SCEJ 50th Autumn Meeting
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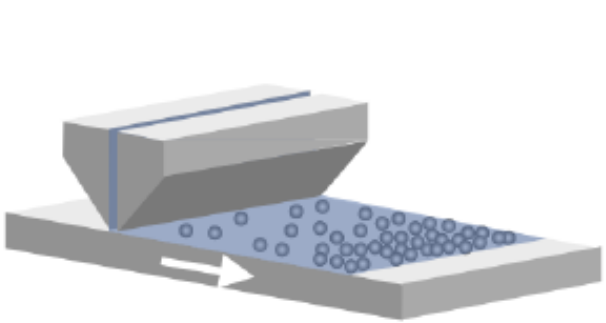
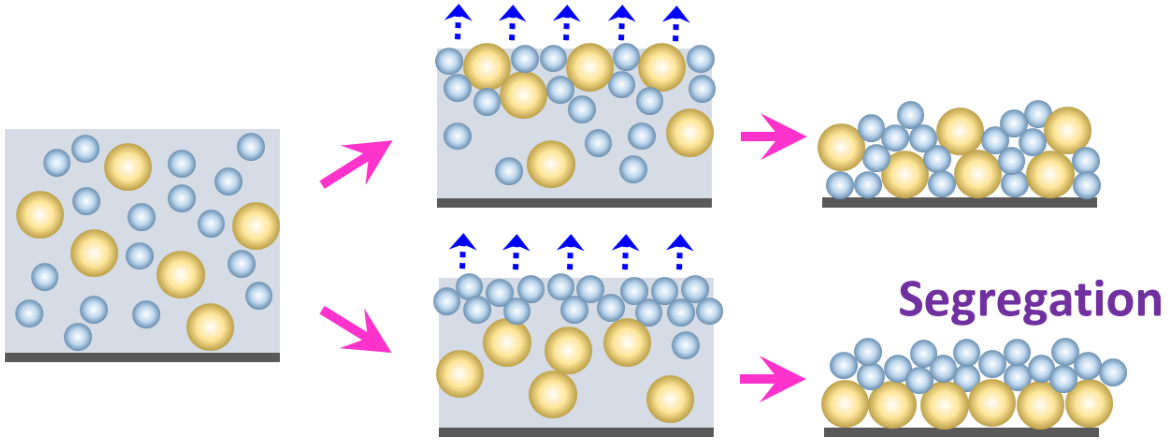
Classification of drying segregation states by generalized diffusion model analysis

一般化拡散モデルによる乾燥偏析状態の解析的分類

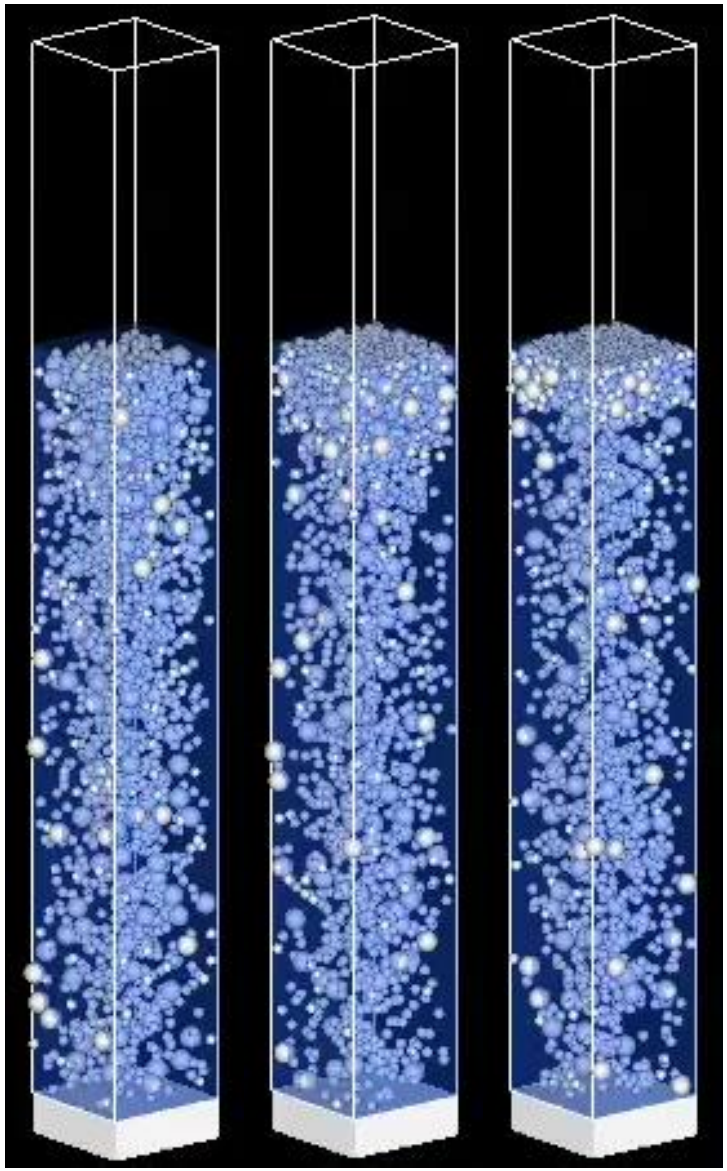
○ 辰巳 怜　（東大環安セ）
小池 修　（PIA）
山口由岐夫　（PIA）
辻 佳子　（東大環安セ/東大院工）



Coating, **Drying**

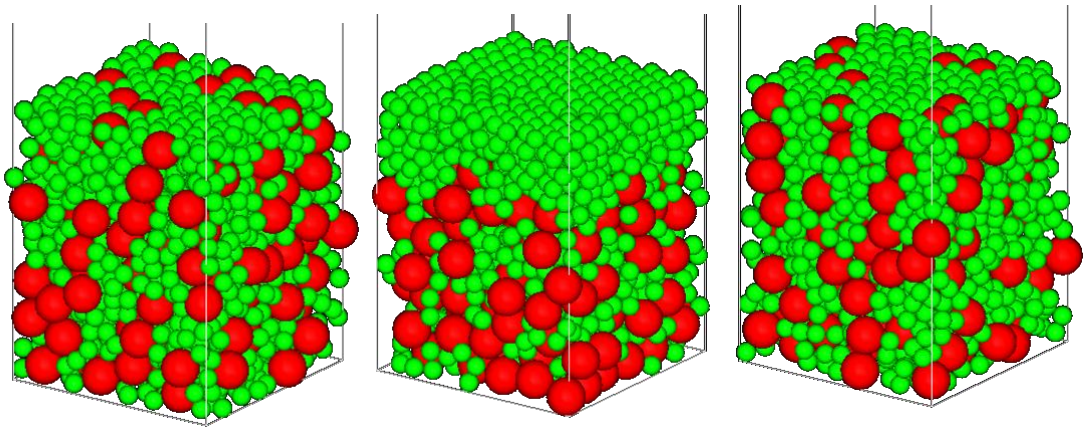


Particle Dynamics



Pe = 0.3 5 100

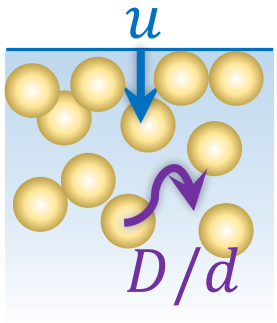
Diameter ratio: 2



Pe = 0.3 5 100

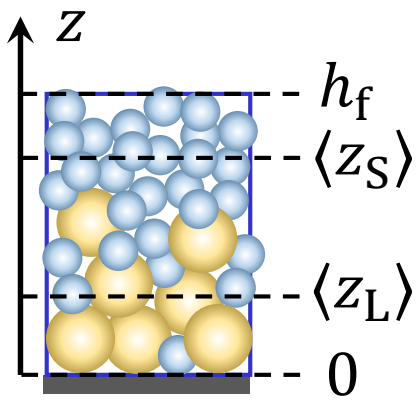
Particle drying Péclet number:

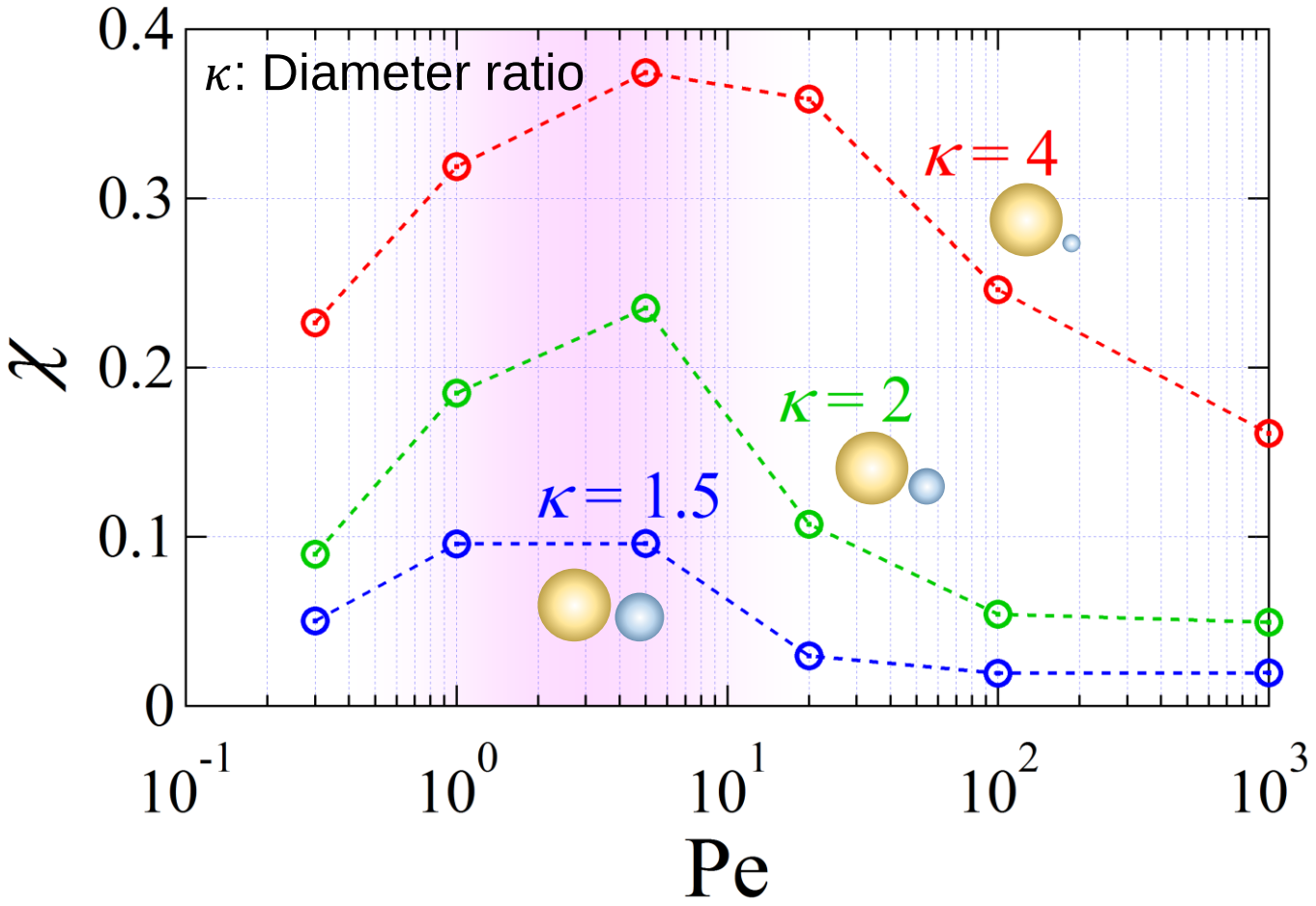
$$\mathrm{Pe} = \frac{(\text{Evaporation rate})}{(\text{Diffusion rate})} = \frac{ud}{D}$$



Tatsumi *et al*., Appl. Phys. Lett. (2018).

Segregation Condition





Indicator of segregation: $\chi \equiv (\langle z_S \rangle - \langle z_L \rangle)/h_f$

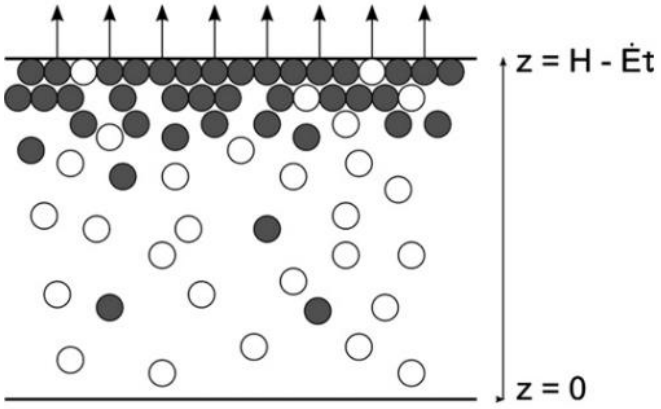
Average z-coordinate of the particles : $\langle z_L \rangle$: $\langle z_S \rangle$

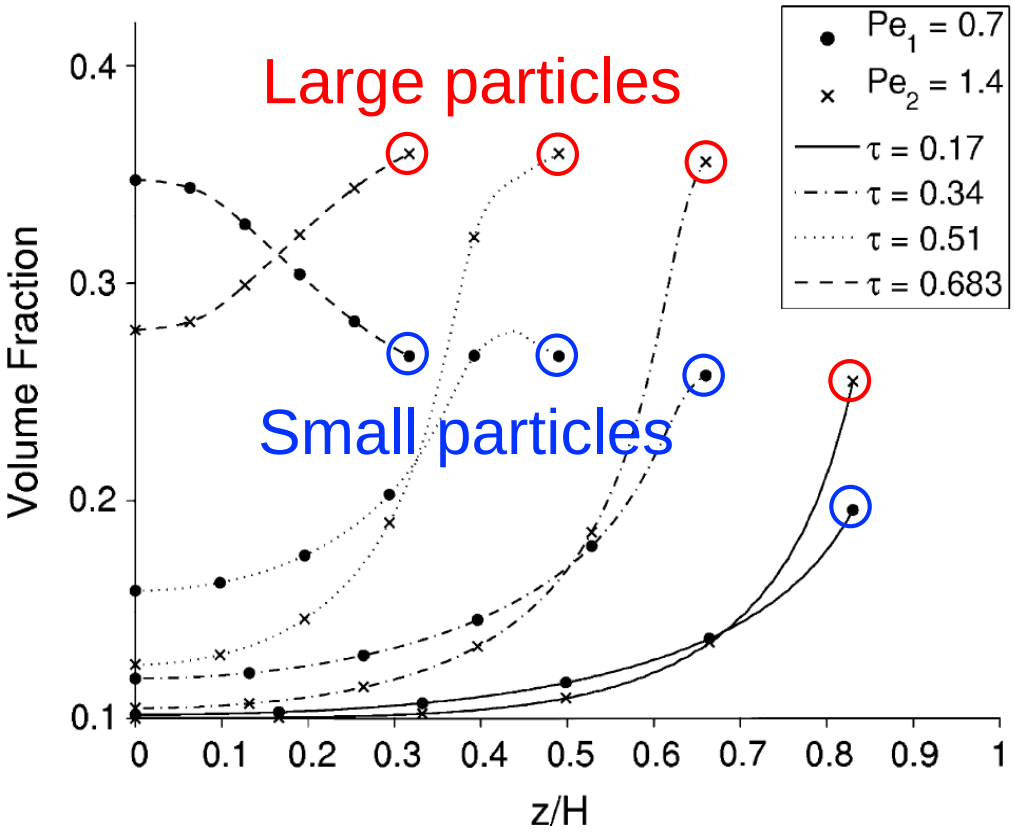
1D Diffusion Model

Time evolution of particle concentration dist.

$$\frac{\partial \phi_1}{\partial t} + \nabla \cdot \phi_1 \mathbf{U}_1 = 0$$

$$\mathbf{U}_1 = -\frac{K(\phi_1, \phi_2)}{6\pi\eta R_1} \nabla \mu_1$$





Segregation of Large particles to the top surface

← Diffusion rate: (Large particles) < (Small particles)

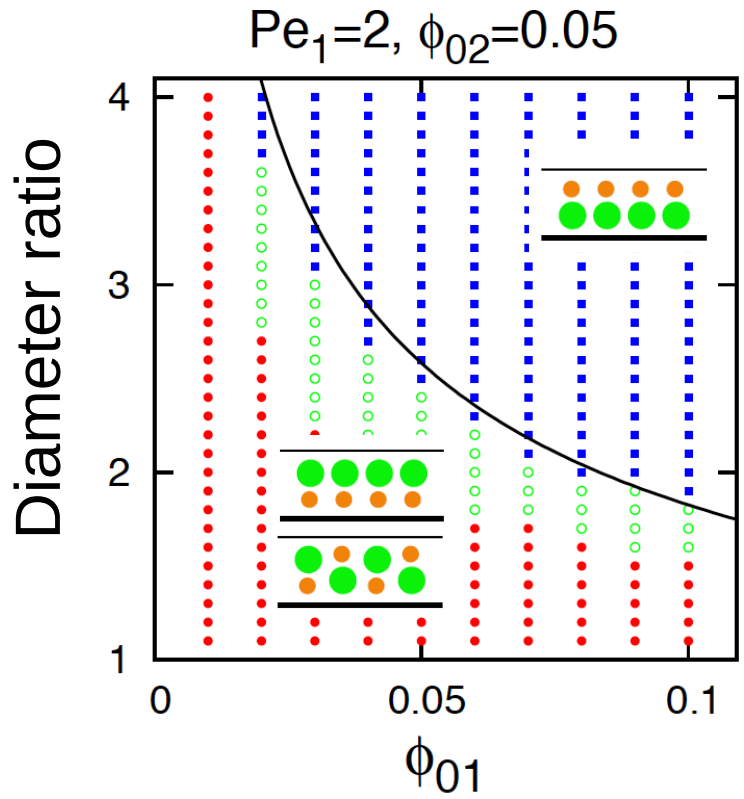
Trueman *et al*., J. Colloid Interface Sci. (2012).

1D Diffusion Model

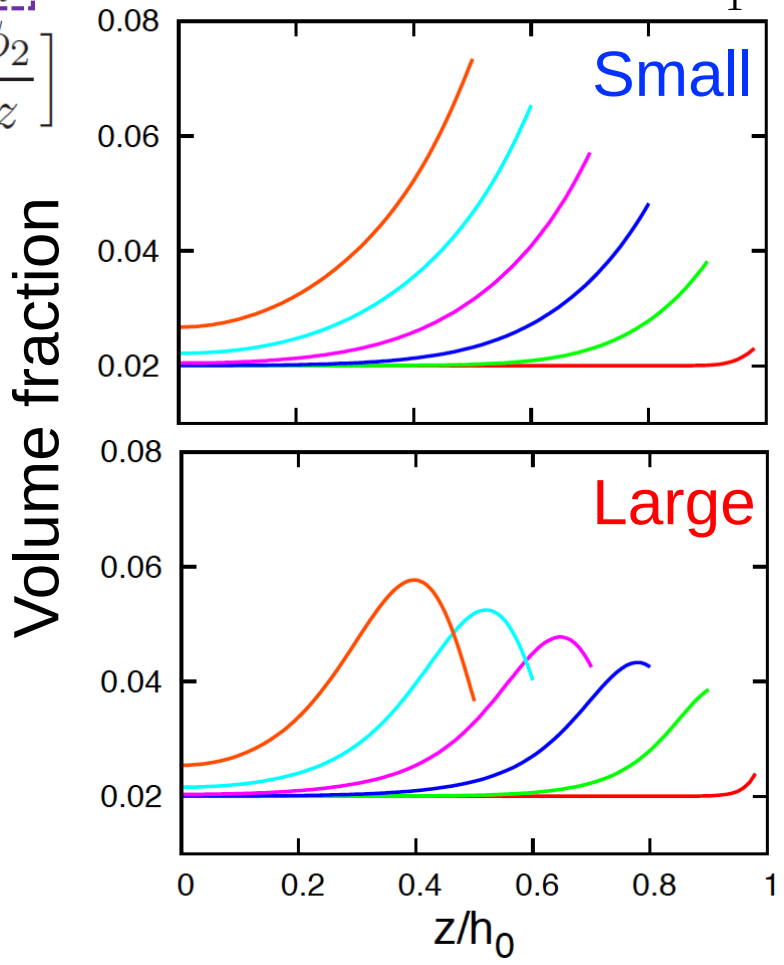
Cross-diffusion → Segregation of Small particles to the top surface

$$\frac{\partial \phi_1}{\partial t} = D_1 \frac{\partial}{\partial z}\left[(1+8\phi_1)\frac{\partial \phi_1}{\partial z} + (1+\frac{1}{\alpha})^3 \phi_1 \frac{\partial \phi_2}{\partial z}\right]$$

$$\frac{\partial \phi_2}{\partial t} = D_2 \frac{\partial}{\partial z}\left[(1+\alpha)^3 \phi_2 \frac{\partial \phi_1}{\partial z} + (1+8\phi_2)\frac{\partial \phi_2}{\partial z}\right]$$



Diameter ratio 4 $\mathrm{Pe}_1 = \frac{v_e h_0}{D_1} = 5$



Zhou *et al.*, Phys. Rev. Lett. (2017).

Objective

- Classification of drying segregation states by generalized diffusion model analysis

- Examination of factors affect segregation:
  - Cross-diffusion coefficient
  - Particle size ratio (Ratio of diffusion coefficient)
  - Mixing ratio (Mole fraction)
  - Drying time

Model

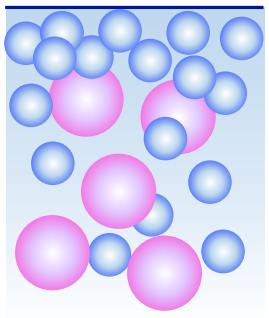
Binary mixture dispersed in a liquid

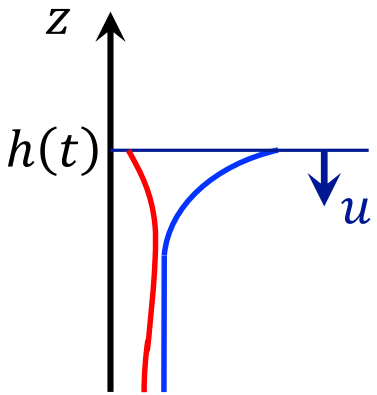
$$\frac{\partial \boldsymbol{c}}{\partial t} = -\frac{\partial \boldsymbol{J}}{\partial z} = \boldsymbol{D} \cdot \frac{\partial^2 \boldsymbol{c}}{\partial z^2}$$

Generalized Fick's law: $\boldsymbol{J} = -\boldsymbol{D} \cdot \frac{\partial \boldsymbol{c}}{\partial z}$

$$\boldsymbol{c} = \begin{pmatrix} c_1 \\ c_2 \end{pmatrix} \quad \boldsymbol{D} = \begin{pmatrix} D_{11} & D_{12} \\ D_{21} & D_{22} \end{pmatrix}$$

$$\rightarrow \quad \begin{aligned} \partial_t c_1 &= D_{11}\partial_z^2 c_1 + D_{12}\partial_z^2 c_2 \\ \partial_t c_2 &= D_{21}\partial_z^2 c_1 + D_{22}\partial_z^2 c_2 \end{aligned}$$





I.C. $\boldsymbol{c}(z, 0) = \boldsymbol{c}_0$

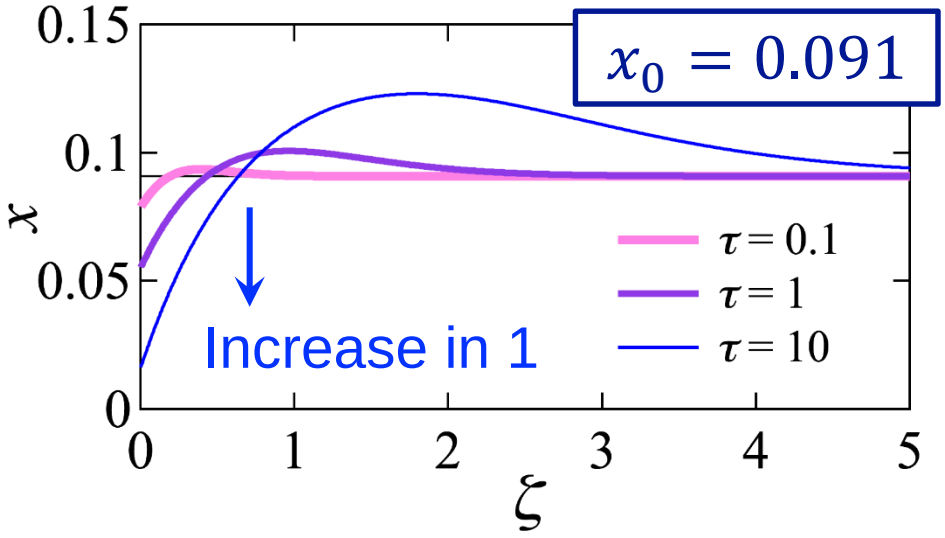
B.C. $\boldsymbol{D} \cdot \partial_z \boldsymbol{c}(h, t) = u\boldsymbol{c}(h, t)$ Free surface

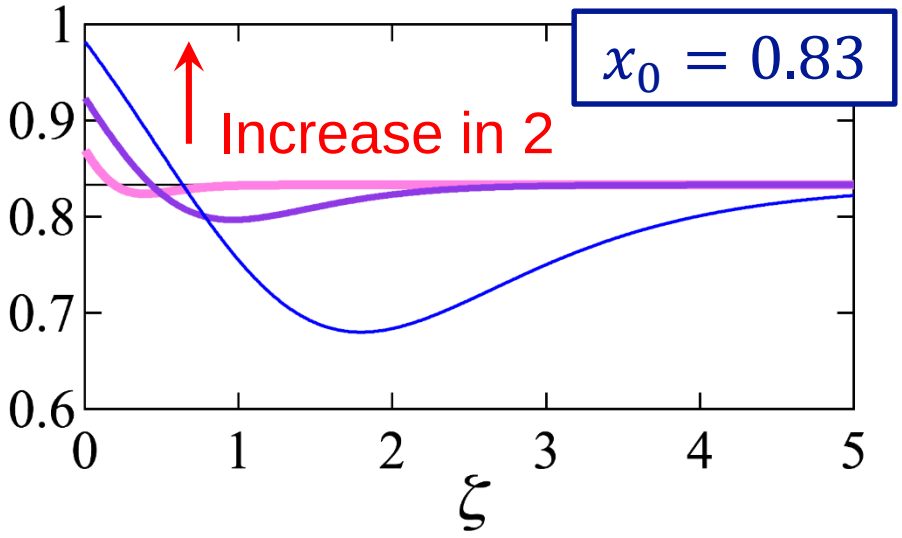
B.C. $\partial_z \boldsymbol{c}(0, t) = \boldsymbol{0}$ Substrate

- D_{ij}: Diffusion of i by the concentration gradient of j
- $D_{11} > D_{22}$; 1: Small particles 2: Large particles
- Approximation: $h \gg D_{11}/u$ → Analytical solutions

Distribution

$\Lambda = 0.5, \quad \Lambda_1 = \Lambda_2 = 0.1$





Mole fraction of 2: $x = c_2/(c_1 + c_2)$

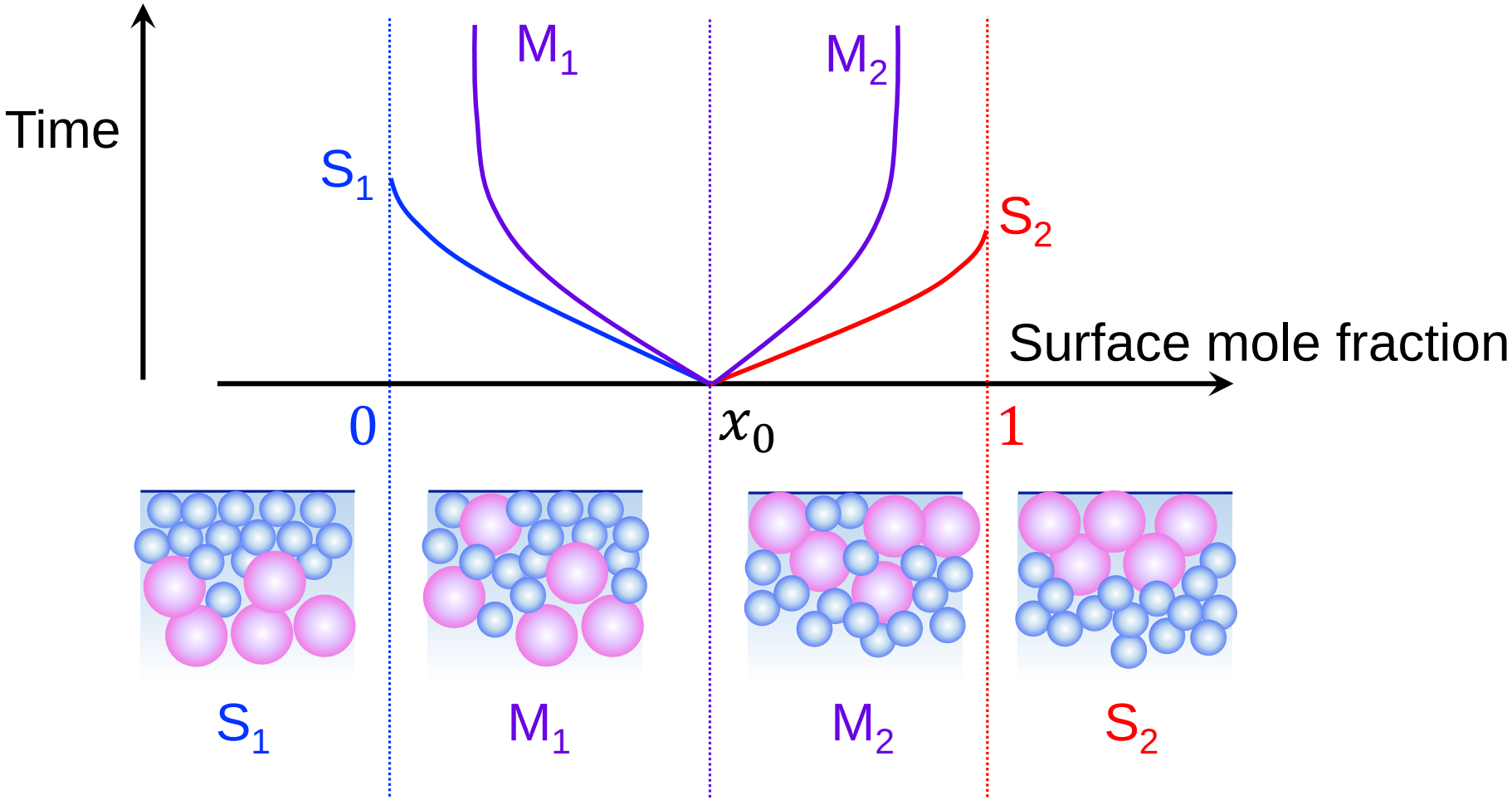
Dimensionless distance from free surface: $\zeta = (u/D_{11})(h - z)$

Dimensionless time: $\tau = (u^2/D_{11})t$

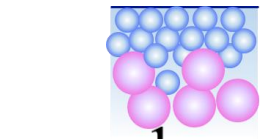
$$\boldsymbol{D} = \begin{pmatrix} D_{11} & D_{12} \\ D_{21} & D_{22} \end{pmatrix} = D_{11} \begin{pmatrix} 1 & \Lambda_2 \\ \Lambda_1 & \Lambda \end{pmatrix}$$

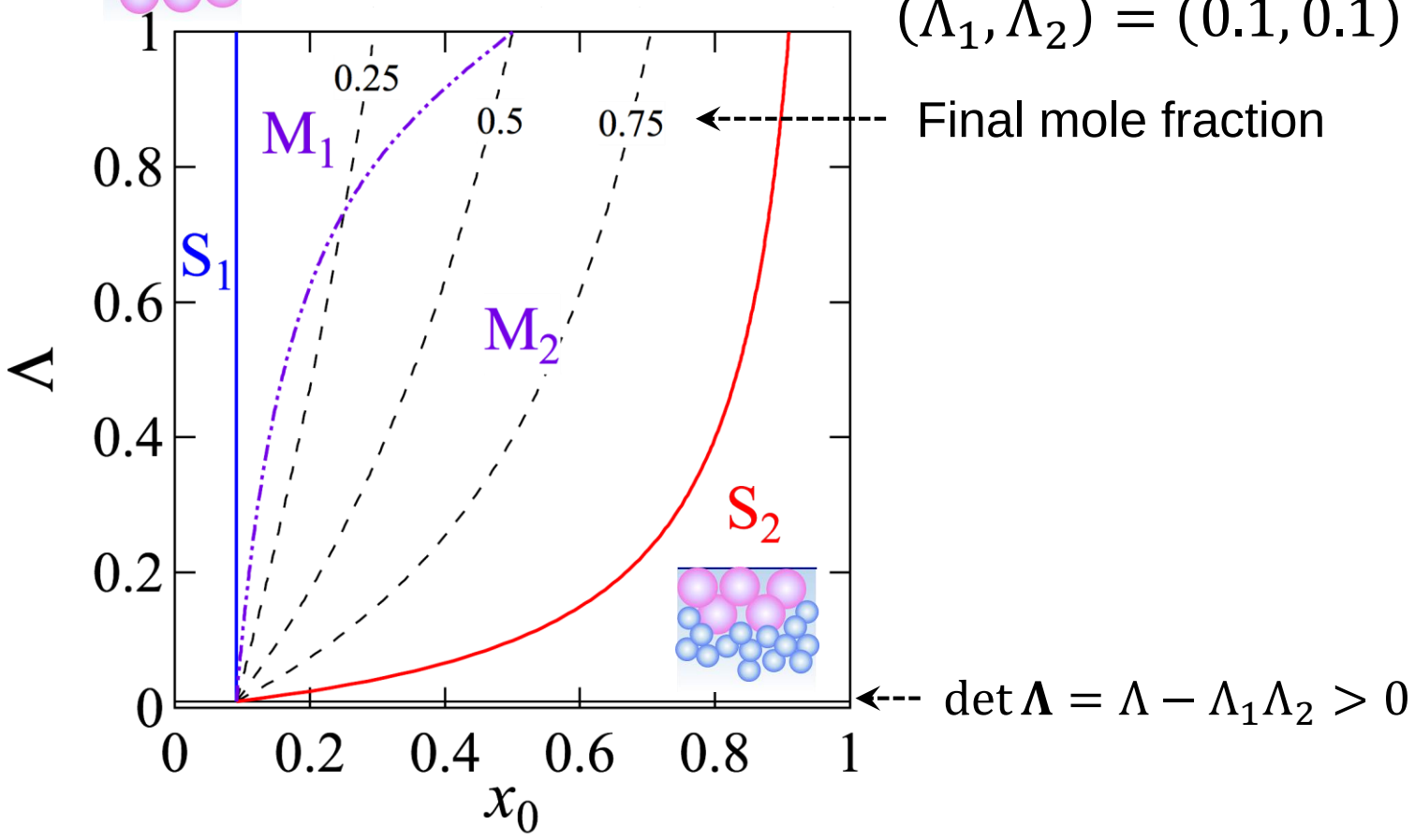
Classification of Segregation States

Classification by surface mole fraction of 2



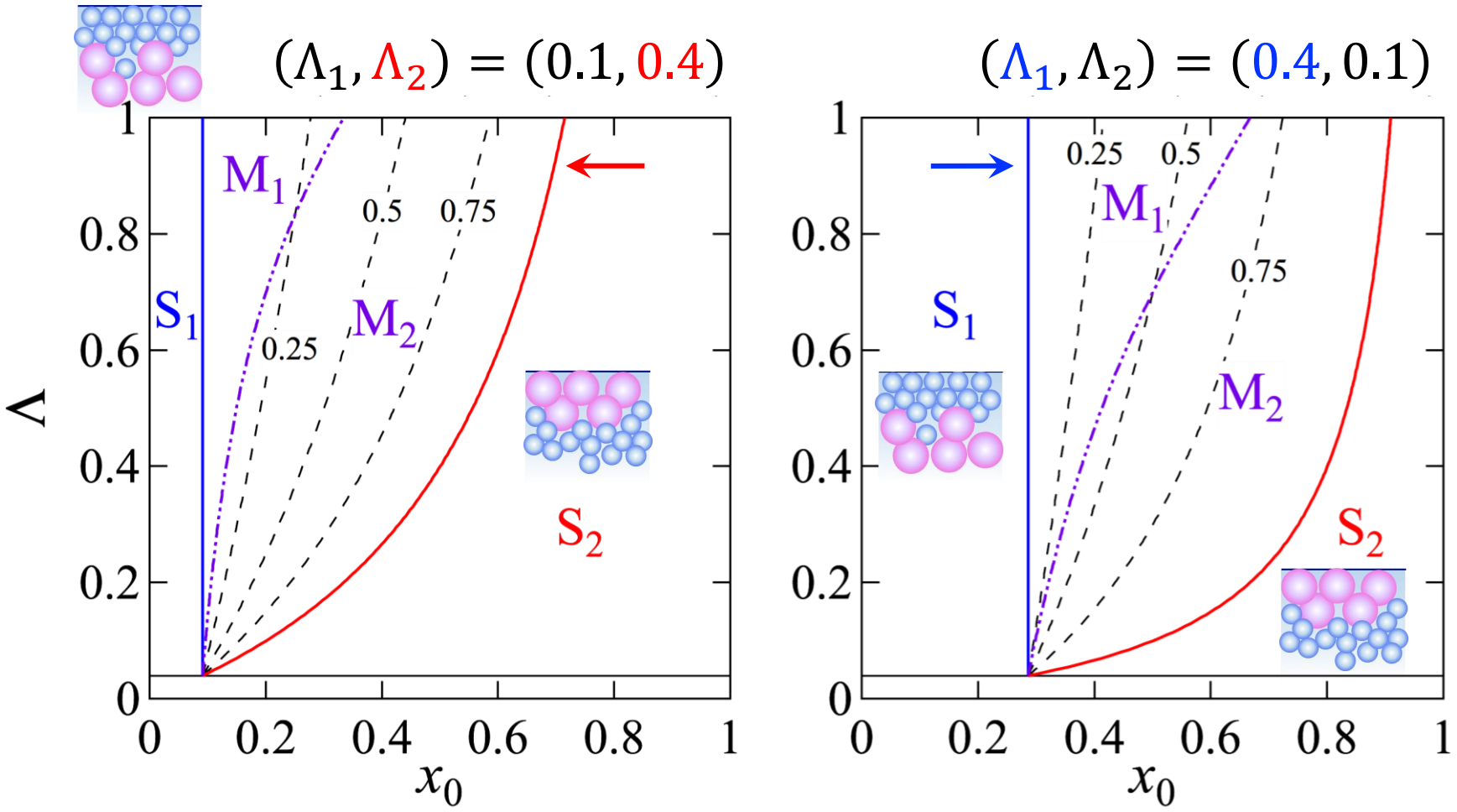
State Diagram





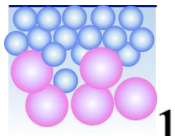
- Larger mole fraction of i → Segregation of i
- Smaller diffusion coefficient of 2 → Segregation of 2
  $1/\Lambda$ ↔ Particle size ratio

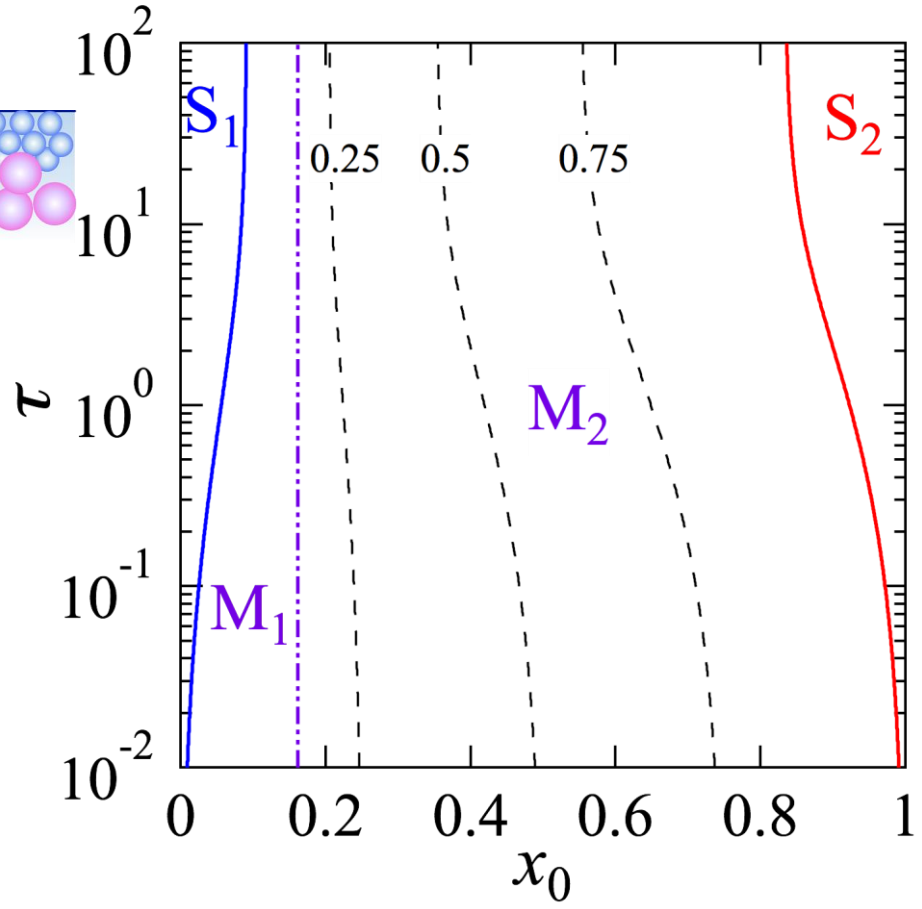
State Diagram



- Λ_i: i-th component excludes another

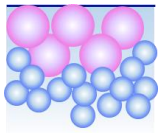
State Diagram





$\Lambda = 0.5$

$(\Lambda_1, \Lambda_2) = (0.1, 0.1)$



- Control of segregation by drying time

Summary

- Segregation conditions derived from the present model:

  - Cross-diffusion coefficient
    Larger $D_{ij} \rightarrow$ Expansion of S_j
  - Particle size ratio
    Smaller size ratio $\rightarrow$ Expansion of S_2
  - Mixing ratio
    Larger mole fraction of $i \rightarrow$ Segregation of i
  - Drying time
    Longer time $\rightarrow$ Segregation