Clathrate Hydrate Single Crystals: Studies of Growth Kinetics and Thermodynamics

Nur Aminatulmimi Ismail^s and Ahmad AA Majid

Center for Hydrate Research, Chemical & Biological Engineering Department, Colorado School of Mines, Golden, CO, U.S.A. Faculty of Chemical & Natural Resources Engineering, Universiti Malaysia Pahang, Pahamg, Malaysia

Luis E. Zerpa

Department of Petroleum Engineering, Colorado School of Mines, Golden, CO, U.S.A.

Reuben T. Collins Department of Physics, Colorado School of Mines, Golden, CO, U.S.A.

Carolyn A. Koh^C Center for Hydrate Research, Chemical & Biological Engineering Department, Colorado School of Mines, Golden, CO, U.S.A. ckoh@mines.edu

Gas hydrates (also known as clathrate hydrates) are non-stoichiometric crystalline solids in which small organic guest molecules (e.g., methane, ethane, tetrahydrofuran) are encaged in a host lattice composed of hydrogen-bonded water molecules. There are many important energy applications for gas hydrates (i.e. gas transportation, energy recovery). Additionally, in the petroleum industry, gas hydrate may form blockages in subsea oil/gas pipelines during production, transportation, and processing of reservoir fluids, which are considered major economic and safety concerns. For the advanced gas hydrate management strategy of gas hydrate blockages in the flowline, as well as the application of gas hydrate technology, it is necessary to understand the kinetics and structural properties of gas hydrate crystals during the growth period. Thus, in this work, single crystals of structure II (sII) gas hydrate and other gas hydrate structures were grown using several approaches, including a seed crystal method, and the use of a cold wire for stimulating nucleation sites. The crystal organization and faceting, and growth rates were investigated under different growth conditions of driving force (i.e., subcooling of 0.4 - 2 °C) and composition. The growth rates obtained for fixed subcooling were found to fall between $0.6 \ \mu m/s$ to $2.5 \ \mu m/s$. In addition, the kinetics together with the thermodynamics of the growth process were studied. The results from this study will provide further insight into the detailed kinetics and thermodynamics of the gas hydrate formation.

Acknowledgment: Funding and support from the CSM Hydrate Center Consortium, William K. Coors Distinguished Chair Fund, and Renewable Energy Materials Research Science and Engineering Center (REMRSEC) are acknowledged.