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# **A new approach of studying mixed gas hydrates involving propane at non-equilibrium conditions and final state: An experimental study and modeling**

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## **Abstract**

Metastable clathrate hydrates are a promising energy source in the shallow geosphere and present challenges in flow assurance, energy storage, and carbon capture sequestration. While they have been widely studied, little pertinent data is available for common propane hydrates concerning hydrate phase composition, nor its volume or the amount of converted water. This was the initial motivation for our work. Therefore, with a novel technique, propane hydrate composition and volume were measured dynamically at non-equilibrium conditions over time and at the final states for slow and quick rates of crystallization. Surprisingly, equilibrium pressure, hydrate volume and composition are different according to crystallization rate. The hydrate volume and water conversion in the quick crystallization process were larger. Moreover, at a slow crystallization rate, in a hydrocarbon mixture, enclathration of propane is more considerable and the hydrate crystals appear to be more homogeneous. Furthermore, the hydrate crystallization of a gas mixture is closer to the thermodynamic equilibrium at slow crystallization rates where the impact of kinetics is slight. A new compilation of propane Kihara parameters was presented. Unlike methane, ethane and carbon dioxide, for propane we strongly recommend two Kihara parameters, one for pure and the other for mixtures of propane. A thermodynamic model based on classical van der Waals and Platteuw model was also used to investigate the effects of kinetics. The simulation results have a satisfactory accordance with the experimental data from literature to predict the hydrate equilibrium pressure. The consequence of this research could have a substantial impact on design calculations in which the assumption of thermodynamic equilibrium are done. For instance, at present there would be excess hydrates volume estimations for pipe-lines, equilibrium conditions in energy storage and transportation or carbon capture sequestration and thus increase expenses or loss of productivity where propane is concerned.

Keywords: Clathrate hydrates, crystallization, thermodynamics, non-equilibrium, modeling, flow assurance

## **1. Introduction**

Clathrate hydrates, are solid compounds composed of cages of water with guest “gas molecules” trapped inside (Sloan and Koh, 2007). Based on the number and type of cavities and the gas

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