Tracking Water Diffusion Fronts in a Highly Viscous Aerosol Particle

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Field measurements indicate that atmospheric secondary aerosol particles can be present in a highly viscous, glassy state [1]. In contrast to liquid state particles, the gas phase equilibration is kinetically limited and governed by condensed phase diffusion. In recent water diffusion experiments on highly viscous single aerosol particles levitated in an electrodynamic balance (EDB) we observed a characteristic shift behavior of the Mie whispering gallery modes (WGM) indicative of the changing radial structure of the particle, thus providing us with an experimental method to track the diffusion
process inside the particle.

When a highly viscous, homogeneous particle is exposed to an abrupt increase in relative humidity, the rapid gas phase diffusion and strong concentration dependence of the diffusion coefficient in the condensed phase lead to extremely steep water concentration gradients inside the particle, reminiscent of diffusion fronts. The resulting quasi step-like concentration profile motivates the introduction of a simple core-shell model describing the morphology of the non-equilibrium particle during humidification. The subsequent particle growth and reduction of the shell refractive index can be observed as red and blueshift behavior of the WGM, respectively. The shift pattern can be attributed to a core-shell radius ratio and particle radius derived from model calculations [2]. If supplemented with growth information obtained from the WGM redshift and thermodynamic equilibrium data, we can infer a comprehensive picture of the time evolution of the diffusion fronts in the framework of our core-shell model. The measured time dependent concentration profile is then compared with simulations solving the non-linear diffusion equation [3]

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