

1 **Supplement of**  
2 **Surfactants Regulate the Mixing State of Organic-Inorganic Mixed**  
3 **Aerosols Undergoing Liquid-Liquid Phase Separation**

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## **S1. Influence of SDS on the Mixing State of OIR = 1:4 and 4:1 Particles**

For the particle with OIR = 1:4 without SDS, LLPS occurs via the growth of a second phase from the particle surface at RH values between 74.3% and 70.9% (Figure S3a). Following efflorescence, the organic phase becomes trapped within the cavity of the AS crystal. These scenarios are consistent with our previous observations (Ma et al., 2021a). Upon adding SDS, the LLPS mechanism changes to spinodal decomposition, as depicted in Figure S3b and S3c. During this process, schlieren patterns appear over the aqueous droplet, followed by the growth and coalescence of separated AS inclusions (Ma et al., 2021a; Ciobanu et al., 2009). Notably, as the SDS fraction increases, such as in the org:sulf:SDS = 1:4:0.0075 mixture, a core-shell morphology is no longer observed; instead, a partial engulfing structure forms (Figure S3c). Further increases in SDS concentration, seen in the 1:4:0.008 and 1:4:0.01 mixtures (Figure S4a and S4b), still result in the persistence of partial organic-phase engulfing.

Figure S5 illustrates the phase transition behaviors of mixed 1,2,6-hexanetriol/AS/SDS particles with org:sulf:SDS ratios of 4:1:0.004, 4:1:0.0045, and 4:1:0.005. For the 4:1:0.004 mixture, LLPS occurs at 77.9% RH through a nucleation and growth mechanism (Ma et al., 2021a; Ciobanu et al., 2009), resulting in a core-shell particle morphology. In contrast, for the 4:1:0.0045 and 4:1:0.005 mixtures, LLPS occurs via spinodal decomposition, leading to the formation of numerous dispersed AS inclusions. Coupled with the particle morphologies observed

after efflorescence, we can infer that these AS inclusions tend to reside on the particle surface, indicative of a partial engulfing structure.

It is evident that the SDS concentration threshold regulating the dominance of core-shell versus partial engulfing morphologies differs between the OIR = 1:4 and 4:1 systems compared to the OIR = 1:1 particles. To further explore this, we compare the changes in SDS concentration and surface tension with decreasing RH for the org:sulf:SDS = 1:1:0.001, 1:4:0.0075, and 4:1:0.0045 systems, as shown in Figure S6. At the SRH, the three systems exhibit distinct SDS concentrations of 2.71, 7.27, and 5.10 mM, with corresponding surface tensions of 68.49, 53.15, and 25.10 mN m<sup>-1</sup>, respectively. These discrepancies imply a complex correlation between particle OIR and the surfactant concentration thresholds.

The change in total surface free energy ( $G_s$ ) during spreading depends not only on the spreading coefficients ( $S_i$ ) but also on the changes in surface and interface areas of the organic and inorganic phases ( $dA$ ) (Torza and Mason, 1970; Shaw, 1992):

$$dG_s = -S_i dA \quad (S1)$$

Thus, while the three systems have varying concentrations of organic and inorganic phases, resulting in distinct surface and interfacial tensions during LLPS, the differences in SDS concentration thresholds can be partially attributed to different volume ratios. This is another key factor affecting total surface free energy, in addition to interfacial tensions. Similar to previous studies, our findings emphasize the importance of considering both interfacial tensions and the volumes of separated

139 phases, related to particle OIR, for accurately predicting final particle morphology  
140 (Kwamena et al., 2010).

## S2. Surface Tension Predictions for Aqueous Systems with Decreasing RH

Wexler and Dutcher (2013) developed an equation based on statistical mechanics to describe the relationship between the surface tension ( $\sigma$ ) of a solution and solute activity ( $a_A$ ):

$$\sigma = \sigma_W + \frac{kT}{rS_W} \ln \left( \frac{1-Ka_A}{1-Ka_A(1-C)} \right) \quad (\text{S2})$$

where  $\sigma_W$  is the surface tension of pure water ( $\text{N m}^{-1}$ ),  $k$  is Boltzmann's constant ( $\text{J K}^{-1}$ ),  $T$  is the ambient temperature ( $\text{K}$ ), and  $S_W$  is the area occupied by a water molecule, typically assumed to be  $0.1 \text{ nm}^2$ . The remaining three unknown parameters,  $r$ ,  $K$ , and  $C$ , are specific to each solute, with  $r$  representing the average number of water molecules displaced from the surface by each solute molecule, while  $K$  and  $C$  relate to sorption energies.

For aqueous AS solution, Boyer et al. (2015) estimated values of  $r = -4.04$  and  $K = 0.99$ , based on electrolyte fits using Equation (S2). Coupled with the surface tension of pure solute ( $\sigma_S = 184.99 \text{ mN m}^{-1}$  for molten AS) (Dutcher et al., 2010), the parameter  $C$  is calculated as 670.39 using the formula  $C = 1 - [1 - (1 - K)\exp(rS_W(\sigma_W - \sigma_S)/kT)]/K$  (Wexler and Dutcher, 2013). Therefore, the surface tension of sulfate-containing aqueous droplets can be predicted as a function of solute activity, which varies with decreasing RH, based on the evolution of AS concentration.

Dutcher et al. (2011) extended the Brunauer-Emmett-Teller (BET) and Guggenheim-Anderson-de Boer (GAB) adsorption models to account for additional



adsorbed monolayers when determining solute and solvent activities in aqueous solutions over a wide concentration range. Assuming each solute entity is surrounded by three hydrated layers of water molecules, the solute molality ( $m_A$ ) can be expressed as a function of water activity ( $a_W$ ) according to the three-layer model (Dutcher et al., 2011):

$$m_A = \left( \frac{1 - a_W K_A}{M_W r_A C_{A,1} a_W K_A} \right) \left( \frac{1 - K_A a_W (1 - C_{A,1}) - K_A^2 a_W^2 C_{A,1} (1 - C_{A,2})}{(1 - a_W K_A)^2 + (2 - a_W K_A) a_W K_A C_{A,2}} \right) \quad (S3)$$

The solute activity ( $a_A$ ) can also be expressed as a function of water activity ( $a_W$ ) (Dutcher et al., 2011):

$$a_A = \left( \frac{1 - K_A a_W}{1 - K_A a_W (1 - C_{A,1}) - K_A^2 a_W^2 C_{A,1} (1 - C_{A,2})} \right)^{r_A} \quad (S4)$$

Here,  $M_W$  is the molecular weight of water (the adsorbate) ( $\text{kg mol}^{-1}$ ),  $r_A$  represents the number of adsorption sites on solute A, while  $K_A$ ,  $C_{A,1}$ , and  $C_{A,2}$  are energy parameters related to the exponent of the difference between the energy of adsorption to a layer and the bulk energy of liquefaction. In Dutcher's later work, the values for the aqueous AS solution were determined to be  $r_A = 1.872$ ,  $K_A = 0.9876$ ,  $C_{A,1} = 0.0812$ , and  $C_{A,2} = 34.77$  (Dutcher et al., 2013). Using the EAIM model, the molality of aqueous AS across the entire RH range for the OIR = 1:1 system can be simulated, allowing  $a_W$  and  $a_A$  at specific RH levels to be calculated using Equations (S3) and (S4). Combined with the relationship between surface tension and solute activity in Equation (S2), the surface tension of the mixed droplets (org:sulf:SDS = 1:1:0.001) can be predicted, considering only AS concentration changes as RH decreases.

For the contribution of SDS to droplet surface tension, the empirical Szyskowski equation can be applied (Szyskowski, 1908):

$$\sigma = \sigma_W - a \ln \left( 1 + \frac{c_{\text{SDS}}}{b} \right) \quad (\text{S5})$$

where  $c_{\text{SDS}}$  is the concentration of SDS ( $\text{mol L}^{-1}$ ), and parameters  $a$  and  $b$  are determined by fitting surface tension data of pure SDS solutions, measured using the hanging droplet method (Figure 3c). The evolution of SDS concentration with decreasing RH is evaluated based on the fixed molar ratio of AS to SDS.

When considering both AS and SDS contributions, the surface tension parametrization for the ternary SDS/AS aqueous solution is obtained with the modified Szyskowski equation (Prisle et al., 2010):

$$\sigma = \sigma_W + \left( \frac{d\sigma_{\text{AS}}}{dc_{\text{AS}}} \right) c_{\text{AS}} - a \ln \left( 1 + \frac{c_{\text{SDS}}(c_{\text{AS}} + b_2)}{b_1} \right) \quad (\text{S6})$$

As the solutions are dilute, the gradient of surface tension for aqueous AS ( $\sigma_{\text{AS}}$ ) with respect to its molar concentration ( $c_{\text{AS}}$ ), denoted by  $\left( \frac{d\sigma_{\text{AS}}}{dc_{\text{AS}}} \right)$ , was measured to be  $2.362 \text{ mN m}^{-1}/\text{mol L}^{-1}$  (Svenningsson et al., 2006). Surface tension depression in SDS/AS aqueous solutions was determined for fixed AS concentrations of 0, 0.1, or 1  $\text{mol L}^{-1}$ , using the hanging droplet method (Figure 4c). The least squares method was applied to determine the optimal values of  $a$ ,  $b_1$ , and  $b_2$ . Consequently, the surface tension can be estimated for specific AS and SDS concentrations at given RH levels, as predicted by the EAIM model.

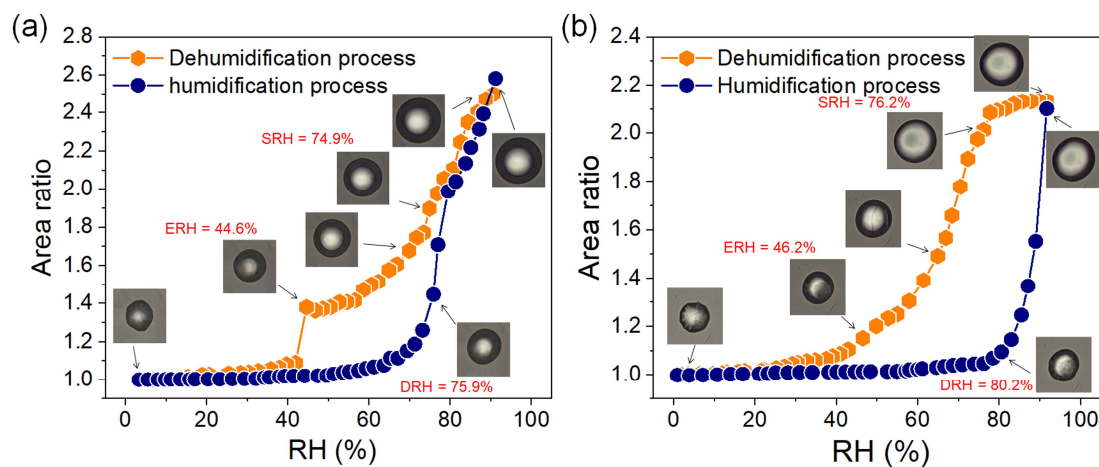
### **S3. Changes in SRH and ERH with Varying Initial Surfactant Concentrations**

Figure S17 illustrates the changes in SRH and ERH values for OIR = 1:4, 1:1, and 4:1 particles in the presence of surfactants (SDS, CTAC, and Triton X-100) at different initial concentrations, with 1,2,6-hexanetriol and AS set at 0.1 or 0.4 mol L<sup>-1</sup>. The original SRH and ERH values for mixed particles without surfactants are assumed to be 76% and 45%, respectively. As shown in Figure S17a, at relatively low surfactant concentrations, surfactant molecules tended to form a thin organic layer on the particle surface, inhibiting water transport across the aqueous-air interface and reducing SRH values (Faust and Abbatt, 2019; Miñambres et al., 2014). For instance, the SRH of OIR = 1:1 particles decreases by approximately 9.0% with the addition of 0.001 mM CTAC. As surfactant concentration increases, the enhanced salting-out effect raises the SRH, reaching up to 90.6% RH for OIR = 1:1 particles with 100 mM Triton X-100.

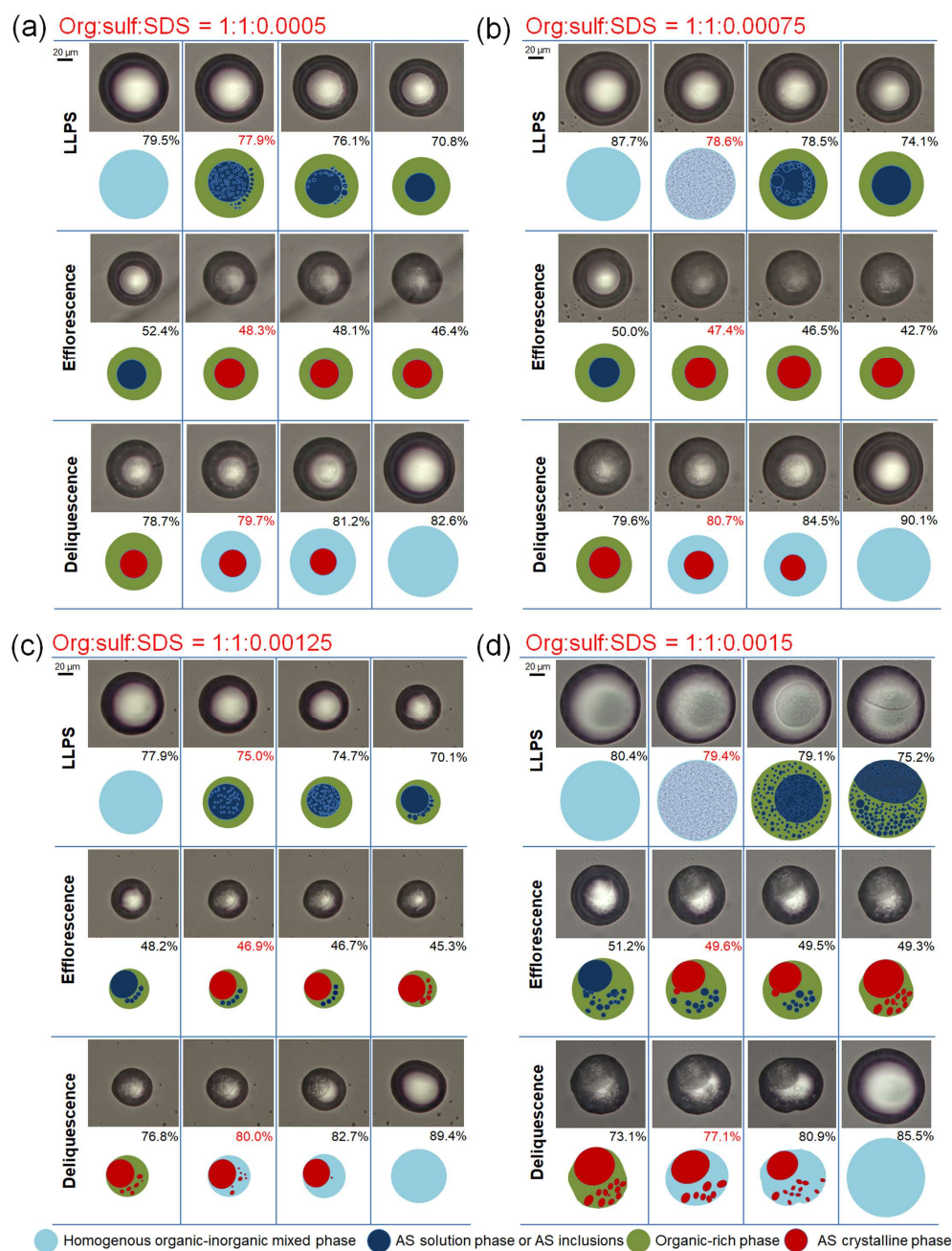
In ERH measurements, low concentrations of ionic surfactants (SDS and CTAC) reduce the ERH of mixed particles (Figure S17b). For example, 0.001 mM CTAC lowers the ERH to 38.3% due to the inhibiting effect of the surface surfactant monolayer (Harmon et al., 2010). Conversely, high concentrations of ionic surfactants promote efflorescence of aqueous droplets at higher RH compared to the surfactant-free system. Considering the very high ERH of the pure SDS system (Figure S18), we infer that the SDS efflorescence induced the heterogenous nucleation of AS (Ma et al., 2021b). Additionally, Triton X-100 inhibits AS

225   efflorescence across the entire concentration range due to its lack of heterogenous  
226   efficacy.

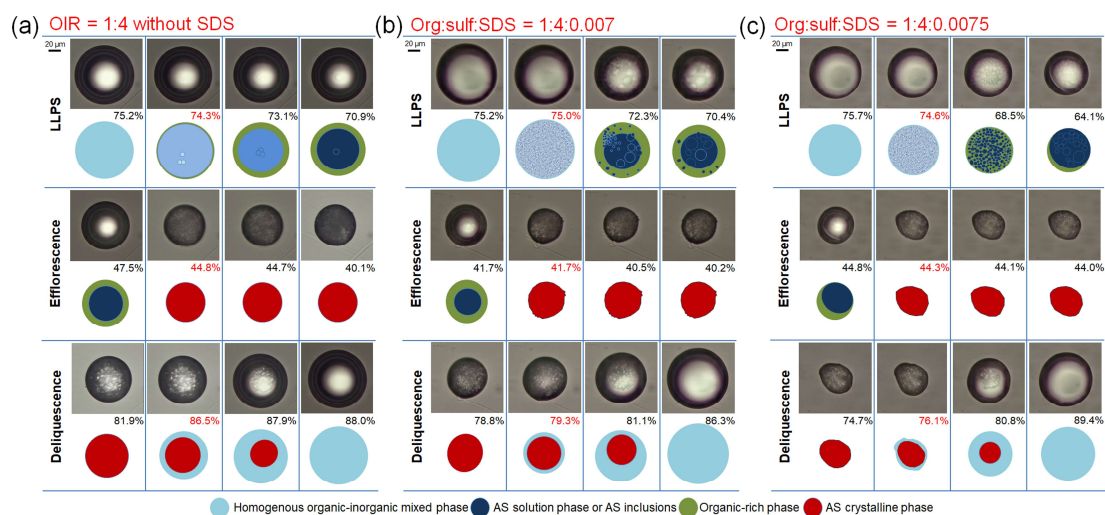
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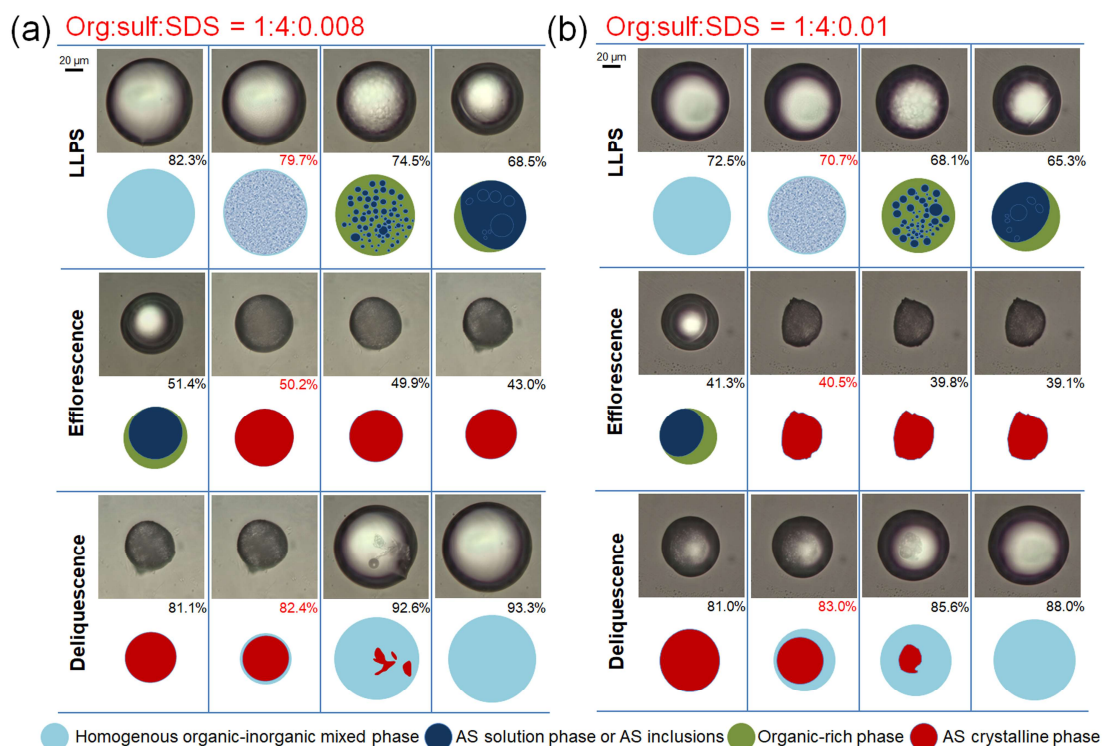
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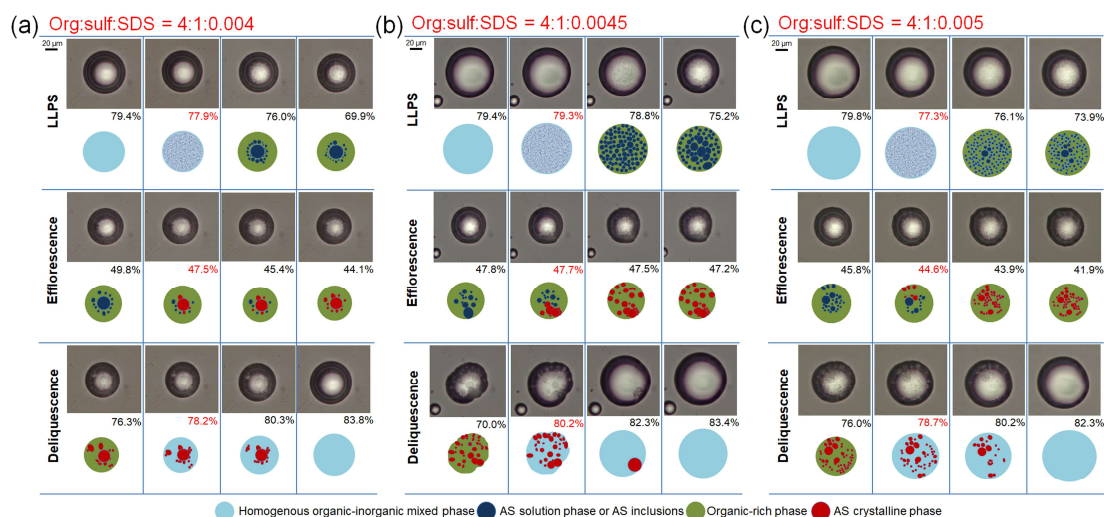


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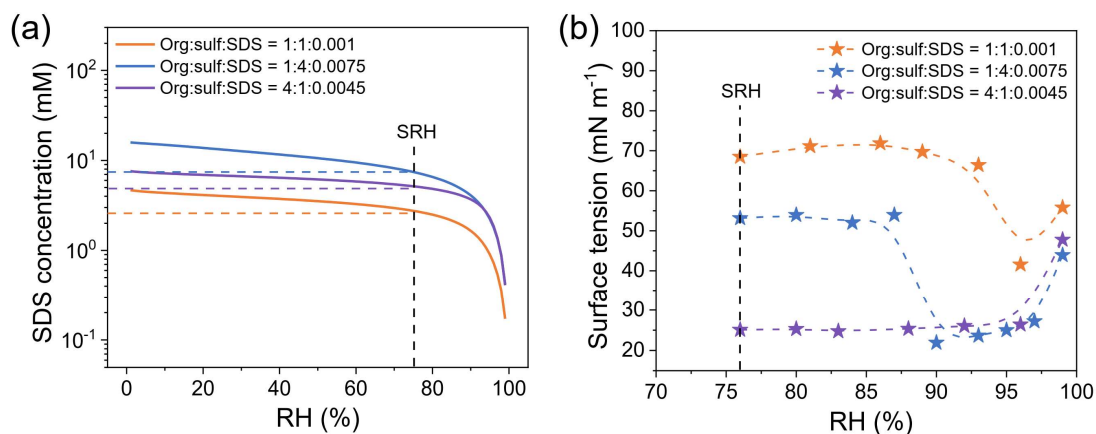


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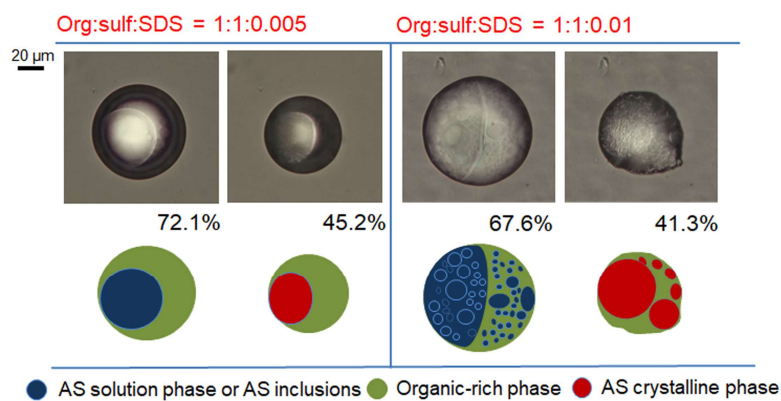




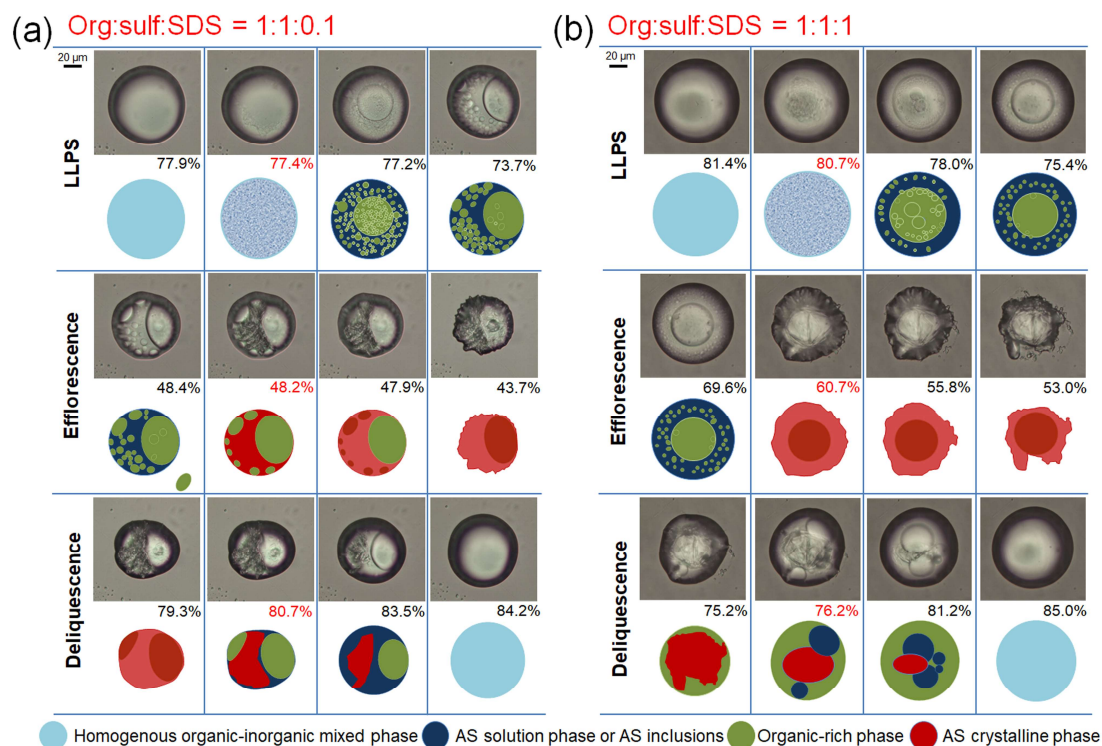
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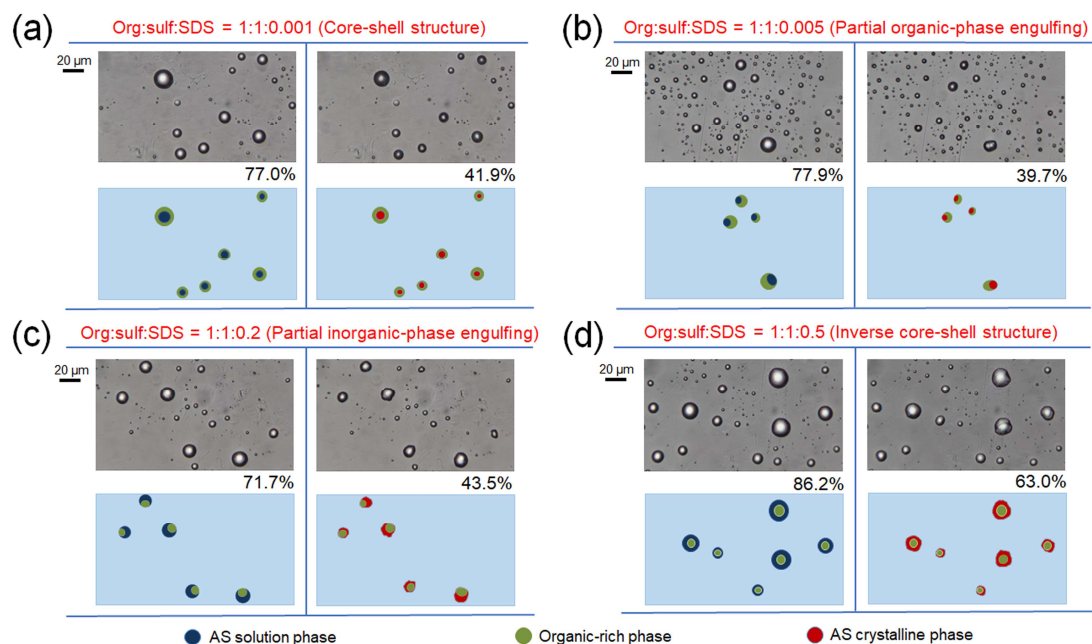
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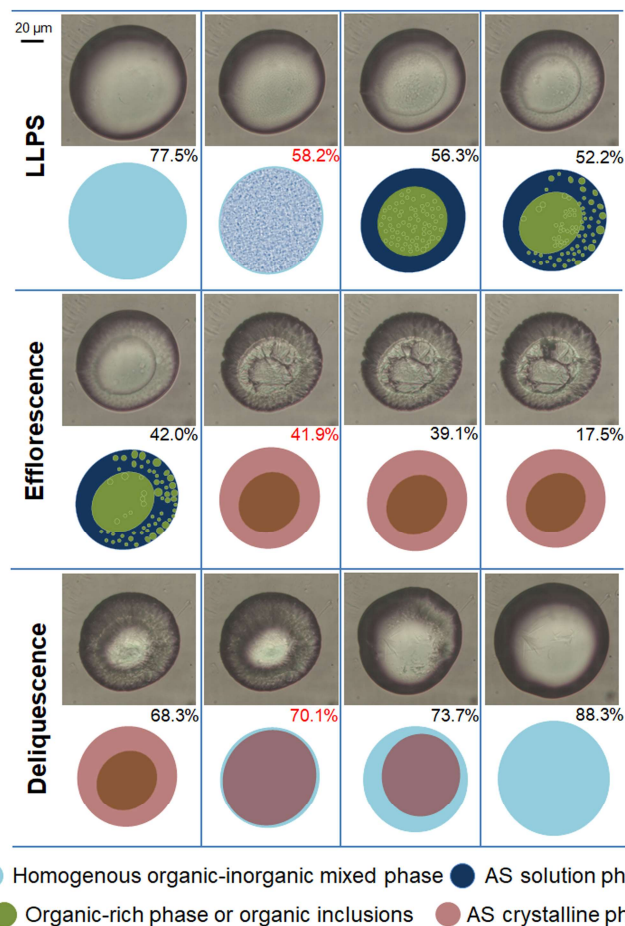
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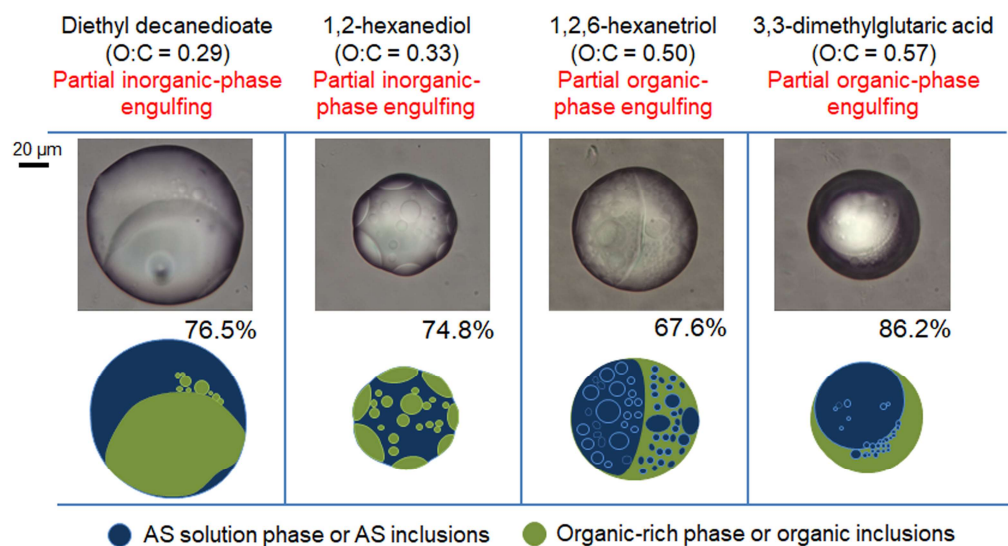
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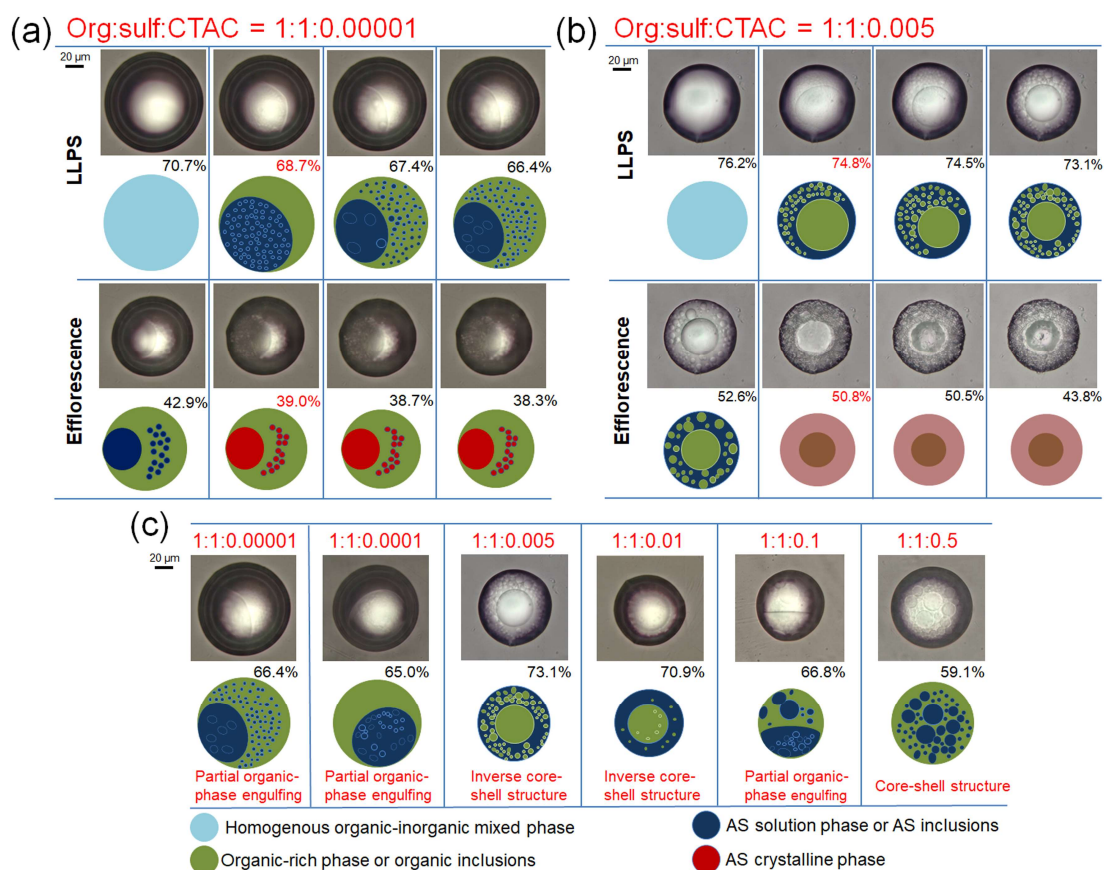


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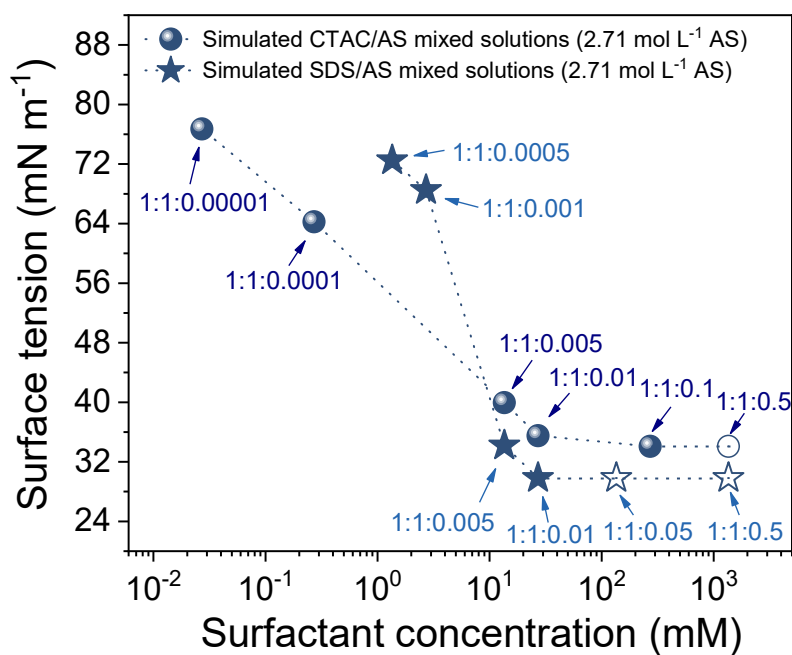
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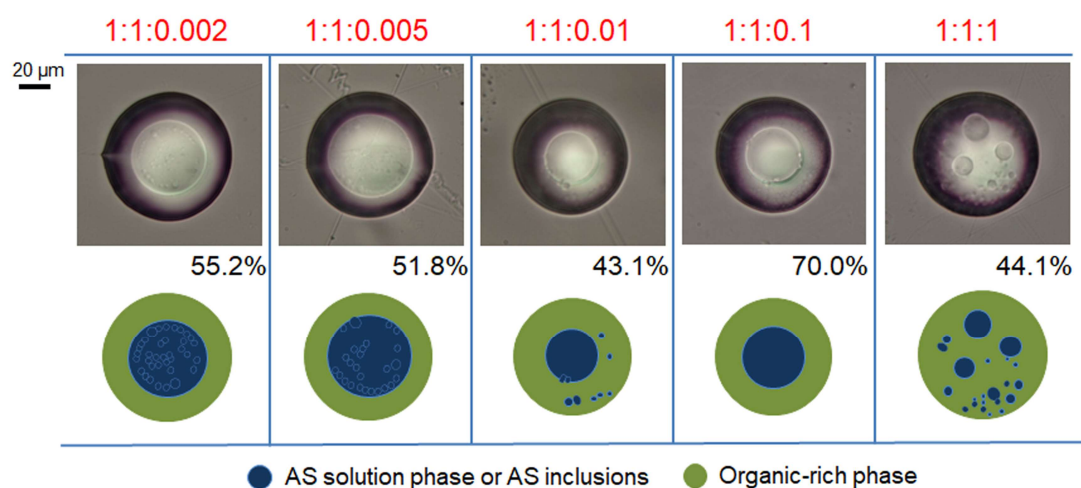


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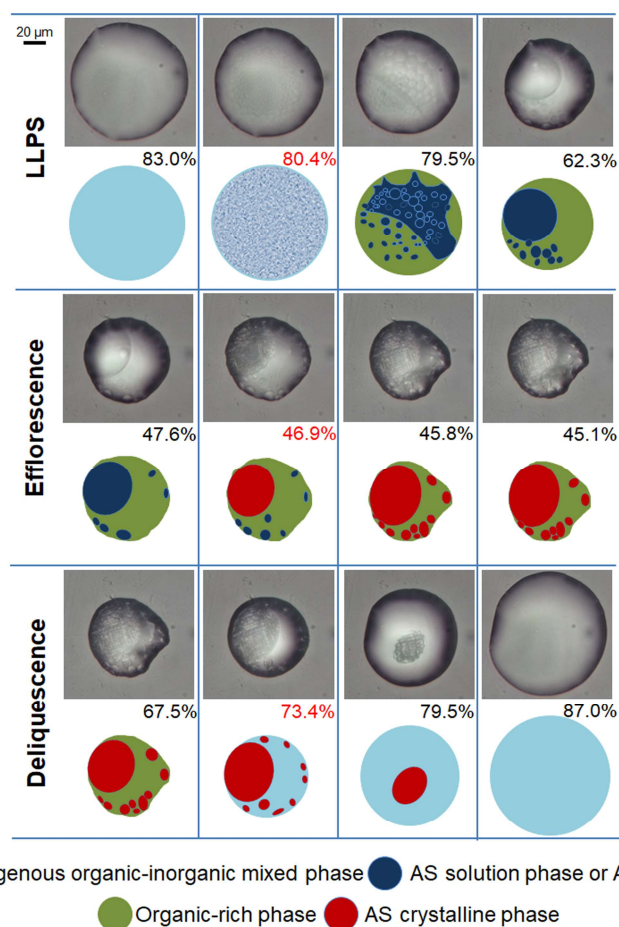




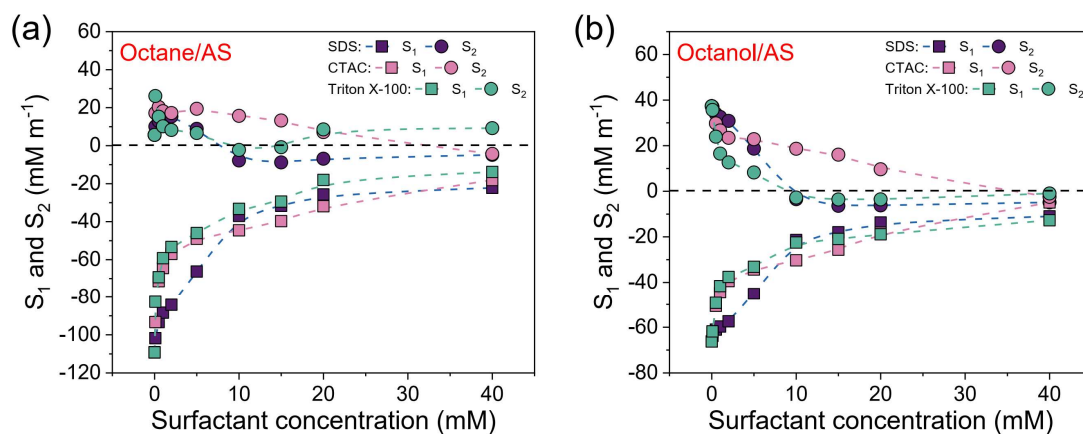
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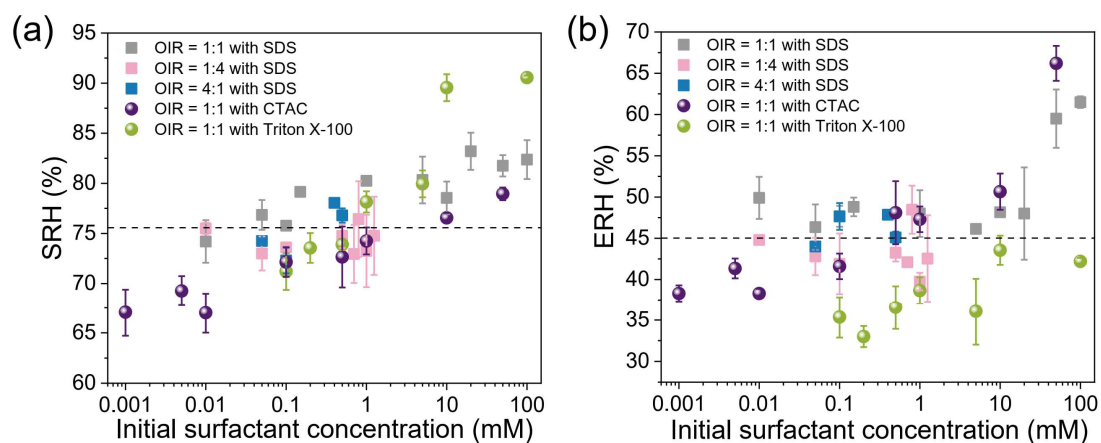
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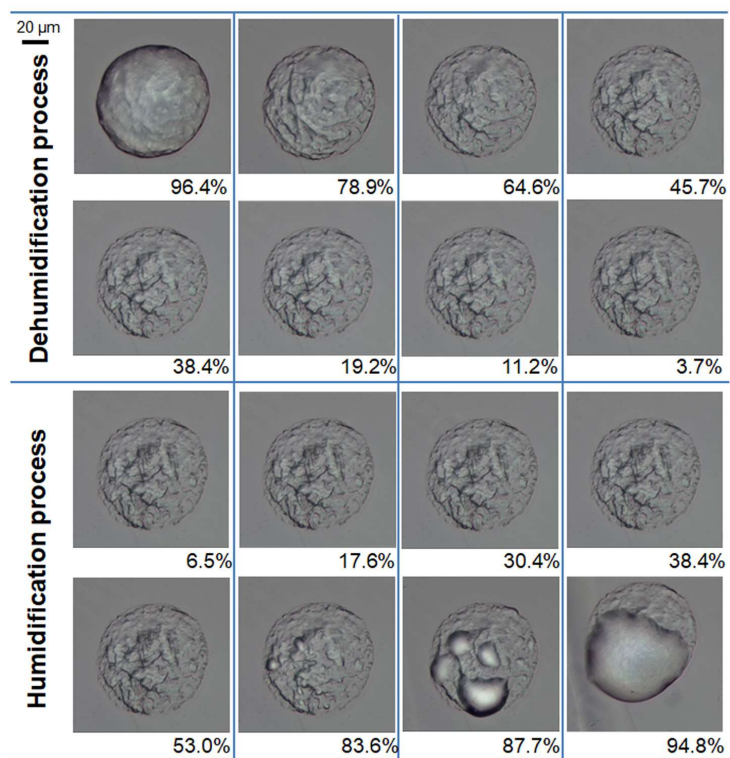
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## References:

- Boyer, H., Wexler, A., and Dutcher, C. S.: Parameter interpretation and reduction for a unified statistical mechanical surface tension model, *J. Phys. Chem. Lett.*, 6, 3384-3389, <https://doi.org/10.1021/acs.jpcelett.5b01346>, 2015.
- Ciobanu, V. G., Marcolli, C., Krieger, U. K., Weers, U., and Peter, T.: Liquid-liquid phase separation in mixed organic/inorganic aerosol particles, *J. Phys. Chem. A*, 113, 10966-10978, <https://doi.org/10.1021/jp905054d>, 2009.
- Dutcher, C. S., Wexler, A. S., and Clegg, S. L.: Surface tensions of inorganic multicomponent aqueous electrolyte solutions and melts, *J. Phys. Chem. A*, 114, 12216-12230, <https://doi.org/10.1021/jp105191z>, 2010.
- Dutcher, C. S., Ge, X., Wexler, A. S., and Clegg, S. L.: Statistical mechanics of multilayer sorption: Extension of the Brunauer–Emmett–Teller (BET) and Guggenheim–Anderson–de Boer (GAB) adsorption isotherms, *J. Phys. Chem. C*, 115, 16474-16487, <https://doi.org/10.1021/jp203879d>, 2011.
- Dutcher, C. S., Ge, X., Wexler, A. S., and Clegg, S. L.: An isotherm-based thermodynamic model of multicomponent aqueous solutions, applicable over the entire concentration range, *J. Phys. Chem. A*, 117, 3198-3213, <https://doi.org/10.1021/jp310860p>, 2013.
- Faust, J. A., and Abbatt, J. P. D.: Organic surfactants protect dissolved aerosol components against heterogeneous oxidation, *J. Phys. Chem. A*, 123, 2114-2124, <https://doi.org/10.1021/acs.jpca.9b00167>, 2019.
- Harmon, C. W., Grimm, R. L., McIntire, T. M., Peterson, M. D., Njegic, B., Angel, V. M., Alshawa, A., Underwood, J. S., Tobias, D. J., and Gerber, R. B.: Hygroscopic growth and deliquescence of NaCl nanoparticles mixed with surfactant SDS, *J. Phys. Chem. B*, 114, 2435-2449, <https://doi.org/10.1021/jp909661q>, 2010.
- Kwamena, N. O. A., Buajarn, J., and Reid, J. P.: Equilibrium morphology of mixed organic/inorganic/aqueous aerosol droplets: Investigating the effect of relative humidity and surfactants, *J. Phys. Chem. A*, 114, 5787-5795, <https://doi.org/10.1021/jp1003648>, 2010.
- Ma, S., Chen, Z., Pang, S., and Zhang, Y.: Observations on hygroscopic growth and phase transitions of mixed 1, 2, 6-hexanetriol/(NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> particles: investigation of the liquid–liquid phase separation (LLPS) dynamic process and mechanism and secondary LLPS during the dehumidification, *Atmos. Chem. Phys.*, 21, 9705-9717, <https://doi.org/10.5194/acp-21-9705-2021>, 2021a.
- Ma, S., Pang, S., Li, J., and Zhang, Y.: A review of efflorescence kinetics studies on atmospherically relevant particles, *Chemosphere*, 277, 130320, <https://doi.org/10.1016/j.chemosphere.2021.130320>, 2021b.
- Miñambres, L., Méndez, E., Sánchez, M. N., Castaño, F., and Basterretxea, F. J.: The effect of low solubility organic acids on the hygroscopicity of sodium halide aerosols, *Atmos. Chem. Phys.*, 14, 11409-11425, <https://doi.org/10.5194/acp-14-11409-2014>, 2014.
- Prisle, N. L., Raatikainen, T., Laaksonen, A., and Bilde, M.: Surfactants in cloud droplet activation: mixed organic-inorganic particles, *Atmos. Chem. Phys.*, 10, 5663-5683, <https://doi.org/10.5194/acp-10-5663-2010>, 2010.
- Shaw, D. J.: Introduction to colloid and surface chemistry: 4th edition, Butterworth-Heinemann, Oxford, U.K., 1992.
- Svenningsson, B., Rissler, J., Swietlicki, E., Mircea, M., Bilde, M., Facchini, M. C., Decesari, S., Fuzzi, S., Zhou, J., Mønster, J., and Rosenørn, T.: Hygroscopic growth and critical supersaturations for mixed aerosol particles of inorganic and organic compounds of atmospheric relevance, *Atmos. Chem. Phys.*, 6,

357 1937-1952, <https://doi.org/10.5194/acp-6-1937-2006>, 2006.  
 358 Szyskowski, B. V.: Experimentelle studien über kapillare eigenschaften der wässrigen lösungen von  
 359 fettsäuren, Zeitschrift für Physikalische Chemie, 64, 385-414, 1908.  
 360 Torza, S., and Mason, S. G.: Three-phase interactions in shear and electrical fields, J. Colloid Interface  
 361 Sci., 33, 67-83, [https://doi.org/10.1016/0021-9797\(70\)90073-1](https://doi.org/10.1016/0021-9797(70)90073-1), 1970.  
 362 Wexler, A. S., and Dutcher, C. S.: Statistical mechanics of multilayer sorption: Surface tension, J. Phys.  
 363 Chem. Lett., 4, 1723-1726, <https://doi.org/10.1021/jz400725p>, 2013.