



## Promoting effect of Al<sub>2</sub>O<sub>3</sub>/ZnO-based nanofluids stabilized by SDS surfactant on CH<sub>4</sub>+C<sub>2</sub>H<sub>6</sub>+C<sub>3</sub>H<sub>8</sub> hydrate formation



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### ABSTRACT

The transportation and storage of gas in a safe, efficient and cost-effective manner is always a challenge. Gas hydrate has been considered as a good alternative for storage and transportation media of natural gas. But the main disadvantage of this method is the slow rate of hydrate formation. Therefore, for large-scale industrial application purposes, the present investigation focuses on increasing the rate of hydrate formation and amount of gas consumed by adding aluminum oxide (Al<sub>2</sub>O<sub>3</sub>) and zinc oxide (ZnO) nanoparticle to water. Three different concentrations (0.1, 0.4 and 0.8% (wt)) of nanoparticles in aqueous solution have been studied to know their effect on the kinetics of CH<sub>4</sub> + C<sub>2</sub>H<sub>6</sub> + C<sub>3</sub>H<sub>8</sub> hydrate formation. To stabilize the prepared solution, 0.03% (wt) SDS has been added to the aqueous solution. The results show that addition of both the nanoparticles increases the gas consumption rate and amount of gas consumed. The amount of gas consumed in presence of both the nanoparticle increases by almost 121% compared with that of pure water system. The storage capacity also increases with the addition of both nanoparticles. Induction time and hydrate growth rate have also been discussed.

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### Introduction

Natural gas is gaining gradually great potential as an essential energy source for the future. Global energy trend indicates the increasingly important role of natural gas in the growth of energy markets through 2035 [1]. It is predicted that the natural gas demand increases at an average rate of 2.4% annually until 2030 in the world [2]. One of the main reason for its rapid consumption is because of growing demand for clean energy and environmental concerns. Its low greenhouse gas emission along with relatively reduced capital cost makes natural gas as the preferred energy source for different sectors.

Consumption of natural gas is very fluctuating. The average variability in the consumption of natural gas is much greater than the average variability in production. Moreover, the location of the demand areas is away from the production areas. Thus, storage and transportation plays a vital role in the competitive natural gas markets [3]. Among different methods, storing and transportation of gas in the form of hydrate gains interest in recent times due to high gas content per volume of hydrate, environmentally friendly and as well as its non-explosive nature possesses no safety risk [4].

Gas hydrates are crystalline, non-stoichiometric compound formed from water and light hydrocarbon. When gas and water molecules are kept at high pressure and low temperature condition they form solid, ice-like hydrate in which gas molecules are trapped inside the cages formed by the hydrogen bonded water molecules. Depending on the size and type of guest molecule all known common hydrate forms three types of structure: structure I (sI), structure II (sII), and structure H (sH). For a unit cell, sI hydrate consists of two 5<sup>12</sup> and six 5<sup>12</sup>6<sup>2</sup> cages; sII hydrate consists of sixteen 5<sup>12</sup> and eight 5<sup>12</sup>6<sup>4</sup> cages; and sH hydrate consists of three 5<sup>12</sup>, two 4<sup>3</sup>5<sup>6</sup>6<sup>3</sup> and one 5<sup>12</sup>6<sup>8</sup> cages [5]. Among many technological applications, natural gas hydrate has unique gas storage properties. It can contain 150–180 v/v (at standard pressure and temperature) of gas [6,7]. But for large-scale industrial application, the main concern regarding storing and transportation of gas as hydrates are the hydrate formation rate. Because higher formation rate reduces the operation cost. So research works are going on to increase the formation rate of hydrate.

Many researchers have studied different methods to promote the hydrate formation process. Among these the use of some surfactants and nanoparticles shows promising results [8–13]. Zhang et al. [14] studied the role of SDS in methane hydrate formation in a non-stirred batch reactor and found that SDS accelerates the hydrate growth. Yoslim et al. [15] reported that during hydrate formation in presence of anionic surfactants, some

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branched porous fibre-like crystals were formed instead of dendritic crystals formed in the absence of any surfactant. They had also noticed that the addition of SDS increases the gas consumption by approximately 14 times when compared with pure water. Kumar et al. [10] studied the effect of nonionic surfactant Tween-80 (T-80), cationic dodecyltrimethylammonium chloride (DTACI) and anionic Sodium Dodecyl Sulphate (SDS) on the CO<sub>2</sub> hydrate formation kinetics in a fixed bed crystallizer. They had found that out of the three surfactants used, SDS was found to be the most effective in enhancing the rate of hydrate formation as well as reducing the induction time. Du et al. [16] studied the effect of four different ionic surfactants on methane hydrate formation kinetics in a static reactor. They had mentioned that SDS reduces the energy barrier to nucleation by adsorbing on the hydrate nuclei.

Li et al. [17] reported that addition of copper nanoparticle enhances the heat and mass transfer process of HFC134a hydrate formation. Park et al. [18] found that addition of multi-walled carbon nanotubes increases the amount of gas consumption by 300% compared with pure water together with the decrease in formation time. Park et al. [19] studied the methane hydrate formation in presence of MWCNTs (multi-walled carbon nanotubes) and OMWCNTs (oxidized multi-walled carbon nanotubes). They had found that carbon nanofluid system accelerated the rate of methane hydrate formation. The amount of gas consumed in presence of oxidized carbon nanofluid was approximately 4.5 higher than that of pure water. Arjang et al. [20] also reported that silver nanoparticle decreased the induction time of methane hydrate formation while it increased the amount of gas consumed. Enhancement of growth rate of methane hydrate during formation in presence of multi-walled carbon nanotubes (MWCNT) had also been noticed by Pasięka et al. [21]. Mohammadi et al. [12] studied the kinetics of CO<sub>2</sub> hydrate formation in presence of silver nanoparticles and SDS. They had found that SDS and silver nanoparticles significantly increases the storage capacity of CO<sub>2</sub> hydrate. It had been also noticed that formation rate of hydrate also increased with the addition of SDS and silver nanoparticles. Choi et al. [22] also studied the effect of different nanoparticle (Aluminum oxide (Al<sub>2</sub>O<sub>3</sub>), Cerium oxide (CeO<sub>2</sub>), Silicon dioxide (SiO<sub>2</sub>)) on CO<sub>2</sub> hydrate formation and noticed that maximum CO<sub>2</sub> hydrate formation rate was obtained with 0.6% (wt) SDS and 0.2% (wt) Al<sub>2</sub>O<sub>3</sub> nanoparticles. Zhou et al. [23] studied the effect of graphite nanoparticle on CO<sub>2</sub> hydrate formation. They had found that addition of graphite nanoparticle reduced the induction time while it increased the gas consumption. Najibi et al. [13] also

reported that SDS and CuO nanoparticle increases the amount of gas consumption, rate of gas consumption and mole percent of water to hydrate conversion during hydrate formation.

In the present work, we have studied the effect of aluminum oxide and zinc oxide-based nanofluids stabilized by SDS surfactant on CH<sub>4</sub> + C<sub>2</sub>H<sub>6</sub> + C<sub>3</sub>H<sub>8</sub> hydrate formation as a promoter. The objective of this work is to see whether those two nanoparticles can be used as an additives for storing gas as hydrates. Induction time, gas consumption, formation rate, water conversion, storage capacity and hydrate growth rate were calculated in presence of both Al<sub>2</sub>O<sub>3</sub> and ZnO nanoparticles.

## Experimental

### Experimental apparatus

The schematic diagram of the experimental setup is shown in Fig. 1. Our previous works [24,25] describe the experimental setup in detail. Briefly, the setup consists of a thermostatic bath, cell, booster and data acquisition system. The bath works with a thermostatic fluid (a mixture of 85% water and 15% glycol) to control the inside temperature of the cell with an operating temperature range from -10 to 60 °C. The bath size is about 225 × 370 × 429 mm. The cell is a stainless-steel cylinder with a capacity of 250 cm<sup>3</sup>. Maximum allowable working pressure is 20.68 MPa. The cell is equipped with a PT100 probe to measure the temperature inside the cell, the uncertainty of the measurement being ±0.05 K. The cell pressure is measured by a pressure transducer with an accuracy of 0.1% (Full scale). A magnetic stirrer with adjustable rotation speed (upto 1000 rpm) is used to agitate the fluid inside the cell. The cell is connected to the gas cylinder through a booster to build up high pressure inside the cell. The vacuum pump seen in the Fig. 1 is used to evacuate air in the cell before inserting gas. The data are recorded by a compute running an AppliLab (v6.0) software program.

### Materials

The gas mixture used in the present study were supplied by Ultra Pure Gases (I) Pvt. Ltd. Gujarat, India. The composition of the gas mixture is given in the Table 1. The specification of nanoparticle used in this study along with purity and their suppliers is reported in Table 2. Distilled water produced by a laboratory purification system was used in all experiments.

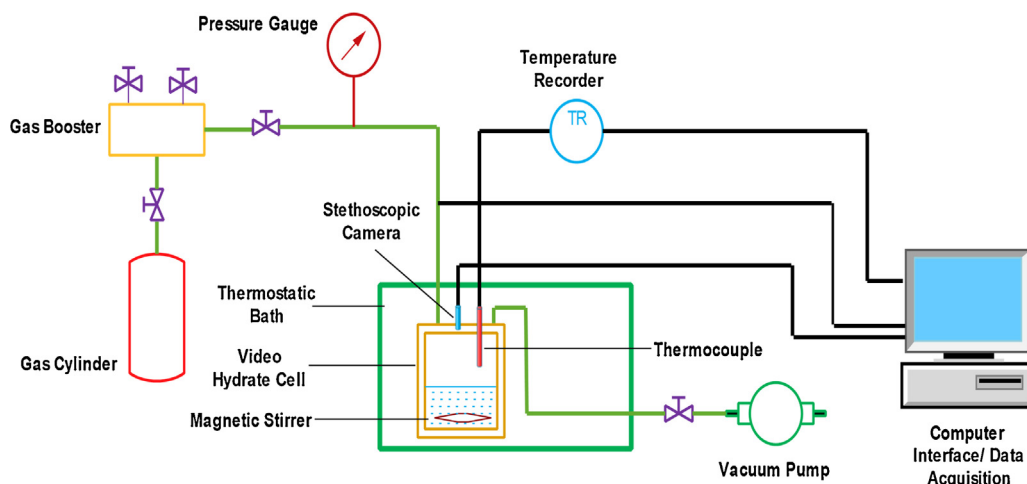


Fig. 1. Schematic diagram of the experimental setup.

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