

**Supporting information for**

**Experimental Phase Diagram and Its Temporal Evolution for Submicron  
2-Methylglutaric Acid and Ammonium Sulfate Aerosol Particles**

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**Summary:** The content of this supporting information includes the size distribution of submicron 2MGA+AS particles, example optical images of micrometer droplets at different RH values, the measured ERH of submicron inorganic aerosol particles and literature values (KCl, K<sub>2</sub>SO<sub>4</sub>), and the AIOMFAC prediction of the viscosity of 2MGA+AS aerosols.

## Experimental Methods

### Supplementary Figures and Tables

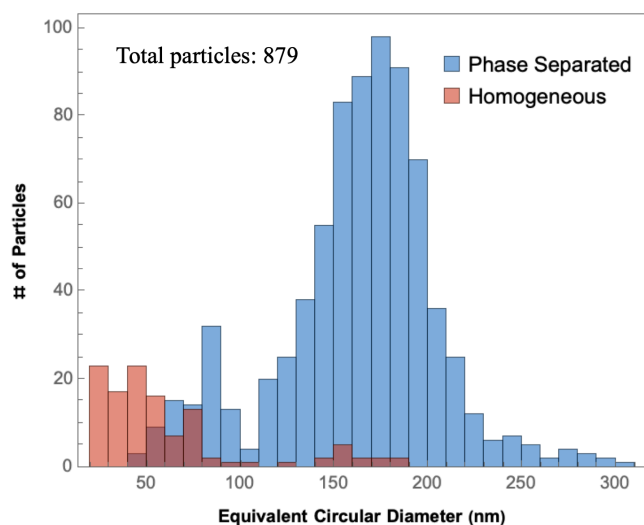


Figure S1. Size distribution of 2MGA+AS aerosols ( $mf_d(AS) = 0.33$ ). The images were collected using a diffusion dryer coupled with cryo-TEM. LLPS is inhibited for particles smaller than 30 nm.

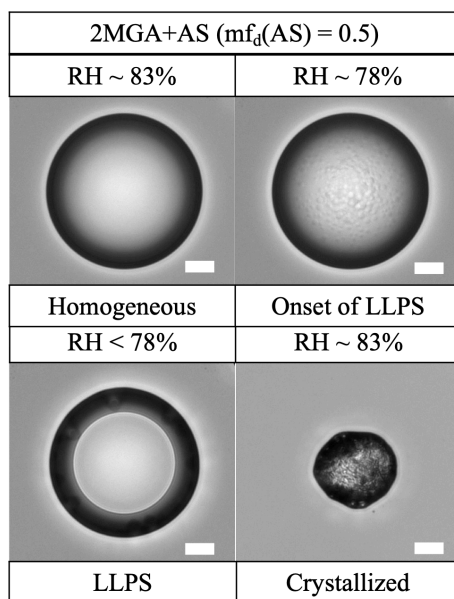


Figure S2. Example optical images of micrometer droplets ( $mf_d(AS) = 0.5$ ), at different RH conditions. The labels under the images denote different phase states of droplets. Scale bar = 2  $\mu\text{m}$ .

Table S1. The ERH of KCl and K<sub>2</sub>SO<sub>4</sub> measured in this study experimentally and measured in the literature by Morris et al.<sup>1</sup> The temperature of this study was 297 ± 1 K, which is overall consistent with Morris et al with variation in temperature about 1K.

Composition	Experimental ERH (%)	Literature value(%)
KCl	56 ± 1	56 ± 2
K <sub>2</sub> SO <sub>4</sub>	59 ± 1	60 ± 1

Table S2. The estimated viscosity of 2MGA+AS aerosol particles with various compositions at ~78% RH and 298K. The 78% RH is close to the SRH of micrometer-sized 2MGA+AS aerosol particles.

mf <sub>d</sub> (AS)	RH	log <sub>10</sub> ( η(Pa · S))
0.10	78.33	-1.612
0.20	78.29	-1.811
0.33	78.50	-2.014
0.50	78.68	-2.172
0.80	78.50	-2.385
0.90	78.34	-2.470

#### References:

- 1 H. S. Morris, A. D. Estillore, O. Laskina, V. H. Grassian and A. V. Tivanski, *Anal. Chem.*, 2016, **88**, 3647–3654.