



Original Research

Formation Methods and Properties of Gaseous Hydrates

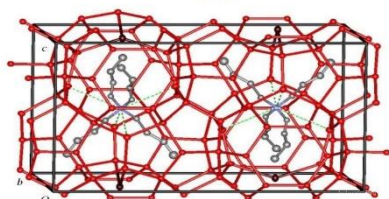
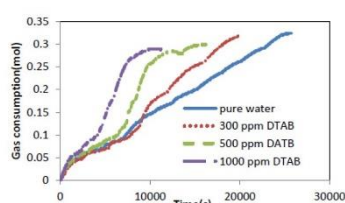
Rozita Kaviani*

Department of Chemical Engineering, Shiraz Branch, Islamic Azad University, Shiraz, Iran

GRAPHICAL ABSTRACT

ABSTRACT

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Underground gas reserves in the form of hydrates are a huge source of energy that researchers are looking for ways to extract. The amount of hydrate gas reserves is estimated by more than 1016 cubic meters, which is more than the discovered sources of other fossil fuels, if only 15% of this gas is extracted. But due to problems such as the slow rate of hydrate formation and the high pressure of its formation, this idea remained in the laboratory. After the discovery of hydrate self-preservation, this idea was raised more seriously and a lot of research was done on the use of hydrate for gas storage and transmission. High gas pressure is the most important negative factor in mass production of hydrates for gas storage and transport. The results showed that using these materials, methane hydrate is formed in much more suitable conditions than temperature and pressure. While the equilibrium pressure of methane hydrate formation at 293 ° K is about 34 MPa, with the addition of tetra-hydro-furan at a concentration of 2.5 mol%, this pressure is reduced to about 2.49 MPa, which shows a decrease of 92.6%. Also, at a pressure of 5 MPa, the equilibrium temperature of methane hydrate is about 278 degrees Kelvin, while at the same pressure, the fuzzy equilibrium temperature of a 6 mol solution of tetra-hydro-furan is about 306 degrees Kelvin.

* Corresponding author: Rozita Kaviani

E-mail: rozita.kaviani@gmail.com

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Introduction

Two classifications of the chemical nature of the guest molecule are proposed. The first one was proposed by Von Stackelberg et al., which was a combination of both size and chemical nature as previously stated [1-5]. The second classification was founded by Jeffrey and McMullan in 1967, in which molecules fall into one of four groups:

- 1) Hydrophobic compounds
- 2) Water-soluble acid gases
- 3) Water-soluble polar compounds
- 4) Water-soluble salts of the second or third type of alkyl ammonium [6-9].

In 1984, Jeffrey stated the classification based on the nature of the chemical in such a way that the guest molecule should not contain a strong hydrogen bond. Molecules of natural gas compounds do not contain hydrogen bonds and therefore their chemical nature is not a determining parameter [10-15].

Restrictions on the movement of guest molecules

Davidson (1971) found that the most important inhibitory interaction against rotation between guest molecules (in adjacent cages) was the bipolar-dipole interaction [16-20]. But these effects are of little importance. The polar and non-polar molecules of the guest, such as ethylene oxide, tetrahydrofuran, and acetone in a cage, feel only a slight impediment to free rotation. Free rotation may be due to the fact that the sum of the water cage dipoles effectively disappears near the center of each

cage. Guest molecules all between the size of argon 3.8 Å and cyclic and butanone 4.6 Å can form hydrates in structures I and II if there is no limit to their chemical nature [21-25].

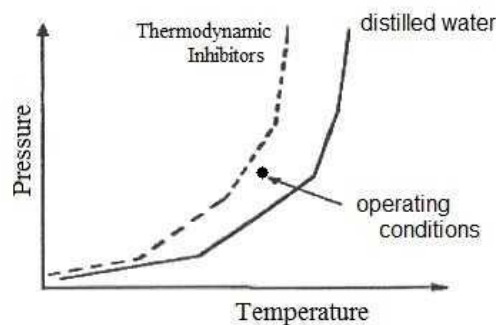


Figure 1. The effect of increasing the thermodynamic inhibitor on the hydrate formation curve

The size of the guest molecule

To specify the size that each compartment provides for a guest, Davidson suggested that the van der Waals radius of the water molecule A be reduced by 1.45 from the mean radius of the chamber. These data also come for two unusual molecules, cyclopropane and triethydm-ethylene, which can form simple hydrates in both constituents [26-30].

The ratios written with the symbol f are those occupied by simple hydrate constituents. The values in the table show that the lower limit for the formation of simple hydrates is about 0.77. Below this ratio, the molecular gravitational forces cannot stabilize the chamber, and the upper limit is about 1, above which the guest molecule cannot fit in the chamber without deformation [31-35].

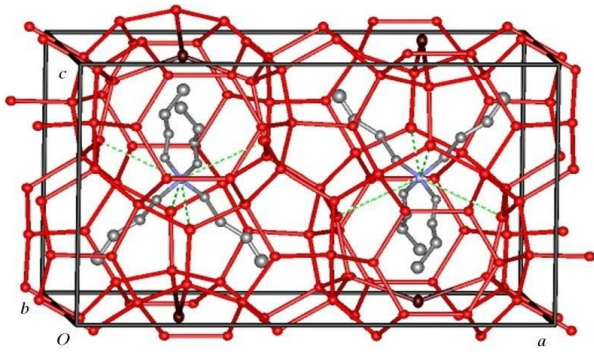


Figure 2. A single cell is formed from the tetra butyl ammonium hydrate

Components that are able to enter the chamber 5^{12} (of both structures) can also enter the large chambers of that structure [36-40] for the natural gas have constituents of the simple hydrate and occupy with a ratio of 0.971 and 1.005. Normal butane cannot form a simple hydrate because it is attributed to a diameter much higher than 1. It has recently been shown that nitrogen in chamber 5^{12} stabilizes type II structures [41-45]. Methane and hydrogen sulfide molecules can occupy chambers 5^{12} in structure I in ratios between 0.886 and 0.931. Methane, methyl silico-hexane, isopentane and 2 and 3 dimethyl butane were combined. Based on the chamber sizes of structure II and H hydrate and the size of the molecule, it is determined that under certain temperature and pressure conditions, structure II hydrate will be formed for benzene and cyclohexane in the presence of an auxiliary gas [46-50]. The structure of H hydrate consists of methyl cyclopentane, methylcyclohexane, 2 and 3 dimethylbutane and isopentane in the presence of an auxiliary gas. Cyclopentane can be formed in the structure of hydrate II in the presence or

absence of an auxiliary gas. The auxiliary gas must fill small chambers and stabilize the structure of the hydrate [51-55].

The shape of the guest molecule

The shape of the guest molecule plays a small role in the structure of the hydrate and its properties. Numerous studies have been performed by Davidson et al [56-59] to estimate molecular reorientation barriers for all components of natural gas hydrates except carbon dioxide. If there are real obstacles to circulation, it can affect the properties of the hydrate. For the constituents of structure, I, basically no barrier is found for methane and hydrogen sulfide, while the barrier for ethane is 1.2 kcal / mol. Medium barriers for structure II constituents are 0.6, 1.2, and 1.4 Kcal/_{moel} for propane, isobutane, and normal butane (double hydrate by hydrogen sulfide, respectively). At most temperatures preferred for natural gas processes above k100, the guest molecules have little limitation for reorientation.

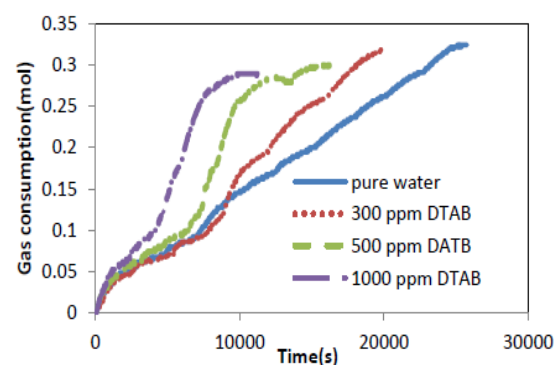


Figure 3. Effect of DTAB on the rate of ethane hydrate formation at 227 K

Features of gas hydrate

Hydrogen bonding: In 1920, hydrogen bonding was proposed using a simplified electrostatic model of a water molecule. The positive pole of a molecule is absorbed by two negative poles of an adjacent molecule. According to this mechanism, each water molecule is attacked by four others. Each proton of the molecule is absorbed by two negative poles of the adjacent molecule, which creates a bond. Also, the two negative poles of the primary molecule are absorbed by the two positive poles of the other two molecules. The four surrounding molecules are arranged in a quadrilateral around the central molecule. In ice, only 34% of the volume is occupied by water molecules, while 37% of the volume in liquid water is occupied by water molecules, which causes ice to float on water. The energy required to break a hydrogen bond ($5 \frac{\text{kcal}}{\text{mol}}$) is much greater than that required

to break a van der Waals bond ($0/3 \frac{\text{kcal}}{\text{mol}}$) as the attraction between two non-polar molecules in a fluid is running. On the other hand, a hydrogen bond is the size of a covalent bond.

($102 \frac{\text{kcal}}{\text{mol}}$) That is, what exists between hydrogen is not oxygen in a separate molecule of water. Based on the strength of these bonds, when hydrates are formed or decomposed, only hydrogen bonds between adjacent molecules are considered. In fact, van der Waals forces are present but can be neglected in contrast to hydrogen bonds.

Formation of methane hydrate under mild conditions using a facilitator

The natural gas hydrate cycle (NGH) consists of three stages of production, transfer and conversion to gas. The first stage, hydrate production, accounts for 61% of the total cycle cost, the transfer stage for 31%, and the gasification stage for 8%. One of the important reasons for the high cost of gas hydrate production is the high pressure of hydrate formation. In addition to creating safety problems, high hydrate formation pressure also increases the cost of designing and manufacturing devices and the cost of implementation. Therefore, reducing the hydrate production pressure will have a significant effect on production costs and process facilitation. In recent years, materials have been used as thermodynamic facilitators of gas hydrate formation, the most important of which is tetrahydrofuran (THF) [60-62].

Construction of gas hydrate decomposition reactor

This reactor was designed and produced by a professor of the University of Oil and Gas to decompose hydrates at high pressure. This reactor is currently operating in the university laboratory. According to him, the mentioned reactor is designed and built with the aim of testing and measuring the equilibrium conditions of gas hydrate decomposition. This device has the ability to measure the conditions of hydrate decomposition and tolerance of 4500 PSI and has reached the construction stage [63-65].

Conclusion

The results show that in summer, despite the decrease in gas temperature after the regulator, it will not be hydrated, but in winter operating conditions, it is possible to form a pressure breaker before and after the regulator at pressures above 252 and 174 psig for before and after the regulator, respectively. In certain circumstances, the use of NGH in natural gas transmission is superior to LNG, but at distances of less than 1000 km, the use of NGH over LNG is not cost-effective at all. The transmission of natural gas over long distances (at least more than 1000 km) and on a large scale can be a desirable idea that must be confirmed by economic considerations. The results showed that these additives are suitable thermodynamic facilitators for methane hydrate and in their presence, the fuzzy equilibrium diagram is significantly shifted to the right.

In this project, the natural thermodynamic path method in the kinetics of chemical reactions was used to predict the kinetics of methane hydrate formation in the constant volume process. The coefficients of this model, tK and Ar , were obtained for several different temperatures. The results showed that this method has the necessary efficiency to predict laboratory data. In experiments performed at high initial pressures, the final system pressure was significantly different from the equilibrium pressure and the system pressure at the end of the test was the equilibrium pressure side which did not advance. This may be the reason for the large difference between the model results and the experimental data. This indicates that when hydrate is formed within the system, there is an

obstacle to the system reaching equilibrium. This factor can be the mass transfer between the gas and water phases. Because at high initial pressures, the rate of hydrate formation is also higher as a result of the volume of the amount. One of the important points for the models used to predict the kinetics of hydrate formation is whether the model has the ability to predict the experimental data obtained by researchers in other laboratories with the same coefficients or not. In this project, it was shown that the tK coefficient can be obtained from the experimental data of other researchers, and thus the results predicted using this model are more consistent with other experimental results.

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