Journée scientifique du CODEGEPRA 2016 17 Novembre 2016 Grenoble INP-Pagora / LGP2- Campus de Saint Martin d'Hères

An experimental study of gas hydrate formation of CH₄-C₃H₈ binary mixture under the non-equilibrium conditions

MAGHSOODLOO Saheb*, LE-QUANG Du, BOUILLOT Baptiste, HERRI Jean-Michel

a Gas Hydrate Dynamics Centre, Ecole Nationale Superieure des Mines de Saint-Etienne, Centre SPINLaboratoire Georges Friedel (UMR 5703)

158, Cours Fauriel, 42023 Saint-Etienne, France

Résumé

Gas hydrates are a crucial issue in many fields, from oil & gas industry, to carbon capture and storage, air conditioning, or even planetary science. They also have an enormous potential as an energy resource because of the tremendous quantity of methane trapped within them. To understand better the gas hydrates phenomena, understanding of the thermodynamics and the crystallization mechanisms of clathrate hydrates are required. The determination of the hydrate composition and volume is also a technical challenge and we recall in the paper the method used by our laboratory. In this work, several experiments were performed to obtain precious experimental data and the gas compositions in hydrate phase. GasHyDyn group has recently found out that the composition of gas hydrate is sensitive to the crystallization conditions. Hence in this work, it was tried to understand better the relation between the composition of hydrate during the crystallization and final state and crystallization rates. Experiments were performed in a batch reactor and there were an online gas chromatography connected to the experimental set-up and an off-line ion-chromatography to measure the gas compositions and liquid compositions, respectively. Based on these data and mass balance, the gas compositions in hydrate phase and hydrate volume can be calculated. During the last decade, our GasHyDyn team has also measured equilibrium data for many gas mixtures, and this document adds new data involving propane. In the present work, the experimental results are discussed after formation following two different experimental protocols: at quick and slow rates of crystallizations. It was observed that, from the same initial conditions, the final pressure and water conversion for two different procedures are slightly different (Figure 1).

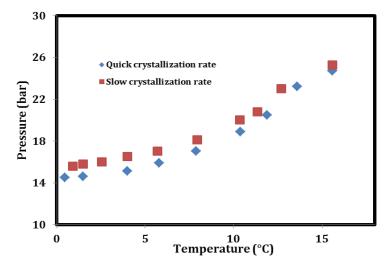


Figure 1. PT diagram for two different experimental procedures with the same initial conditions: quick and slow crystallization rates

-

^{*} Corresponding author : saheb.m@emse.fr

Journée scientifique du CODEGEPRA 2016 17 Novembre 2016 Grenoble INP-Pagora / LGP2 - Campus de Saint Martin d'Hères

Références

- Bouillot, B., Herri, J.-M., 2015. Framework for clathrate hydrate flash calculations and implications on the crystal structure and final equilibrium of mixed hydrates. Fluid Phase Equilibria. 413, 184-195.
- Herri, J.-M., Bouchemoua, A., Kwaterski, M., Fezoua, A., Ouabbas, Y., Cameirao, A., 2011. Gas hydrate equilibria for CO_2 – N_2 and CO_2 – CH_4 gas mixtures-Experimental studies and thermodynamic modelling. Fluid Phase Equilibria 301, 171–190.
- Le Quang, D., Le Quang, D., Bouillot, B., Herri, J.-M., Glenat, P., Duchet-Suchaux, P., 2015. Experimental procedure and results to measure the composition of gas hydrate, during crystallization and at equilibrium, from N_2 -CO₂-CH₄-C₂H₆-C₃H₈-C₄H₁₀ gas mixtures. Fluid Phase Equilibria. 413, 10-21.
- Le Quang, D., Herri, J-M., Ouabbas, Y. A method to calculate the composition of clathrate hydrate of CO₂-CH₄, at equilibrium and during crystallization. 2013. Energies nouvelles et procédées (104), pp.No 99.
- Nixdorf, J., Oellrich, L.R., 1997. Experimental determination of hydrate equilibrium conditions for pure gases, binary and ternary mixtures and natural gases. Fluid Phase Equilibria 139, 325–333.
- Sloan, E.D., Koh, J., Carolyn, 2007. Clathrate Hydrates of Natural Gases, 3rd ed. Taylor & Francis.