Abstract

Homogeneous nucleation of condensing water (H_2O) vapor was studied using a Laminar Flow Diffusion Chamber (LFDC), a Free Expansion Chamber (FEC) and a Supersonic Nozzle (SSN). Furthermore, in the SSN the subsequent freezing of the water nano-droplets was characterized. For temperatures T between 240 and 270 K, the nucleation rates J determined in the LFDC are in the range $10^3 < J$ / $cm^{-3}s^{-1} < 10^6$, which closes a gap in existing nucleation rate measurements. Critical cluster sizes n^* were determined by applying *Kashchiev's* nucleation theorem ($\ln J/ \ \ln S)_T = n^*$. Agreement with experimental literature values is good while the *Gibbs-Thomson* equation predicts larger n^*. In the FEC, the temperatures T_{on} and pressures p_{on} corresponding to the onset of nucleation were measured and compared to data generated in the Nucleation Pulse Chamber (NPC) and SSN. The FEC data lie between those from the NPC and SSN, and thus, the FEC nucleation rates are estimated as $J \sim 10^{13}$ $cm^{-3}s^{-1}$. In the SSN position resolved Pressure Trace Measurements (PTM), Small Angle X-ray Scattering (SAXS) and Fourier Transform Infrared (FTIR) spectroscopy experiments were conducted. The PTMs yield the flow properties including the temperature and supersaturation profiles, as well as the characteristic nucleation time. The SAXS experiments determine the particle number densities *N,* $1.5 \cdot 10^{12} < N$ / $cm^{-3} < 8 \cdot 10^{12}$, and the average droplet radii $<r>$, $3.2 < <r>$ / nm <5.8. From the decrease in N with flow time, the coagulation of nanometer sized water droplets was investigated for the first time. Measured coagulation rate coefficients are up to 4.46 times higher than theoretical predictions. This enhancement is assumed to stem from the increased importance of *van der Waals* forces as particle size decreases, and is consistent with literature values reported for other materials. Combining the results from PTM and SAXS yields nucleation rates in the SSN of $7 \cdot 10^{17} < J$ / $cm^{-3}s^{-1} < 11 \cdot 10^{17}$ for temperatures $198 < T$ / K < 220, and these rates are 4 to 5 times higher than past measurements. Applying FTIR spectroscopy yields the first strikingly clear evidence of freezing of highly sub-cooled H_2O nano-droplets in the nozzle. The spectra corresponding to a partially frozen aerosol were fit to linear combinations of liquid and ice spectra in order to determine the fraction of frozen droplets. The time dependence of the fraction of frozen droplets yields the volume based nucleation rates for the freezing transition, $6.61 \cdot 10^{22} < J_{ice}$ / $cm^{-3}s^{-1} < 3.64 \cdot 10^{23}$, and these are in good agreement with literature values.