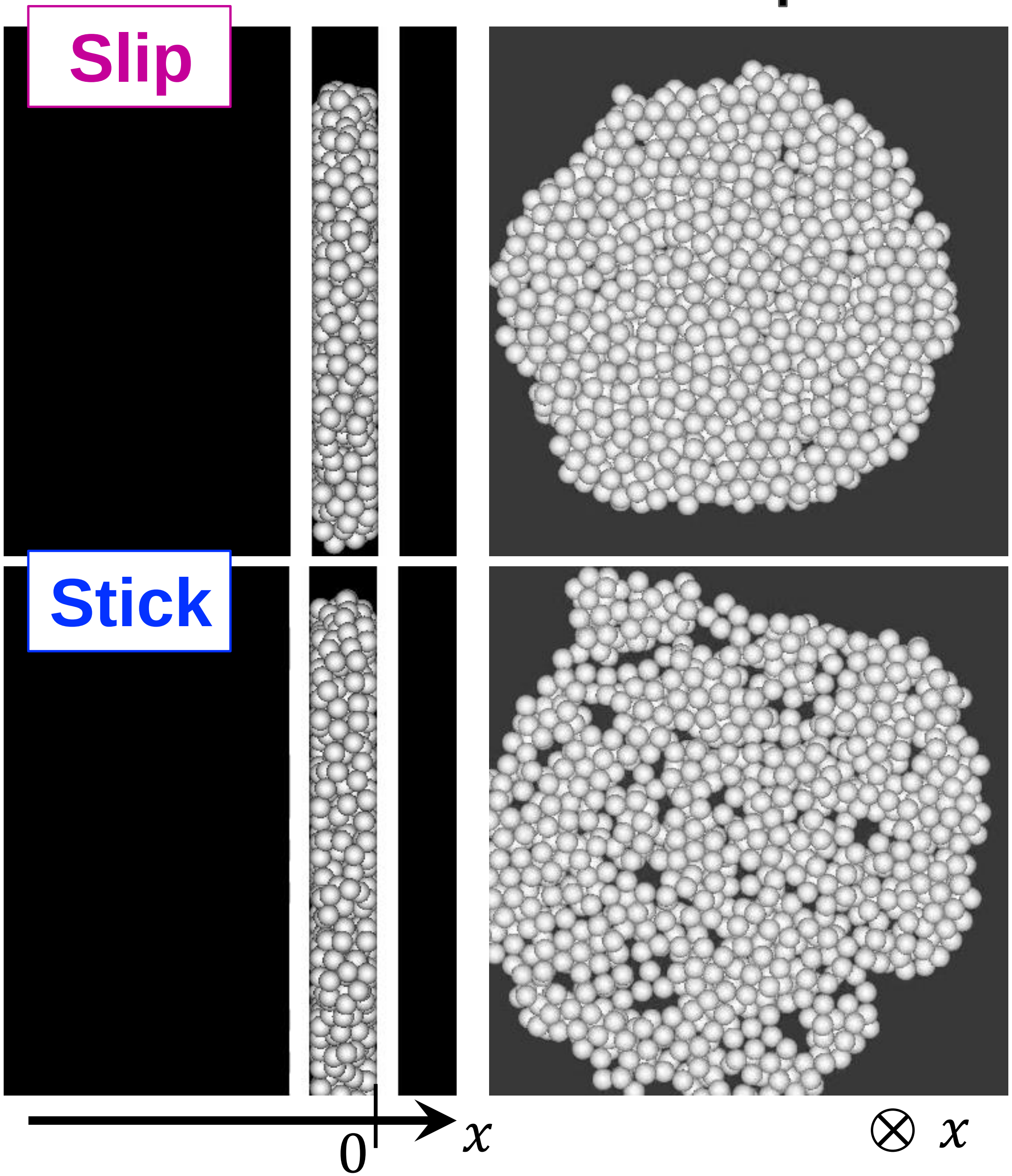




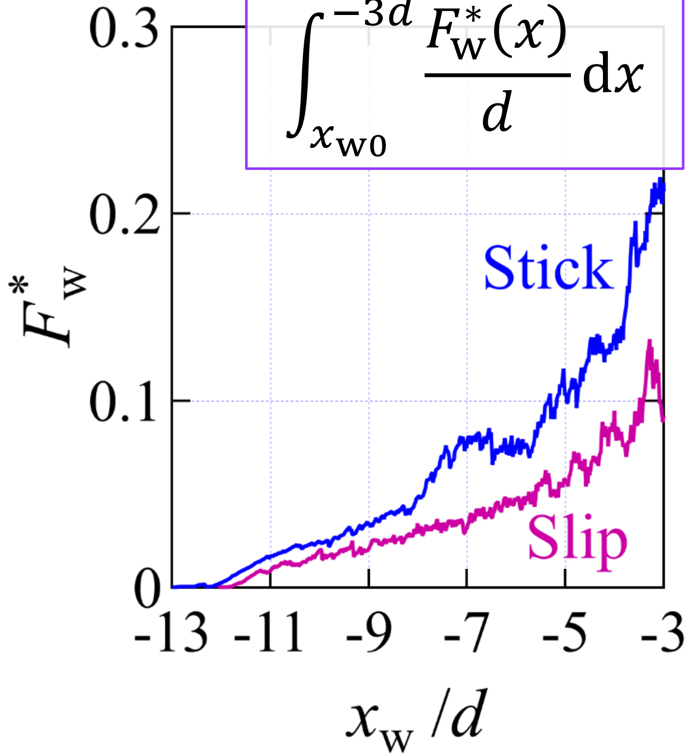
Constant drying rate: U

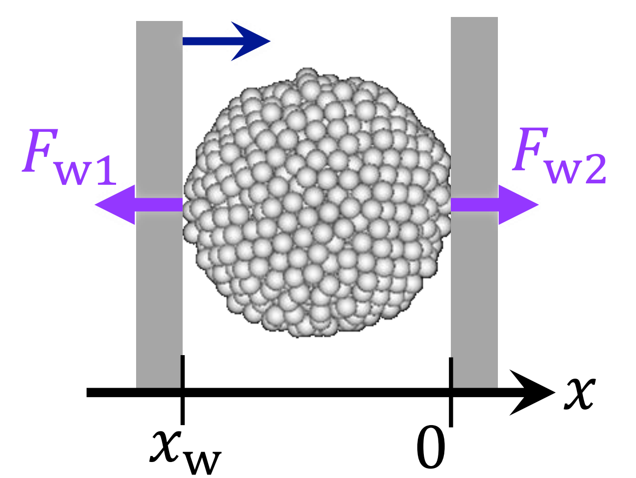
Particle diameter: $d = 20\ \mathrm{nm}$, Zeta potential: $-50\ \mathrm{mV}$

Compression

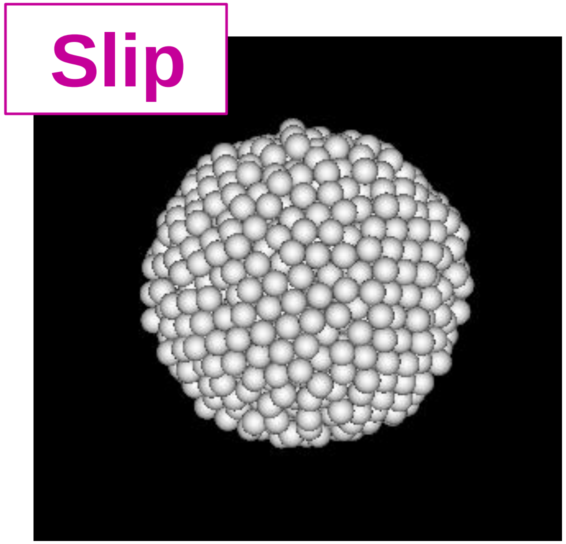


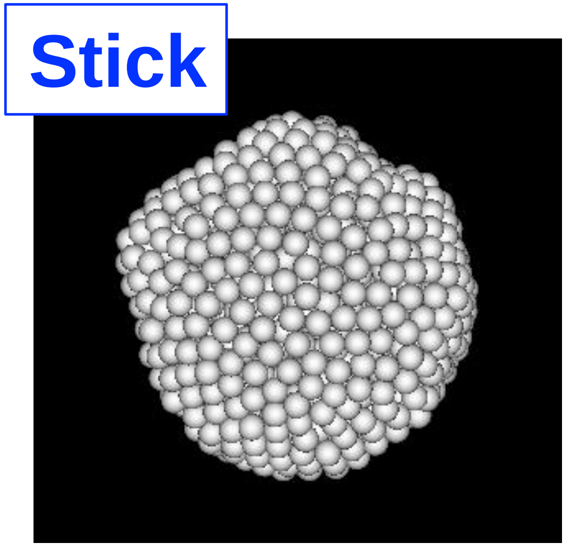
Strength: $\int_{x_{\mathrm{w0}}}^{-3d} \frac{F_{\mathrm{w}}^{*}(x)}{d}\,\mathrm{d}x$





Structure

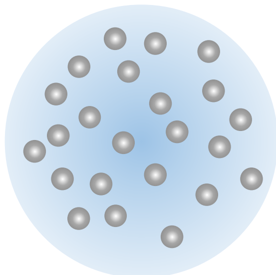




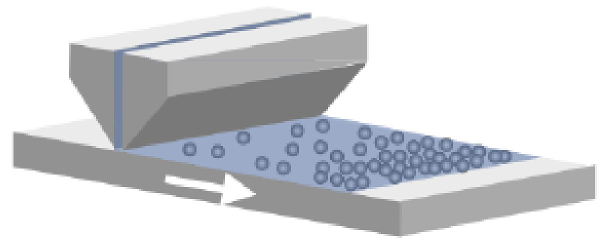
	Diameter	Porosity	Strength
Slip	10.2	0.07	0.35
Stick	11.5	0.34	0.64

Strong granule with high porosity

Network Formation of Particles



Aqueous suspensions
of nanoparticles



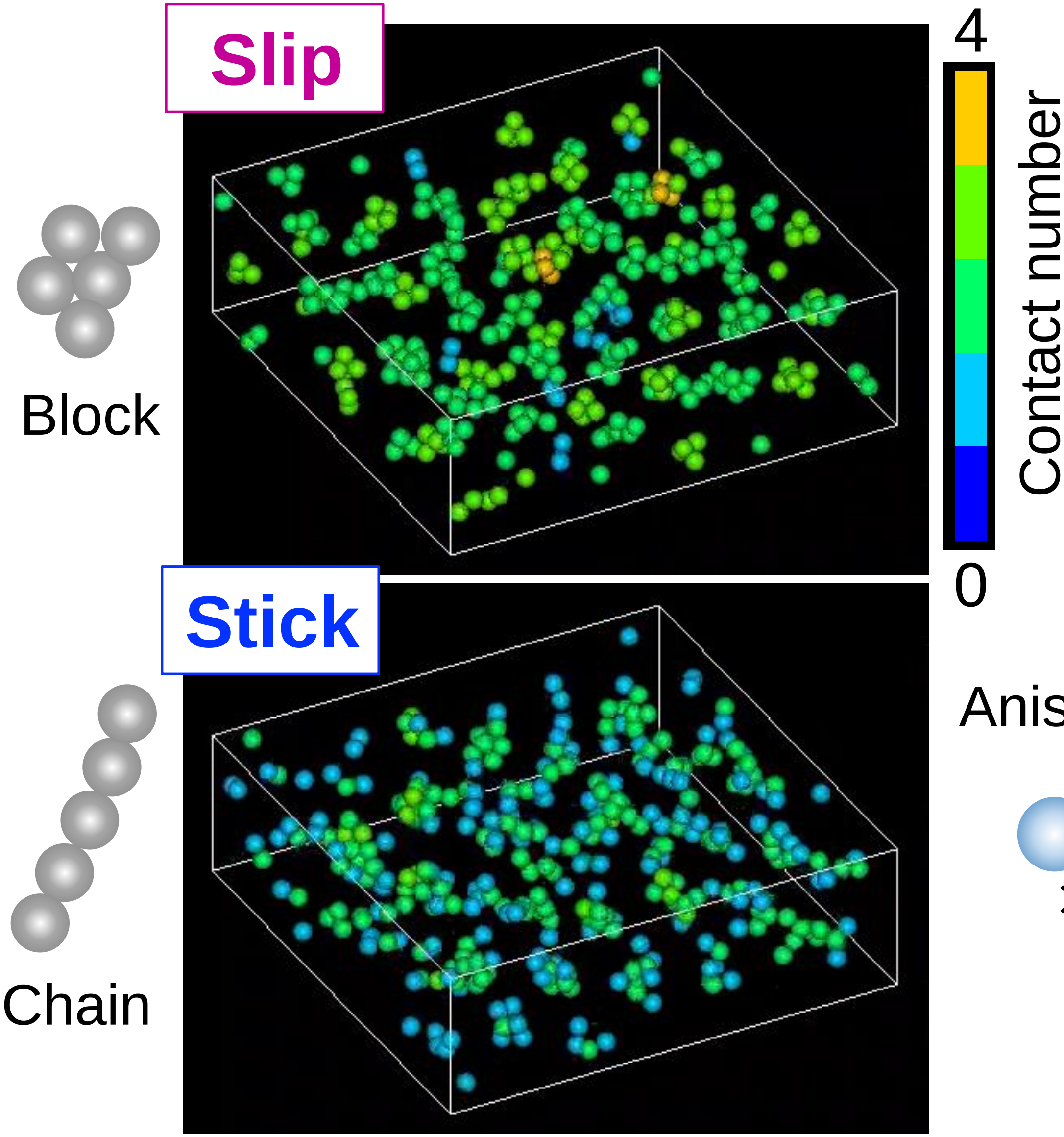
Coating
Drying

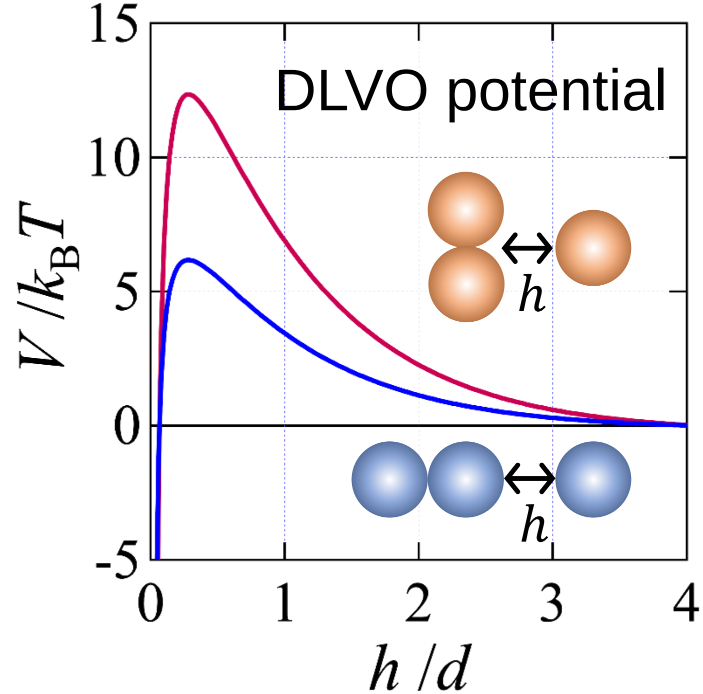
Transparent conductive films

Example of network structure:
Wakabayashi *et al*., Langmuir (2007).

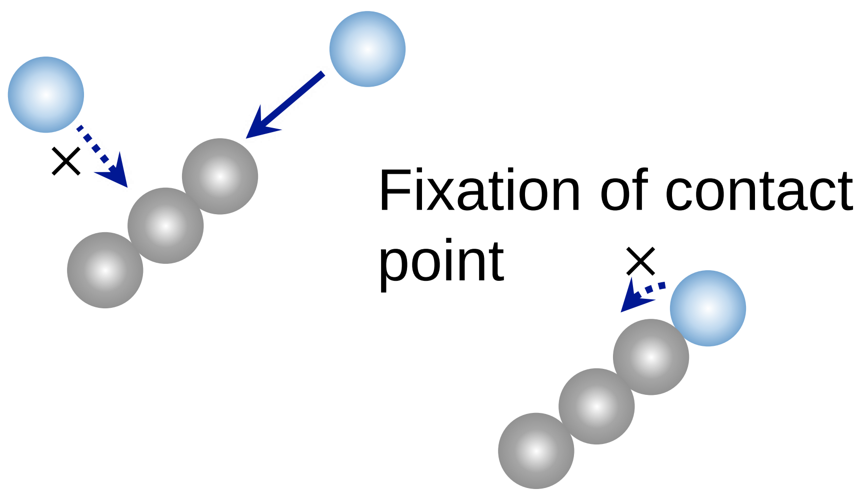
How do network structures form?

Aggregation



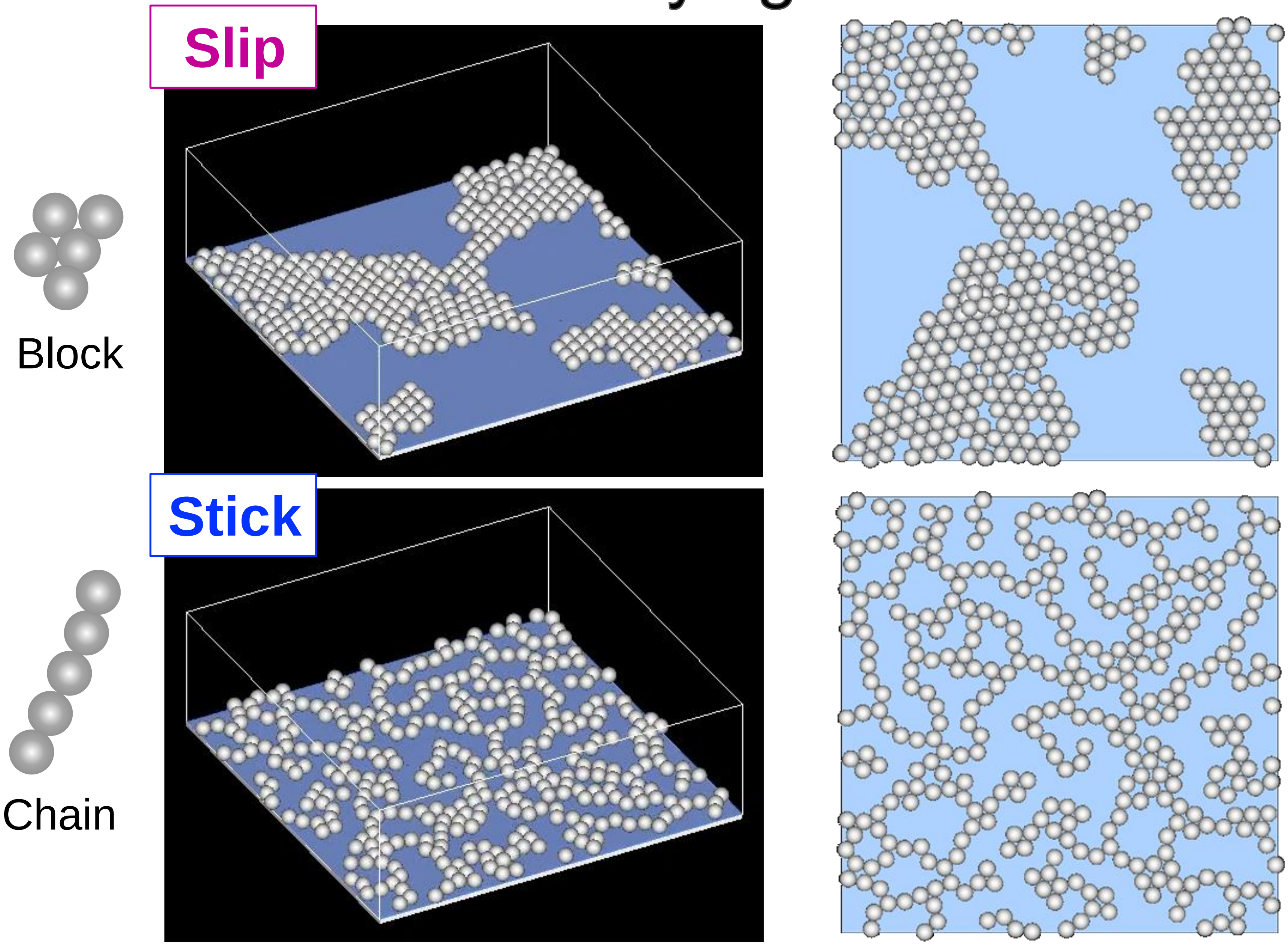


Anisotropic aggregation



Particle diameter: $d = 10$ nm

Drying



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

- ◆ Adhesion → Structures with high porosity formed during drying
  - High permeability (drying rate)
  - High strength
  - Network structures → Transparent conductive films