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Observations of Liquid-Liquid Phase Separation in Single Organic/Inorganic Aerosol Particles

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Aerosol particles containing both organic substances and inorganic salts are abundant in the atmosphere. The mixed organic/inorganic aerosol particles can undergo phase transitions (crystallization, dissolution, and liquid-liquid phase separation) when exposed to changing relative humidity (RH). The physical and chemical properties of the particles, such as the scattering and absorption of solar radiation, the rates of heterogeneous chemical reactions, and the kinetics of water uptake should be affected by the phase transitions. Therefore, the fundamental knowledge about the physical states of mixed organic/inorganic aerosol particles is indispensable for climate predictions. In previous experiments, micrometer-sized organic/inorganic particles deposited on hydrophobic substrates were usually investigated under an optical microscope. Since the solid substrates might act as seeds for the heterogeneous nucleation, the influence of the substrates on liquid-liquid phase separation is still controversial. Noncontact levitation of single micrometer-sized water droplets in air can be achieved by a laser trapping technique. Recently, we developed the trapping chamber equipped with a RH controller and demonstrated the reversible control of the equilibrium size of a single droplet levitated in air through a change in RH^[1]. Thus, the laser trapping technique is a powerful means to study on the phase transitions in single aerosol particles. In this study, we demonstrate a novel approach for in situ observation of liquid-liquid phase separation in mixed organic/inorganic aerosol particles levitated in air by means of a laser trapping technique.

Prior to the laser trapping experiments, liquid-liquid phase separations in mixed organic/inorganic aqueous droplets containing poly(ethylene glycol)-400 (PEG-400) and ammonium sulfate (AS) were observed on a hydrophobic glass substrate at different organic-to-inorganic ratio (OIR). Liquid-liquid phase separations were induced by decreasing RH. Depending on the OIR, two different mechanisms of liquid-liquid phase separation were successfully observed as shown in Fig. 1, namely, nucleation-and-growth (Fig. 1 (a)) and spinodal decomposition (Fig. 1 (b)) mechanisms.

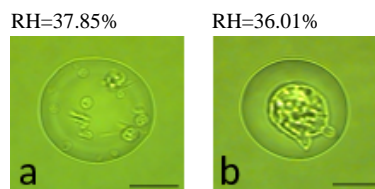


Fig. 1 Morphology of PEG-400/AS particles
(a) OIR = 8 : 1, (b) OIR = 1 : 1.
The length bar in each image corresponds to 20 μ m.

[1] S. Ishizaka, K. Yamauchi, and N. Kitamura, *Anal. Sci.*, **30**(11), 1075-1079 (2014).