



Effect of ultrasonic waves on dissociation kinetics of tetrafluoroethane (CH₂FCF₃) hydrate



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ABSTRACT

The effect of ultrasonic waves on formation and dissociation kinetics of gas hydrates has been experimentally studied by several researchers. Some results indicated that ultrasonic waves have the effect of reducing gas hydrate induction time before the initiation of hydrate formation, while others reported on the effect of prohibiting the growth kinetics during hydrate formation. There have also been reports regarding an increase in the amount of gas consumed and a decrease in the hydrate formation time. The present study examines the influence of ultrasonic waves on gas hydrate dissociation. Gas hydrate formation and dissociation occurred in a temperature-controlled 15 L horizontal cylindrical reactor. The reactor was agitated with a pair of flat blades installed in radial direction and a pair of rollers, which were driven by a motor connected through a magnetic coupler. R-134a, a gas that can form hydrates at a relatively low pressure, was used to form gas hydrate slurry. Dissociation was augmented by 20 kHz ultrasonic waves generated with a piezoelectric converter and transmitted through a vibrating horn whose tip was in contact with the hydrated slurry in the reactor. To compare the effect of energy input in the form of ultrasonic waves with that of the same amount of heat input, equivalent amount of heat was provided to the reactor. Dissociation rate and the accumulated dissociation time were compared between the cases of heat addition and ultrasonic wave irradiation. Results show that not only does the dissociation rate increase with an increase in the ultrasonic wave power, but the irradiation of ultrasonic waves also enhances the dissociation of R-134a hydrate more effectively than the application of an equivalent amount of energy in the form of heat at the same location.

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Introduction

Gas hydrates are clathrate solid compounds where guest molecules are trapped inside cages consisting of hydrogen bonded water molecules, and are thermodynamically stable under pressures higher than equilibrium values at a given temperature. Of the various guest molecules which can form hydrates, hydro-fluorocarbon (HFC) hydrate is stable at relatively low pressures for a given temperature, making them attractive candidates for economical hydrate formation [1–4]. Many experimental studies have been carried out to date on the formation and dissociation of refrigerant hydrates [5–11]. Negma et al. [6–8] and Eslamimanesh et al. [10] developed thermodynamic models to predict the dissociation conditions of some refrigerant hydrates experimentally and theoretically.

The relatively less harsh conditions under which it can form gas hydrates make HFCs an economically attractive guest molecule candidate for obtaining potable water from seawater via gas hydrate desalination [11]. The desalination of seawater utilizing gas hydrate technology has received continuous attention since the 1940s as an alternative method to the multi-stage flash distillation and reverse osmosis processes [12–14], together with treatments of industrial waste water [15]. To compete with the conventional desalination methods, hydrate formation and dissociation processes have to be economical in energy consumption. Moreover, for commercial application it is necessary to drastically increase the kinetics of formation/dissociation processes [16–19]. There have been several studies to increase the rate of gas hydrate formation/dissociation, which include exposure of hydrates to ultrasonic waves during formation/dissociation processes [20–22]. Ultrasonic wave irradiation is also known to be an effective method to enhance mass transfer in alcoholysis [23]. Some have reported on the hydrate inhibition mechanisms of ultrasonic waves [24,25], and there has been a report on the enhancement of methane

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Nomenclature

C_p	Specific heat at constant pressure ($\text{J kg}^{-1} \text{K}^{-1}$)
E_{total}	Total energy of heat supplied to the reactor (kJ)
$E_{\text{dissociation}}$	Energy used for dissociation out of the total energy supplied (kJ)
E_{sensible}	Energy used in changing the temperature of the gas hydrate and the water (kJ)
Δf	Fugacity difference, fugacity(equilibrium)-fugacity(experiment) (kPa)
$\Delta h_{\text{hydrate}}$	Heat of hydrate formation (kJ kg^{-1})
n_{hydrate}	Number of moles, hydrate (mol)
$n_{\text{hydrate_diss}}$	Number of moles, dissociated hydrate
$n_{\text{hydrate_water-bath}}$	Number of moles, amount of hydrate dissociated from the water jacket without additional energy input
$n_{\text{hydrate_net}}$	Number of moles, additionally dissociated hydrate
n_{water}	Number of moles, water (mol)
P_c	Critical pressure (kPa)
t	Time (min)
T_c	Critical temperature (K)

Greek letters

η	Thermal energy efficiency
ω	Acentric factor

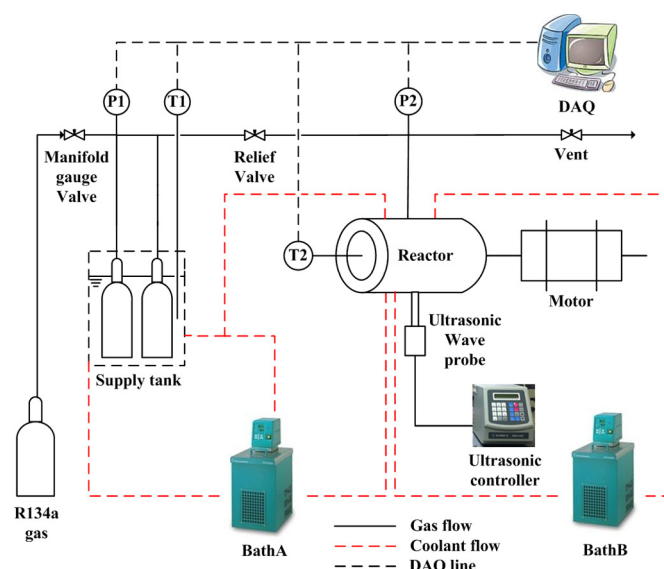


Fig. 1. Schematic diagram of experimental apparatus (P1, P2: pressure transmitter, T1, T2: T-type thermocouple, solid line: gas flow, red dotted line: coolant flow, dotted line: data acquisition). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

presented in Fig. 1. The cylindrical high pressure reactor, which provides approximately 15 L, has an inner diameter and length of 250 mm and 500 mm respectively, and is made of SUS306 material. To increase the rate of hydrate formation and dissociation, efficient mixing was facilitated with an impeller, consisting of two rollers and two flat blades that are perpendicular to each other and attached to a horizontal axis installed along the center of the cylindrical reactor. The blades, which are 5 mm thick, 40 mm wide and 500 mm long, rotate while maintaining a 5 mm gap from the reactor's inner surface. The rollers, pressed against the cylindrical reactor surface by springs, are used to scrape off the hydrates formed on the surface. The rotating axis of the impeller is attached to the driving motor through a magnetic coupling to ensure sealing and can be rotated at a speed of up to 64 rpm. The shape and layout of the rollers and the blades are presented in Fig. 2.

hydrate formation resulting in a decrease in formation time [20]. Others have studied the effect of ultrasonic waves specifically on hydrate dissociation. Miura et al. [21] looked into the effect of underwater ultrasonic irradiation and found that it promoted the dissociation of methane hydrates as a facilitator and reduced the dissociation time. Tsuyuki et al. [22] carried out experiments on methane hydrate dissociation in sediments comparing the cases of using depressurization only, using ultrasonic irradiation only, and using both. It was found that the dissociation rate increased dramatically when the two methods were combined.

In this study we investigate the effect of ultrasonic wave irradiation on the dissociation kinetics of tetrafluoroethane (CH_2FCF_3) gas hydrate. Tetrafluoroethane, also known as R-134a, is a haloalkane refrigerant gas in the family of HFC refrigerants. We constructed a 15 L horizontal cylindrical reactor with rotating blades attached to the center axis to promote formation. A 20 kHz ultrasonic wave horn was inserted into the reactor and operated during the dissociation process. Since there already are many studies that report a definite increase in the dissociation rate, our aim is not merely to confirm the enhancement of the dissociation rate but more specifically to isolate the rate enhancement that can be attributed solely to the ultrasonic wave effect, by comparing it with the enhancement achieved through the application of an equivalent amount of energy in the form of heat. By comparing the dissociation characteristics we intend to ascertain the effect of energy addition in the specific form of ultrasonic waves on the dissociation rate of gas hydrates.

Hydrate formation/dissociation experimental apparatus and procedures

Experimental apparatus

A schematic diagram of the experimental apparatus used for formation and subsequent dissociation of R-134a hydrates is

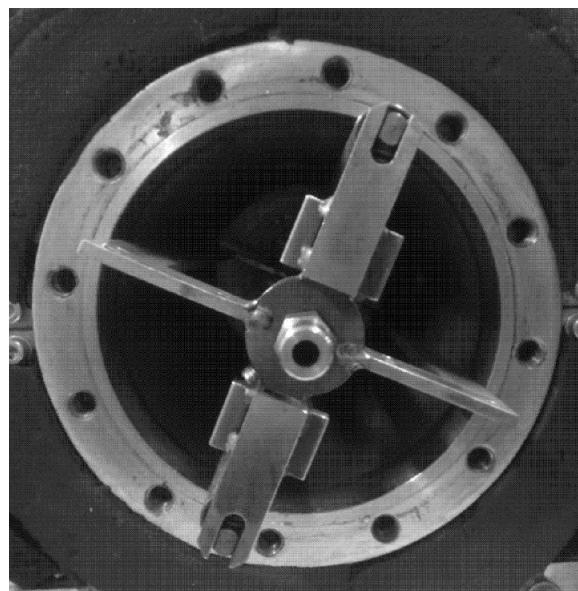


Fig. 2. Side view of the reactor with rollers and blades.

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