73a The Properties of Small Water Clusters from Isothermal Nucleation Rate Measurements

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We have made direct measurements of the stationary, homogeneous nucleation rates, $J=N/\Delta t$, in supersonic Laval nozzles. The number densities, N, of droplets formed are measured using small angle neutron scattering (SANS) experiments and the time intervals during which nucleation occurs, $\Delta t < 10$ μs, are derived from static pressure measurements along the axis of the nozzle. By using nozzles with different expansion rates, we obtain the first isothermal nucleation rate measurements as a function of supersaturation for these devices with a relatively small error margin in J of +/-50%. At temperatures T of 210, 220, and 230 K, the maximum nucleation rates for D_2O range between $4 \cdot 10^{16} < J / cm^{-3} s^{-1} < 3 \cdot 10^{17}$ for supersaturations S ranging from 46 to 143. Applying the first and second nucleation theorems to isothermal nucleation rate data directly yields estimates for the number of molecules in the critical cluster n* and the excess internal energy $E_x(n^*)$, respectively. The agreement between these values and the classical values predicted assuming the critical cluster is a compact spherical object are really quite good even though under our conditions n* is less than 10 and the water is highly supercooled.