

Microfluidic Measurements of CO₂ Hydrates: Equilibrium Water Content, Film Thickening, and Raman Spectroscopy

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Carbon dioxide hydrates may form during carbon dioxide capture and storage (CCS) processes. These solid, ice-like compounds may form in pipelines during the transportation of carbon dioxide, especially in offshore cases where temperatures can be low. In order to prevent carbon dioxide hydrates from forming, water can be removed from the stream before transportation. The degree of drying required is informed by the solubility limit of water in carbon dioxide at equilibrium with the hydrate, which is dependent on temperature and pressure conditions. Using a high pressure micro-reactor as an equilibrium cell, the water solubility in carbon dioxide was measured at different conditions. These smaller scale equilibrium experiments allow for a much higher experimental throughput than can be achieved in larger systems. The optical transparency of the chip allows in situ Raman spectroscopy to be used, which confirmed the absence of metastable phases during water solubility measurements. Additionally, the thickening of carbon dioxide hydrate films crystallizing in the micro-reactor has been investigated to better understand hydrate formation in pipelines and in other CCS processes. The influence of subcooling, pressure, and carbon dioxide flow rate was explored. In situ Raman spectroscopy was also used in these thickening studies to better understand hydrate annealing phenomena.

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