

Supporting Information

Reactive Uptake of Dimethylamine by Ammonium Sulfate and Ammonium Sulfate – Sucrose Mixed Particles

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S1. Reactivity of sucrose particles against dimethylamine (DMA)

The reaction between sucrose particle and dimethylamine (DMA) gas was also investigated. Figure S2 shows that at 70% RH, the mass of a sucrose droplet remained unchanged (within experimental uncertainty) under the exposure of 1 ppm DMA vapor for 265 min. Reaction between sucrose and DMA at low RH was not investigated, since drying a sucrose droplet readily induces a transition from liquid to glassy state and leads to ultra-slow diffusion of water.^{1,2} According to Zobrist et al., a sucrose particle with $\sim 7\ \mu\text{m}$ in diameter has never reached water equilibrium at 2% RH after one day.² In our case where the particles were even larger, the drying process is expected to take longer time and may result in significant particle charge loss and thus an overestimated particle mass. Nevertheless, it is clear that DMA uptake onto sucrose alone is negligible.

S2. Confirmation of the presence of sucrose-rich coating layer at < 5% RH for AS – sucrose ($F_{\text{su}} = 0.50$) particle

The presence of a sucrose-rich coating layer was confirmed based on vertical distribution of Raman intensities captured by the CCD detector, as described in our previous EDB experiments.^{3,4} In the CCD, the horizontal pixel is the Raman shift (cm^{-1}) and the vertical pixel represents the vertical position of the particle. Color represents the Raman spectral intensity. As shown in Figure S3 (see below), the C–H stretching band ($2970 - 3035\ \text{cm}^{-1}$) intensities was more evenly distributed at 70% RH

(Figure S3c) along the vertical direction than that at < 5% RH (Figure S3a), although the AS – sucrose ($F_{\text{su}} = 0.50$) particles may have focusing effect that exhibits bright spots near the particle top and bottom. Besides, the C–H stretching peak intensities were higher at the polar regions and lower at the core of the particle, if the particle is dried under < 5% RH for 11 hours (Figure S3b) compared to that for only 2 hours (Figure S3a). This confirms the heterogeneity of the particle, i.e., presence of a sucrose-rich coating layer.

Figures

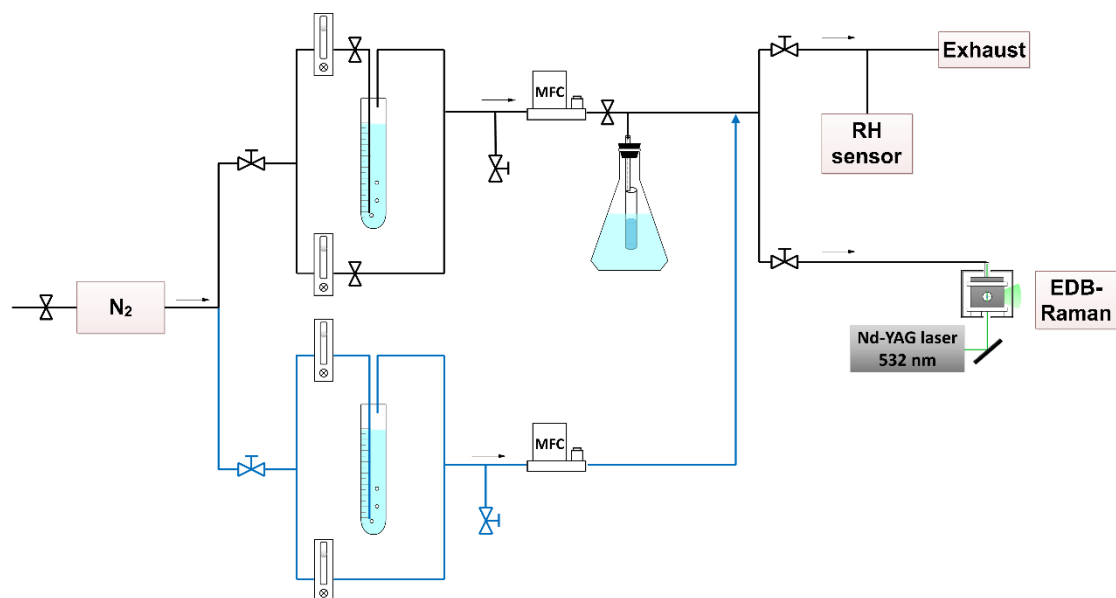


Figure S1. A Schematic of the flow system. The DMA-free stream, shown in blue, is connected to the EDB for conditioning the levitated particle at particular RH before DMA uptake as well as drying the particle at the end of each experiment.

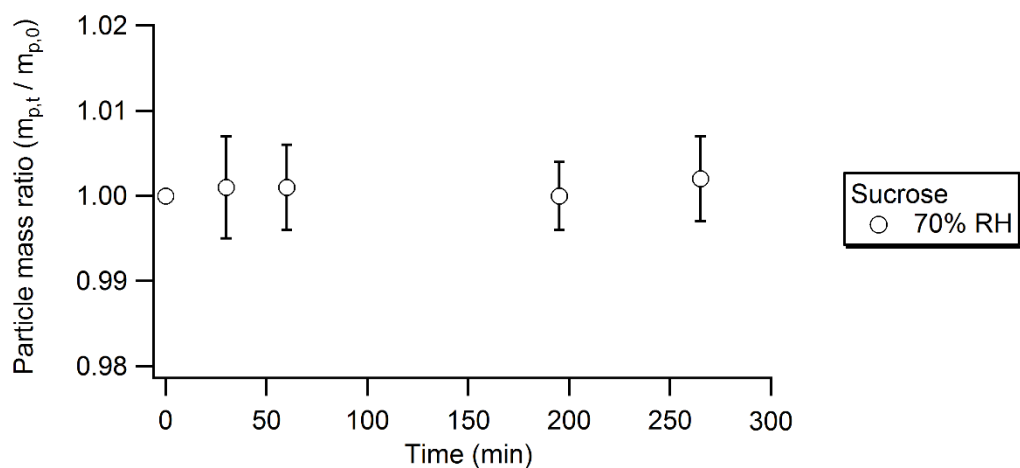


Figure S2. Relative mass changes in pure sucrose particle in the presence of DMA vapor at 70% RH.

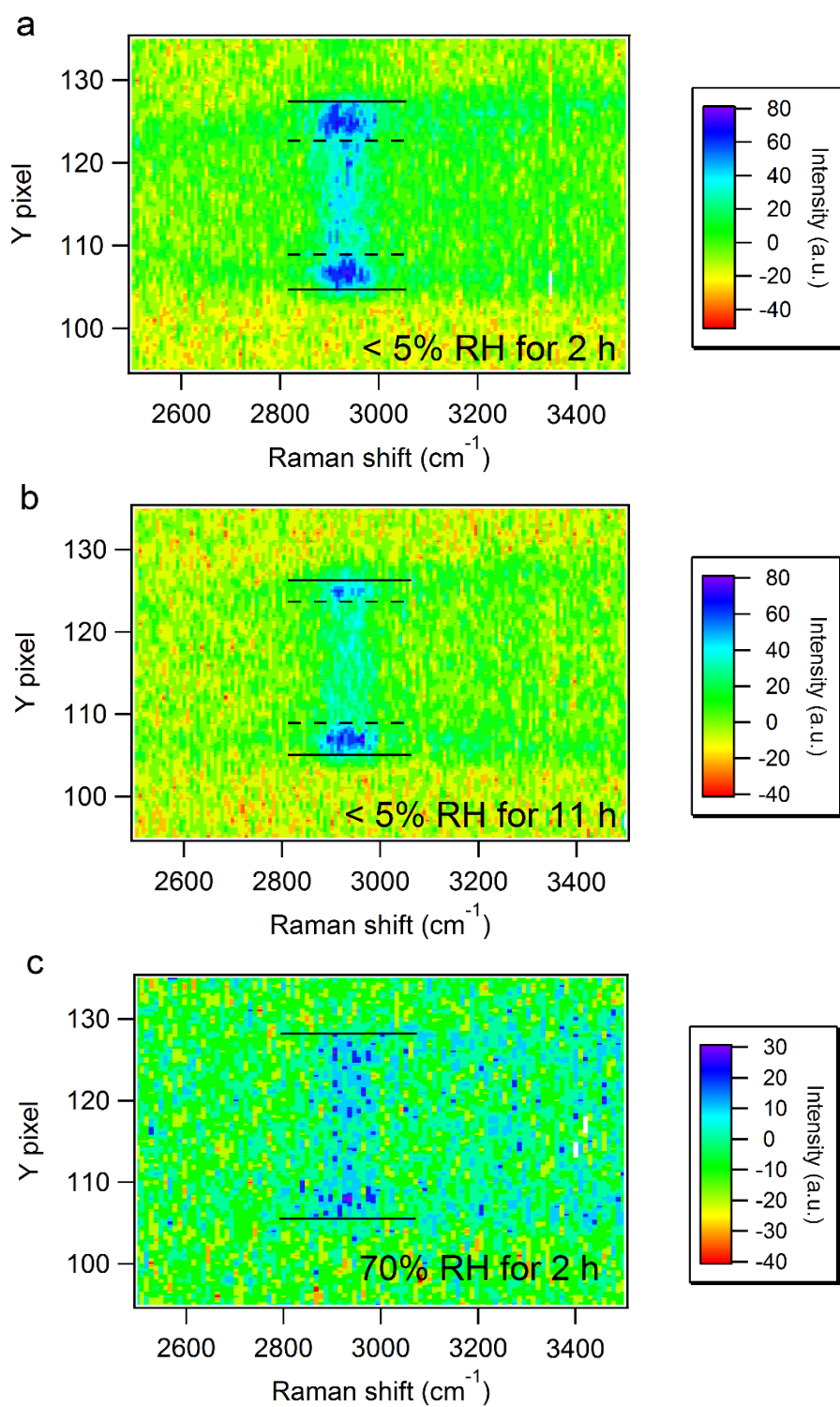


Figure S3. Spatially resolved Raman intensities of AS – sucrose ($F_{\text{su}} = 0.50$) particles equilibrated at < 5% RH for (a) 2 hours and (b) 11 hours and at (c) 70% RH for 2 hours. Solid lines indicate the particle upper and lower edges. The outer regions between the dashed and solid lines are sucrose-rich in (a) and (b).

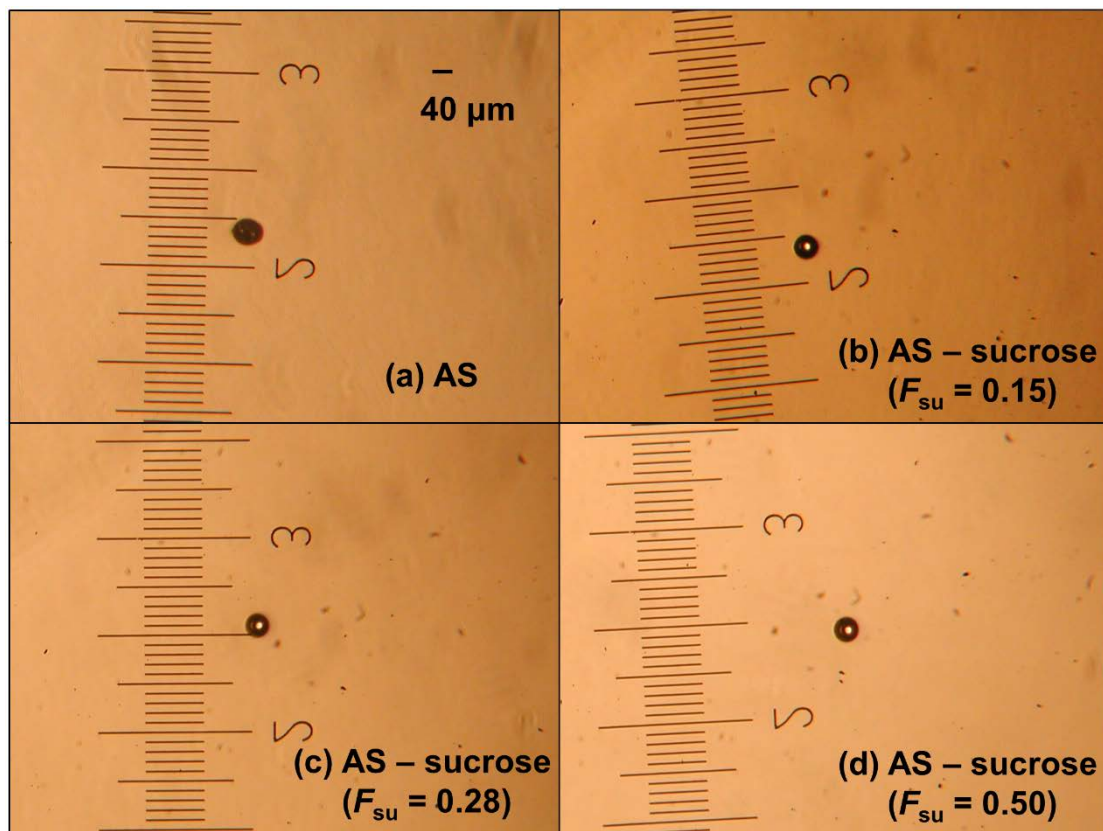


Figure S4. Microscopic images of (a) AS and AS – sucrose particles at $F_{\text{su}} =$ (b) 0.15, (c) 0.28 and (d) 0.50 after drying at $< 5\%$ RH. The shiny particle images in (b – d) are indicative of a sucrose-rich particle shell. The bar in Figure S4a indicates a length of 40 μm .

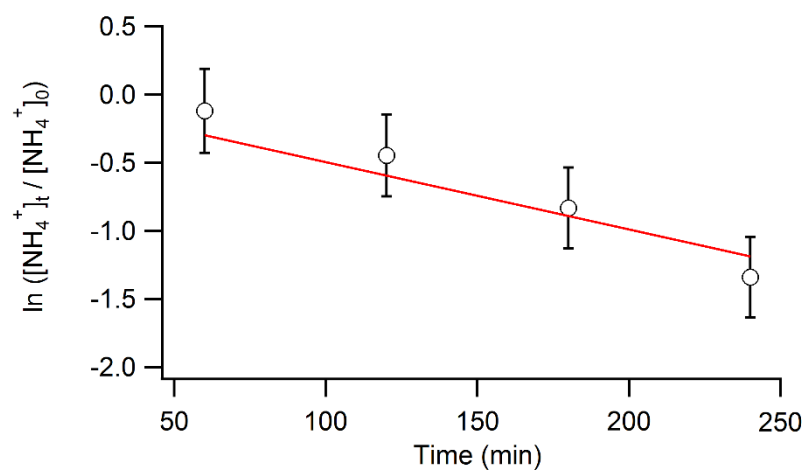


Figure S5. An example plot of $\ln ([\text{NH}_4^+]_t / [\text{NH}_4^+]_0) - t$, where DMA vapor reacted with the initial AS particle at 30% RH. The red line represents the linear regression function.

References

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