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APPENDICES

Nomenclature

| | |
|-------------------------------|------------------------------------------------------------------------------------------------------------|
| T | absolute temperature |
| N_A | Avogadro's number ($6.02 \times 10^{23} \frac{\text{molecules}}{\text{mol}}$) |
| k_B | Boltzmann constant ($1.38 \times 10^{-23} \frac{\text{kg} \cdot \text{m}^2}{\text{s}^2 \cdot \text{K}}$) |
| β | collision efficiency |
| K_{ij} | collision kernel |
| D | diameter |
| K_B | diffusion parameter |
| η | dynamic viscosity of medium |
| e | elementary charge ($1.60 \times 10^{-19} \text{ C}$) |
| R_h | hydrodynamic radius |
| ϵ_0 | permittivity of free space ($8.85 \times 10^{-12} \frac{\text{C}^2}{\text{J} \cdot \text{m}}$) |
| D_p | primary particle diameter |
| R_p | primary particle radius |
| R_{gy} | radius of gyration |
| $\epsilon_{r,\text{mixture}}$ | relative dielectric constant of the mixture at analogous condition |
| P_{ij} | RLA correction matrix |
| B_{ij} | size dependence matrix |
| R_s | smallest radius of the sphere enclosing the aggregate |
| W | stability ratio |
| R_g | universal gas constant ($8.314 \frac{\text{J}}{\text{mol} \cdot \text{K}}$) |

Appendix A: Derivation of Reaction-Limited Aggregation Kernel

The reaction-limited aggregation kernel (RLA) kernel used in the study was derived from the following equation (Sandkühler *et al.*, 2005);

$$K_{ij} = K_B W^{-1} B_{ij} P_{ij} \quad (\text{A.1})$$

where
$$K_B = \frac{8R_g T}{3\eta} \quad (\text{A.2})$$

$$W = \frac{1}{\beta} \quad (\text{A.3})$$

$$B_{ij} = \frac{1}{4} \left(\frac{1}{R_{h,i}} + \frac{1}{R_{h,j}} \right) (R_{s,i} + R_{s,j}) \quad (\text{A.4})$$

$$P_{ij} = \left[\left(0.94 \left(\frac{R_{gy,i}}{R_p} \right)^{2.05} \right) \left(0.94 \left(\frac{R_{gy,j}}{R_p} \right)^{2.05} \right) \right]^{0.45} \quad (\text{A.5})$$

The subscripted i and j denote the i -th and j -th aggregate, respectively. Equation (A.1) becomes

$$K_{ij} = \left(\frac{8R_g T}{3\eta} \right) \left[\frac{1}{4} \left(\frac{1}{R_{h,i}} + \frac{1}{R_{h,j}} \right) (R_{s,i} + R_{s,j}) \right] \left[\left(0.94 \left(\frac{R_{gy,i}}{R_p} \right)^{2.05} \right) \left(0.94 \left(\frac{R_{gy,j}}{R_p} \right)^{2.05} \right) \right]^{0.45} \beta \quad (\text{A.6})$$

R_s and R_h can be related to R_{gy} using the following relationships;

$$R_s = 1.65R_{gy} \quad (\text{Lattuada } et al., 2003) \quad (\text{A.7})$$

$$R_h = 1.0R_{gy} \quad (\text{Lin } et al., 1990) \quad (\text{A.8})$$

Substituting those two terms yields

$$K_{ij} = \left(\frac{8R_g T}{3\eta} \right) \left[\frac{1.65}{4} \left(\frac{1}{R_{gy,i}} + \frac{1}{R_{gy,j}} \right) (R_{gy,i} + R_{gy,j}) \right] \left[\left(0.94 \left(\frac{R_{gy,j}}{R_p} \right)^{2.05} \right) \left(0.94 \left(\frac{R_{gy,i}}{R_p} \right)^{2.05} \right) \right]^{0.45} \beta \quad (\text{A.9})$$

Assuming silica particles to be spherical, the particle diameter (D) is approximately $2R_{gy}$. Rearranging equation (A.9) gives

$$K_{ij} = \frac{1.0404R_g T (D_i + D_j)^2}{\eta D_p^{1.85} D_i D_j} (D_i D_j)^{0.92} \beta \quad (\text{A.10})$$

By rounding the two non-integral exponent terms to be D_p^2 and $D_i D_j$, Equation (A.10) can be simplified to give

$$K_{ij} = \frac{1.0404R_g T}{\eta D_p^2} (D_i + D_j)^2 \beta \quad (\text{4.2})$$

Appendix B: Electrical Double Layer Thickness Calculation

The double layer thickness ($1/\kappa$) is defined as the reciprocal term of Debye length (κ). The debye length is a characteristic distance which the electric potential energy of a charged particle is shielded by particles having charges of the opposite sign, resulting in the decrease in the potential energy from Φ to $(1/e)\Phi$ (0.369Φ) where Φ is the total potential energy at the surface. The expression of Debye length is given by (Hiemenz and Rajagopalan, 1997)

$$\kappa = \left[\frac{1000e^2 N_A}{\epsilon_{r,mixture} \epsilon_o k_B T} 2I \right]^{1/2} \quad (B.1)$$

From equation (B.1), one notes that the Debye length is proportional to the square root of ionic strength and is also a function of the relative dielectric constant of the mixtures, and temperature. The relative dielectric constant of the mixture at 5°C ($\epsilon_{r,mixture}$) is calculated using the Lichteneker's empirical logarithmic mixing rule (Langton and Matthews, 1958);

$$\log \epsilon_{r,mixture} = v_{water} \log \epsilon_{r,water,5^\circ C} + v_{HCl} \log \epsilon_{r,HCl,5^\circ C} + v_{salt} \log \epsilon_{r,salt,5^\circ C} \quad (B.2)$$

where $\epsilon_{r,mixture}$ = relative dielectric constant of the final mixture,
 $\epsilon_{r,water,5^\circ C}$ = reactive dielectric constants of water at 5°C,
 $\epsilon_{r,HCl,5^\circ C}$ = reactive dielectric constants of HCl at 5°C,
 $\epsilon_{r,salt,5^\circ C}$ = reactive dielectric constants of salt at 5°C,
 v_{water} = volumetric fraction of water,
 v_{HCl} = volumetric fraction of HCl,
and v_{salt} = volumetric fraction of salt,

such that $v_{water} + v_{HCl} + v_{salt} = 1$.

The calculated double layer thicknesses of silica particles in different medium are listed in Table B.1.

Table B.1 Calculated double layer thicknesses in different mediums.

| Medium | Double layer thickness, Å |
|-------------------------------|---------------------------|
| 4M HCl (base case) | 1.290 |
| 4M HCl + 1M CsCl | 1.072 |
| 4M HCl + 1M NaCl | 1.116 |
| 4M HCl + 1M MgCl ₂ | 0.928 |
| 4M HCl + 1M CaCl ₂ | 0.895 |
| 4M HCl + 1M AlCl ₃ | 0.750 |
| 4M HCl + 1M NaNO ₃ | 1.095 |
| 4M HCl + 1M NaBr | 1.106 |
| 4M HCl + 1M NaI | 1.096 |

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