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Study of sodium halide aqueous solutions treatment for ethane hydrate formation kinetics



Ali Dehghanpoor *, Farshad Varaminian

School of Chemical, Gas and Petroleum Engineering, Semnan University, Semnan, Iran

A R T I C L E I N F O

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ABSTRACT

The aim of this work is to present the experimental investigation of ethane hydrate formation kinetics in the presence of sodium halide salt solutions containing NaF, NaCl, NaBr, and Nal. To achieve this aim, the variations of pressure versus time have been measured during hydrate formation process. The temperature, volume of reactor and stirring velocity are 277 K, 500 ml, and 840 rpm, respectively. The influence of anion type salts and their concentrations on induction time and hydrate formation rate have been studied. The result prove that sodium halides (NaF, NaCl, and NaI) at a concentration of about 0.6 wt% and sodium bromide (NaBr) at concentration ranged 0.3–1.5 wt% have minimum promotion effect on the ethane hydrate formation rate. Also, the results of present study show a significant decrease in hydrate formation rate at a concentration higher than 1.5 wt% for sodium halides.

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1. Introduction

Gas hydrates are crystalline non-stoichiometric compounds in which water (host) molecules are brought into contact with small molecules such as methane, ethane, propane, carbon dioxide and some refrigerants at elevated pressures and/or low temperatures. Because of hydrogen bonding, the molecules of water forms a network of cavities in which cavities of water surround the gas (guest) molecules. This structure is thermodynamically stabilized due to van der Waals forces between the guest molecules and the water cavities. On the basis of the size of cavities and molecular size of the guest molecules, three main crystallographic structures of gas hydrates exist, namely structure I (sl), structure II (slI) and structure H (sH) [1,2].

In some chemical and oil industry applications, gas hydrate is a nuisance. In production lines, plugs due to gas hydrate formation are problematic in the oil and gas industry during drilling, and in work-over operations. In some of the gas and oil industry applications, gas moves toward deep-water exploration and production in which the pressure and temperature conditions are appropriate for hydrate formation. Because of these problems, hydrate inhibition in oil and gas industry is of importance. Increasing the temperature or heating (containing insulation, bundles, electric or hot water heating), operating at low pressures and shifting the thermodynamic conditions of hydrate formation by adding thermodynamic inhibitors are several ways of avoiding the formation of hydrate plugs. These methods are often very expensive to be applied. The high concentrations of thermodynamic inhibitors

* Corresponding author. *E-mail address:* ali_dehghanpour@semnan.ac.ir (A. Dehghanpoor). are required in aqueous solution so that the ways with lower cost should be utilized; therefore, one way to overcome this problem is to use the thermodynamic inhibitors at very low concentration and investigate the influences of them on the kinetics of hydrate formation. In previous studies, the thermodynamic inhibitors such as salts were used to increase hydrate formation pressure. On the other hand, a few investigations have been conducted on effects of salts on hydrate formations kinetics.

In recent years, a few studies have been done to determine the hydrate formation rate in the presence of thermodynamic inhibitors. Farhang et al. [3] investigated the influences of sodium halide salts on kinetics of carbon dioxide hydrate. They also determined the impacts of anion type and concentration on the maximum uptake of the carbon dioxide, conversion, storage capacity, induction time and growth rate of hydrate crystals. They found that halide ions had a significant influence on CO_2 hydrate formation rate. Yang et al. [4] studied the effects of halogen ions on thermodynamics and induction time of methane hydrate. They concluded that halogen ions decrease the equilibrium temperature and have influences on induction time of methane hydrate.

Due to lack of enough investigations on effects of sodium halide salts on the kinetics of hydrate formation, the effects of sodium halide salts containing NaF, NaCl, NaBr and NaI on ethane hydrate formation rate have been investigated experimentally at very low concentrations. The operational temperature, volume of reactor and stirring velocity are 4 °C, 500 ml and 840 rpm, respectively. To achieve this aim, seven concentrations of sodium halide salts ranged 0.15–10 wt% are applied for hydrate formation kinetic experiments. Furthermore, the induction times of ethane hydrate have been measured in the presence of sodium halide salts at different concentrations. Table 1

Test materials used for experiments.

	1		
Component	Chemical formula	Purity	Supplier
Ethane Sodium fluoride Sodium chloride Sodium bromide Sodium iodide Water	C ₂ H ₆ NaF NaCl NaBr NaI H ₂ O	99.95% 99% >99.5% >99.99% 99.5% Deionized, distilled	Technical Gas Services Merck,Germany Dr.m, Iran Merck,Germany Merck,Germany Cheshme Zolal Sabalan, Iran

2. Experimental

2.1. The applied materials

An analytical grade ethane with the chemical formula of C_2H_6 is used as hydrate formers with a purity of 99.95% that is supplied by Technical Gas Services. It was selected to form a hydrate with double distilled water. The information of the applied materials and salts has been shown in Table 1.

2.2. Apparatus

The schematic of experimental hydrate forming set-up has been shown in Fig. 1. It can be seen that the ethane (C_2H_6) hydrate formation experiments are done in the cylindrical, and high-pressure stainless steel cell with a total capacity of 500 cm³ which can operate pressures up to 10 MPa and temperatures ranging from 253.15 K to 373.15 K. The hydrate formation cell is also equipped with a three-blade mixer. The coolant of the system contains (water + ethylene glycol) mixture. The coolant was circulated through this jacketed cell, and the cell temperature was controlled with the flow of coolant through the jacket. The cell is also insulated. It also contains a coolant bath with controllable circulator which is used to circulate the coolant. The temperature is measured by using a PT100 thermometer (Pro-Temp Controls, Santa Ana, California, United States) with \pm 0.1 K accuracy. A BD-Sensors-Str.1 pressure transmitter with \pm 0.1 bar accuracy is utilized to measure the pressure of the cell. To measure the pressures and temperatures of the cell, the cell contains a data acquisition system which is connected to a personal computer.

2.3. Experimental procedure

Before conducting any experimental measurement, the reactor was washed and rinsed with the water which has been distilled for four times. Then the cell was evacuated with a vacuum pump. Subsequently, the cell was charged with 150 cm³ of pure water or aqueous solution of sodium halide salts. All experiments were conducted at the 277.15 K. First, the cell was pressurized to approximately 0.36 MPa below the equilibrium pressure of ethane hydrate formation (the equilibrium pressure of ethane hydrate is 7.6 bar at 277.15 K). Then the coolant bath was turned on and the reactor was allowed to reach the desired temperature for the formation of the hydrate. After the operating temperature was stable, gas was slowly charged into the cell, and the system was allowed to reach a supersaturated condition. After the specified temperature was maintained, the reactor was pressurized to the specified initial pressure with ethane. Then the mixer was turned on. All experiments are also are conducted at a stirring velocity of 840 rpm. Subsequently, temperature and pressure data were collected during the each experiment. Each test was carried out until the system reached an equilibrium condition.



Fig. 1. Experimental setup and hydrate formation apparatus.

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