Crystallization, nucleation and droplet growth for the combined gas treatment and liquefaction of natural gas

Abstract

Molecular dynamics (MD) simulations were carried out to study the vapour to liquid homogeneous transition. Understanding and controlling the phase behaviour of gas mixtures is of great importance in the field of natural gas industry. Where the homogeneous nucleation phenomenon constitutes the fundamental process of phase transitions. The key property of nucleation is the nucleation rate, defined as the formation rate of stable clusters (droplets) per unit volume. Atomistic simulations, such as MD, prove to be promising tools in gaining complete description of the kinetics of non-equilibrium dynamics. Previously, Yasuoka *et al.* [1] evaluated water nucleation rates from MD simulations using the threshold method. The applied methodology enables us to study various gas mixture systems.

In this work we focus on the system of water as condensate and argon, methane, and helium as comparable carrier gases. We first assess the effect of the water molecular model, by adopting the TIP4P and TIP4P/2005 models [2]. From this nucleation study of water in argon at 350 K we find the nucleation rate of the TIP4P/2005 model twice as large as the rate of the TIP4P model. Also the stability of all identifiable clusters in our simulations is strongly dependent on the random impingement with the neighbouring molecules. Even though a cluster is in a dominantly growing process, we notice a significant change in its composition. The improved TIP4P/2005 model was further used in the simulations with helium and methane [3]. From the comparison between the carrier gases used, methane is found to be a slightly better thermalizer gas for water than helium and argon. Therefore, MD can be further used to accurately characterize condensation in various gas mixtures at the chosen system conditions.

[1] K. Yasuoka and M. Matsumoto, *J. Chem. Phys.* **109**, 8463, 1998.

[2] L. R. Dumitrescu, D. M. J. Smeulders, J. A. M. Dam, and S. V. Gaastra-Nedea, *J. Chem. Phys.* **146**, 084309, 2017.

[3] L. R. Dumitrescu, H. Huinink, D. M. J. Smeulders, J. A. M. Dam, and S. V. Gaastra-Nedea, (unpublished).