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Simulating the nucleation of water/ethanol and water/n-nonane mixtures: Mutual enhancement and two-pathway nucleation

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Abstract

A combination of the aggregation-volume-bias and configurational-bias Monte Carlo algorithms and the umbrella sampling technique was applied to investigate two different binary vapor–liquid nucleation systems: water/ethanol and water/n-nonane. The simulations are able to reproduce the different non-ideal nucleation behaviors observed experimentally for these two systems, i.e., the mutual enhancement of nucleation rates for water/ethanol mixtures and the two-pathway nucleation for water/n-nonane mixtures. Structural analysis provides microscopic explanations for the observed nucleation behavior. In particular, the simulations show a large and size-dependent surface enrichment of ethanol in the water/ethanol droplets, which confirms the previous experimental interpretation for this system. The immiscibility observed even for small water/n-nonane clusters causes the two-pathway nucleation mechanism.