Contact-free Hydrate Formation on Water Droplets Acoustically-Levitated in High-Pressure Natural Gas

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Hydrate nucleation at the gas-water interface is of direct relevance to the field of flow assurance in the energy industry. However, hydrate nucleation can be difficult to characterize due to its stochastic nature and potential apparatus dependencies. Conventionally, studies have been performed using stirred fluids in high-pressure vessels wherein solid-liquid interfaces generate potential hydrate nucleation sites that are non-trivial to characterize. To overcome such constraints, we have used an acoustic levitator to enable the study of contact-less hydrate formation on a levitated water droplet. During the experiment, water droplets with well-defined geometries were acoustically levitated within a high-pressure natural gas atmosphere. Hydrate formation on the gas-liquid interface of the µL-scale droplets was then directly imaged and used to obtain induction times. Cumulative probability distributions describing hydrate formation probability as a function of time were subsequently constructed at subcoolings of approximately (12, 13.2 and 14.5) K and fitted to a model from classical nucleation theory, enabling extraction of nucleation rates. To compare these values with rates observed in much larger systems, the extracted nucleation rates were normalized to the gas-water interfacial area of the droplets. Nucleation rates measured at lower subcoolings in a stirred reactor experiment (HPS-ALTA; high pressure, stirred, automated lag time apparatus) were similarly normalized. An extrapolated trendline based on the subcooling-dependent, area-normalized nucleation rates from the levitator aligned well with those from the HPS-ALTA. This implies that the gas-water interfacial area is the characteristic area that determines the experimentally observed nucleation rate in well mixed systems (in the levitator, mixing is generated via the acoustic field).