



Effect of methane–sugar interaction on the solubility of methane in an aqueous solution

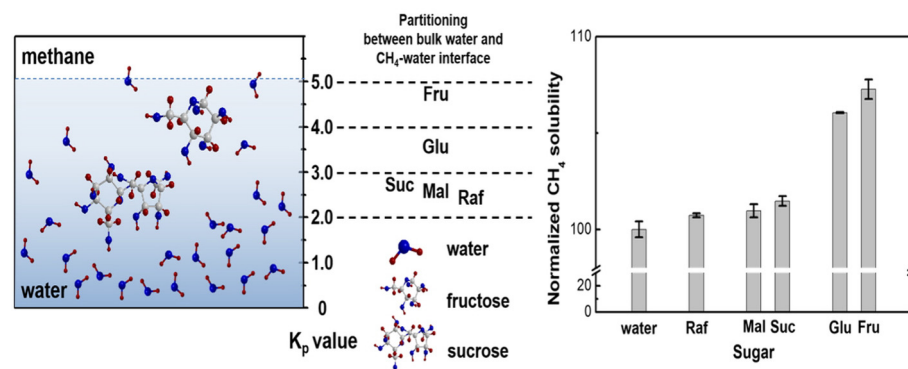


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GRAPHICAL ABSTRACT



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ABSTRACT

In this study, the effect of methane–sugar interaction on the solubility of methane in an aqueous solution at ambient pressure was investigated. Various sugars, such as fructose, glucose, sucrose, maltose, and raffinose, were used, and depending on the type and concentration of sugar, the methane solubility increased from 21.72 mg/L (in pure water) to 24.86 mg/L. Sugars with a low hydrogen-bonding number between the water and sugar molecules exhibited a large enhancement in methane solubility. The solute partitioning model and molecular dynamics simulations were employed to verify the results obtained for the experimental solubility of methane in aqueous sugar solutions.

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1. Introduction

In various industrial engineering fields, one of the important parameters to consider is the solubility of gases in liquid solutions [1,2]. Such interactions are observed in gas anti-solvent crystallization systems and gas hydrate fermentation processes, as well as in petroleum refineries [3,4]. In particular, methane,

which represents the major component of shale gas, has recently attracted immense attention from the gas fermentation industry [5,6]. In gas fermentation, methane dissolved in an aqueous culture medium acts as the major carbon source for microorganisms present in the medium to produce desired products [7,8]. However, the low solubility of methane in the culture medium (less than 21.7 mg/L at STP) limits the methane conversion efficiency [9]. Hence, novel approaches are required to enhance the solubility of methane in an aqueous solution.

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The solubility of a gas in an aqueous solution depends on several factors, such as temperature, the partial pressure of the gas over the liquid, and the characteristics of both solvent and gas [10]. The addition of certain additives (e.g., nanoparticles and electrolytes) to water has been reported to either increase or decrease the solubility of a gas in water [11,12]. Among the various mechanisms that can be utilized to interpret a change in the solubility of a gas in a solution, the relationship between the surface tension of water and the solubility of gas in water has been extensively studied [13,14]. In accordance with this interaction, additives known as chaotropes serve to decrease the surface tension of water under a certain gas, thereby increasing the solubility of a gas in the solution [14]. Conversely, additives known as kosmotropes serve to increase the surface tension of water under a certain gas, thereby decreasing gas solubility [15,16]. The scaled particle theory (SPT) has been widely investigated for understanding the solvation of gases in fluid media via the use of molecular parameters and intermolecular interactions [17,18]. However, the hard sphere model employed in SPT limits the use of such a theory; for example, SPT cannot be used to explain the increase in gas solubility observed with the use of additives that decrease the surface tension of the solution [19]. Recently, Pegram and Record have introduced the solute partitioning model (SPM) for investigating the relation between the solubility of a gas in a solution and the surface tension change of the solution [20,21]. According to SPM, additives that decrease the solubility of gas in a solution are repelled from the gas–water interface, while additives that increase the solubility of gas in a solution are located close to the gas–water interface [21,22]. As compared to the SPT, SPM offers an instinctive interpretation of the inner working of such systems [22].

The additives used for efficient fermentation of methanotrophs should exhibit the following characteristics: (i) they should increase the solubility of methane in the aqueous solution; (ii) they should not hinder the growth of methanotrophs; and (iii) methanotrophs should not use the additives as a carbon source [23]. Considering these properties, sugars are good candidates for use as additives because of their bio-friendly nature; it is well known that methanotrophs do not use sugars as a carbon source [24,25]. However, the effect of sugar molecules on the surface tension of water is unclear. Some researchers have reported that sugar molecules serve to increase the surface tension of water, while other researchers have reported a decrease in the surface tension of an aqueous solution with the addition of sugar [26,27]. To the best of our knowledge, the solubility of methane in various aqueous sugar solutions has not been reported; hence, in this study, the effect of sugar on the surface tension of water under methane was investigated, and the solubility of methane in the aqueous solution was examined.

In this study, the maximum solubility of methane in various aqueous sugar solutions was measured as a function of sugar concentration. Various sugars with different numbers of carbohydrate rings and different chemical structures, such as fructose, glucose, sucrose, maltose, and raffinose, were employed. Furthermore, SPM and molecular dynamic simulations were employed in order to corroborate the trend in the solubility of methane in aqueous sugar solutions by measuring the surface tension of water under methane.

2. Experiment and theoretical methods

2.1. Reactor setup

Bubble column reactor with 4-pitched blade impeller was used to measure solubility of methane in aqueous solution in this study. Rubber cap was located at the side of the reactor to harvest solutions with syringes. Temperature of the solution was maintained

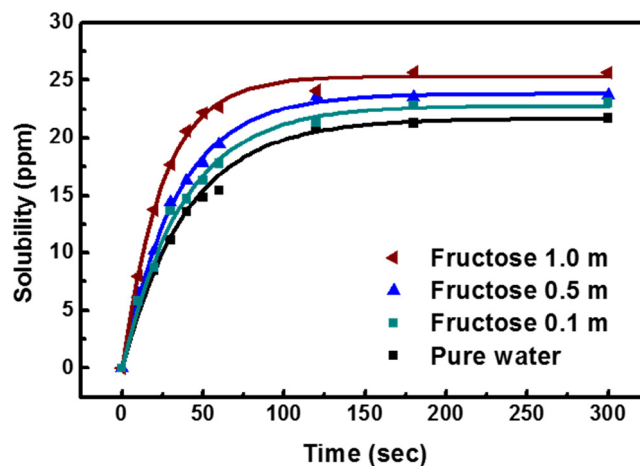


Fig. 1. Concentration of methane in aqueous fructose solutions with various concentrations.

at 30 °C during experiment by water-jacket and water-circulator (Wisecircu, Wisd Laboratory Instruments Co.) around the reactor [28]. Methane gas was injected continuously into the reactor at 3 L/min near the impeller (300 rpm) by alumina bubbler.

2.2. Determination of methane solubility

Aqueous sugar solutions of x molal concentration ($x = 0.1, 0.5$ and 1.0) were prepared by dissolving sugars in distilled water. While methane gas was flowed into the reactor, the sugar solutions were harvested from the reactor at different times of 10, 20, 30, 40, 50, 60, 120, 180, and 300 s by disposable syringes. Then, the solutions were transferred into vials with sealed cap and were heated and maintained at 95 °C for 1 h in a heater (DMB-2, Misung Instrument Co.) to evaporate methane completely. Then, 0.4 mL of head space gas was taken by gas-tight syringe from the vials, and was measured by gas chromatography (ACME 6100, Young Lin Instruments Co.) with flame ionization detector [29].

2.3. Measurement of surface tension

Surface tension of aqueous sugar solutions was measured using computer-controlled surface tensionmeter (Biolin scientific; model, Sigma702) by Du Nouy's ring method under methane-saturated environment [30].

2.4. Thermodynamics of methane–water surfaces and analysis on surface tension using the solute partitioning model

According to investigation by Pegram and Records, Gibbs–Duhem equation can be converted to Eq. (1) [21,22].

$$\frac{d\gamma}{dO_{sm}} = -\frac{RT}{\bar{m}_1} \left(\frac{n_1^s}{A} \right) (K_p - 1) \quad (1)$$

where γ is the surface tension, O_{sm} is the osmolality, R is the ideal gas constant, T is the absolute temperature, \bar{m}_1 is the molality of water (55.5 mol/kg), n_1^s/A is the number of water molecules in the surface region per unit area of surface, and K_p is the surface-bulk partition coefficient (See supporting information for detailed calculation) [31,32]. The $d\gamma/dO_{sm}$ value can be calculated from the slope of surface tension change as a function of osmolality (Fig. 3). The n_1^s/A value will be used as $0.20 \cdot \text{H}_2\text{O} \text{ \AA}^{-2}$ as used in previous literatures [21,22]. From these calculation, we tried to get K_p by using surface tension increment (STI), which is $d\gamma/dO_{sm}$. K_p is the

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