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Pore-scale morphologies of CO2 hydrate formation in microfluidics with in-situ Raman spectroscopy for CO2 sequestration

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CO2 hydrates are cage-like solid compounds consisting of CO2 gas molecules and water molecules. The properties of CO2 hydrates, such as high gas storage capacity and moderate formation conditions of pressure-temperature, are desirable for long-term and safe CO2 storage in geological setting. The purpose of work is to probe the feasibility of CO2 storage in hydrate in marine subsurface. Compared to natural gas hydrates, it is difficult to investigate morphological CO2 hydrate dynamics in microfluidics, as the morphologies of CO2 in liquid and water phases are similar. In this work, two scenarios were investigated regarding different CO2 states controlled by different pressures above or below CO2 liquid liquefaction pressure (PL-CO2), by means of pore-scale microscopic observation coupled with in-situ Raman spectroscopy in microfluidic chip. In one scenario, the system pressures were kept lower than PL-CO2 to form CO2 hydrate. The system was tested by both microscope and Raman spectroscope to distinguish CO2 in gas, water and hydrate. A pressure difference was observed in the system indicating hydrate blocked pore channels and prevented unwanted CO2 flow. In another scenario, the system pressures were increased over PL-CO2, which was the typical reservoir pressure, to observe CO2 hydrate formation in the system containing liquid CO2. The differences between both morphological patterns and Raman peak shift were observed, verifying CO2 could be stored in states of both hydrate and liquid. The quantitate calculation of CO2 storage capacity showed about 80 volumes of CO2 was retained in the system having only CO2 hydrates, compared with about 150 volumes of CO2 stored in the system having both CO2 hydrates and liquid CO2. The morphological patterns, Raman spectra and calculation of storage capacity indicates that CO2 hydrate storage could serve as a secondary storage option for geological CO2 storage in marine subsurface. The results of this work are beneficial to understand marine CO2 hydrate storage in confined space of porous media, and the enhanced CO2 hydrate storage capacity can be explored by coupling with hydrate promotors and thus achieve more efficient CO2 storage in marine subsurface conditions.

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Primary author: OUYANG, Qian

Co-authors: PANDEY, Jyoti Shanker (Technical University of Denmark); Prof. VON SOLMS, Nicolas (Technical University of Denmark)

Presenter: OUYANG, Qian

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