

Direct Surface Tension and Viscosity Measurements of Aqueous-Organic Aerosol Droplets from Particle Coalescence Measurements

J. P. Reid¹, B. R. Bzdek¹, R. E. H. Miles¹, A. E. Haddrell¹, A. D. Hall¹

¹School of Chemistry, University of Bristol, Bristol, BS8 1TS, UK

Keywords: aerosol surface tension, coalescence, optical tweezers, viscosity.

Presenting author email: chjpr@bristol.ac.uk

The coalescence of airborne droplets is relevant to a wide range of processes including aerosol mixing state and lifetime, cloud dynamics, spray drying, and intranasal drug delivery. The time-dependent changes in composite droplet shape during the coalescence event are governed by composite droplet size, surface tension and viscosity. Aerosol surface tension impacts the ability to describe particle activation as cloud condensation nuclei via the Köhler equation. Aerosol viscosity impacts semivolatile partitioning from the aerosol phase, molecular diffusion in the bulk of the particle, and reaction kinetics. However, direct, quantitative measurements of both properties are challenging and thereby hinder accurate modelling of aerosol physical state and atmospheric impacts. In this contribution, we discuss precise measurement of the surface tension and viscosity of airborne picolitre droplets in both sub- and super-saturated solute states using holographic optical tweezers. Additionally, coalescence sampling of flowing aerosol into an optically trapped droplet will be discussed.

Aerosol is nebulized and captured into two optical traps formed from 532 nm light. The relative position of the two traps is varied so as to induce coalescence. Elastic and inelastic backscattered light are directed to a photodiode connected to an oscilloscope and to a Raman spectrometer, respectively. Blue light illuminates the droplets from above and is directed to a camera to image the droplets. Scattered Raman intensity is amplified at wavelengths commensurate with the whispering gallery modes and, through comparison with Mie scattering calculations, the radius and refractive index of the trapped droplet is accurately determined with precisions of ± 2 nm and 0.001, respectively. Coalescence excites shape oscillations which decay as the droplet relaxes to a sphere. In the limit of low viscosity, the time-dependent elastic backscattered light intensity takes the form of a damped oscillator and permits quantification of the shape relaxation timescale and oscillation frequency. These parameters permit precise, simultaneous determination of the droplet viscosity (from the relaxation timescale) and surface tension (from the oscillation frequency).

Figure 1 provides an example of measurements from picolitre droplets of aqueous glutaric acid solution. This illustrates that surface tension and viscosity can be measured with precisions of the order $1 \text{ mN}\cdot\text{m}^{-1}$ and $<1 \text{ mPa}\cdot\text{s}$, with measured values agree well with bulk measurements and parametrizations under sub-saturated conditions. Additionally, trapped droplets can be equilibrated to a relative humidity below their

deliquescence point but above their efflorescence humidity, thereby enabling the determination of the surface tension and viscosity of droplets containing supersaturated solute states.

The surface tensions and viscosities of subsaturated and supersaturated droplets of a wide range of atmospherically relevant model organic compounds including mixtures of both organic and inorganic components will be presented and compared with model predictions.

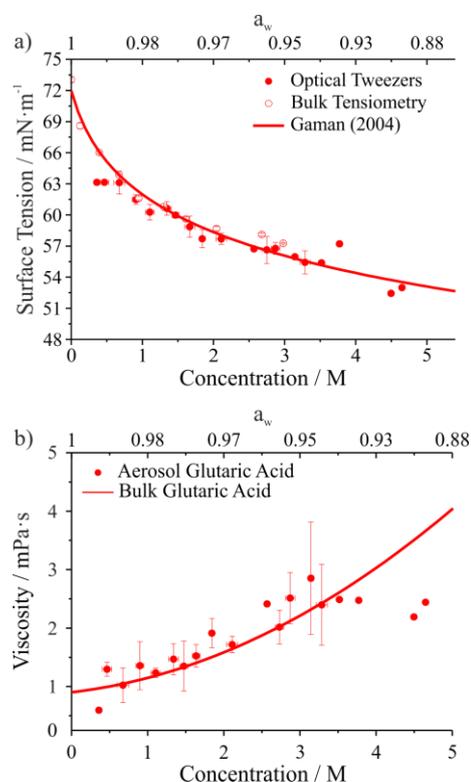


Figure 1. Simultaneous measurements of the surface tension and viscosity of picolitre glutaric acid droplets (Bzdek et al., 2016). Surface tension measurements are compared to a parametrization of bulk measurements (Gaman et al., 2004).

The authors acknowledge the Engineering and Physical Sciences Research Council, UK (grant EP/L010569/1).

Bzdek, B. R., Power, R. M., Simpson, S. H., Reid, J. P., and Royall, C. P. (2016) *Chem. Sci.* **7**, 274-285.

Gaman, A. I., Kulmala, M., Vehkamäki, H., Napari, I., Mircea, M., Facchini, M. C., and Laaksonen, A. (2004) *J. Chem. Phys.* **120**, 282–291.