



Full Length Article

Molecular dynamics simulations of natural gas-water interfacial tensions over wide range of pressures

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ABSTRACT

Natural gas-water interfacial tension (IFT) plays an important role in gas production and transportation as well as hydrate formation. While experiments can measure IFT from macroscopic perspective, the interfacial phenomena remain less clear at molecular level. In this work, we use molecular dynamics (MD) simulations to study hydrocarbon-water IFT up to 5000 bar at various temperature conditions. At each temperature, we study the IFT of C₁-H₂O system, C₁ + C₂ mixture-H₂O system, C₁ + C₃ mixture-H₂O system, and C₁ + C₂ + C₃ mixture-H₂O system. We find that IFTs decrease with increasing temperature at low pressure conditions, while the differences become insignificant at high pressures. Addition of C₂ and C₃ can lower IFT in line with the previous experimental findings, while C₃ has a more pronounced effect than C₂. However, after a certain pressure, IFT becomes similar for various hydrocarbon mixture-H₂O systems. As pressure further increases, IFT gradually increases. At low and intermediate pressures, hydrocarbons can form adsorption layers on gas-water interfaces but become less significant at high pressures. We find that IFT decreases when the relative adsorption obtained from density distributions is positive but increases for negative relative adsorption at high pressures. This finding agrees well with previous molecular simulation work on C₁-water interfacial tension. At low and intermediate pressures, relative adsorption becomes more significant when the heavier components (C₂/C₃) are added, resulting in a more pronounced IFT reduction effect. Our study provides fundamental understandings about the interfacial phenomena between hydrocarbon and water, and important insights into the energy prospection and flow assurance problems.

1. Introduction

Natural gas-water interface is a ubiquitous phenomenon in natural gas production and transportation as well as gas hydrate formation. During natural gas production from sub-formation, gas is usually in contact with water while flowing in porous media [1–3]. The gas-water interfacial tension (IFT) can significantly affect capillary pressure which can cause serious pore blocking problems during two-phase flow, especially in tight gas reservoirs [4,5]. Gas-water IFT also plays an important role in gas hydrate formation, which is one of important unconventional energy resources with massive amount deposited on the deep ocean floor [6,7]. On the other hand, gas hydrate formation can cause flow assurance problems when gas flows in pipelines and wellbores [8–10]. Hydrate formation occurs from crystal growth at gas-water interface when gas molecule concentrations are sufficiently high. Then, a thin film of hydrates on the water surface forms, and further growth is dominated by mass transfer through the film [7,9,11–13]. A number of works have concluded that low IFT can accelerate hydrate

formation [12,13].

In the past few decades, there have been a large number of experimental works [1,2,7,13–20] on the measurement of IFT between water and methane which is the major constituent of natural gas. These works have shown that as pressure increases, methane-water IFT presents a sharp decrease first, then followed by a moderate decrease, and finally slowly increases [1,2,16]. In addition, IFT generally decreases as temperature increases [7]. In addition to methane, other hydrocarbon molecules, such as ethane and propane, are also important constituents of natural gas. Recently, Khosharay and Varaminian [20] measured the IFTs of methane-water, ethane-water, and propane-water from 284.15 K to 312.15 K and pressure up to 60 bar. They reported that IFTs between gas and water in all three systems decrease with increasing pressure and temperature. Moreover, the decrease in IFTs with pressure for the heavier hydrocarbons is more significant than the lighter ones. Hayama et al. [11] measured the IFT between natural gas mixtures (methane + ethane + propane) and water from 283.2 K to 298.2 K and up to 100 bar. They found that natural gas-water IFT decreases as

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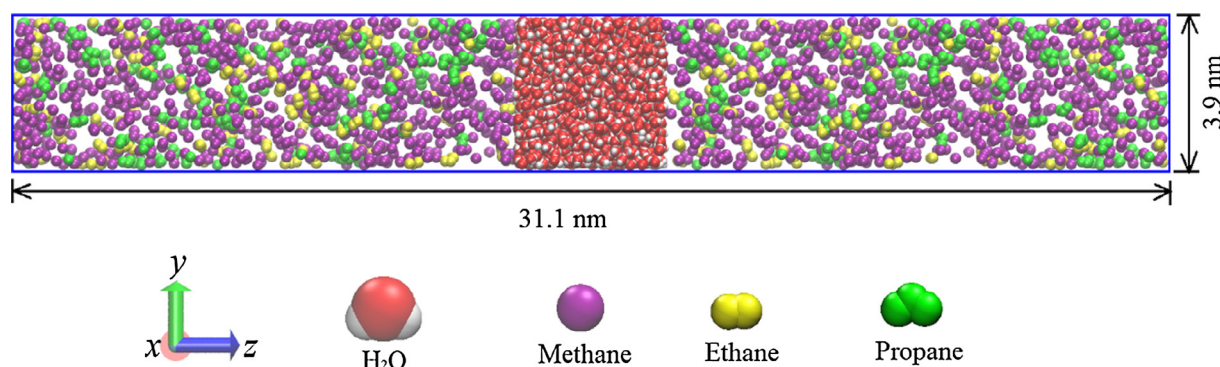


Fig. 1. Schematic representation of hydrocarbon mixture-water system. Red spheres represent oxygen atoms, white spheres represent hydrogen atoms, purple spheres represent methane pseudo atoms, yellow spheres represent ethane pseudo atoms, and green spheres represent propane pseudo atoms. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Table 1
LJ parameters and partial charges of hydrocarbon and water models.

	σ (Å)	ϵ/k_B (K)	q (e)
Hydrocarbon (TraPPE model) [37]			
CH ₄	3.73	148	0
CH ₃ -	3.75	98	0
-CH ₂ -	3.95	46	0
Water (TIP4P/2005 model) [38]			
O	3.1589	93.2	0
H	0	0	0.5564
M	0	0	-1.1128

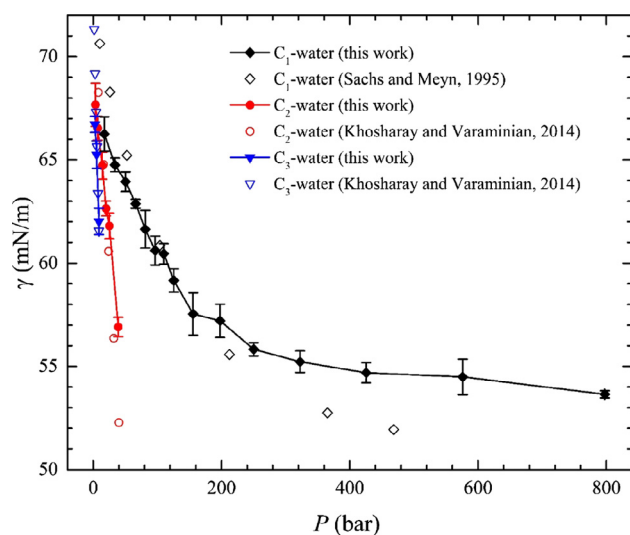


Fig. 2. IFTs of pure C₁-water, C₂-water, C₃-water systems from MD simulations and experimental data at 298.15 K.

pressure increases at all temperature conditions. Furthermore, they observed the larger content of ethane and propane, the faster IFT decrease as pressure increases. While experimental measurements can provide some insights into the IFTs between gas and water, these works are generally limited to a small range of temperature and pressure conditions and the underlying mechanisms about the IFTs between gas and water are still less clear.

On the other hand, a number of modeling [2,21–28] and molecular

simulation [8,26,27,29–31] works have been reported on the IFT between methane and water. Most of modeling works are based on statistical thermodynamics and take into account the density gradients at interfaces, such as integral and density functional theories [21,22] as well as density gradient theory (DGT) [2,23–28]. Simulation works, either based on Monte Carlo (MC) simulation [26,27,29] or Molecular Dynamics (MD) simulation [8,27,30,31], which explicitly consider intermolecular interactions, have been applied to study the IFT. In general, all the theoretical and simulation results show that the IFT between methane and water decreases steeply with increasing pressure first and then followed by a moderate decrease, and finally gradually increases at elevated high pressures [1,20,26,31]. In particular, molecular simulation can study the interfacial phenomena at molecular scale, which is hardly accessible by experiments [11,31,32]. Although these works have provided the underlying mechanisms about interfacial phenomena of pure methane, CO₂ and their mixtures with water, the IFTs between hydrocarbon mixtures-water remains less understood.

Therefore, in this work, we use MD simulations to study hydrocarbon-water IFT under wide range of pressure (from about atmospheric pressure up to ~5000 bar) and various temperature (278.15 K and 298.15 K) conditions. We first calibrate our simulations by comparing the IFT between methane-water, ethane-water, and propane-water to that from experimental measurements. Then, we study the IFT between various binary, ternary hydrocarbon mixtures and water and explicitly investigate the effects of pressure, temperature, gas mixture compositions. The underlying mechanism of IFT decrease due to the addition of ethane and propane is explained by density profiles and relative adsorption [33,34].

The remainder of this paper is organized as follows. In Section 2, we introduce simulation systems and procedures. In Section 3, we present simulation results and investigate the effects of pressure, temperature, and gas phase compositions on the IFT between pure and hydrocarbon mixtures-water. In Section 4, we summarize the key conclusions and discuss the potential applications.

2. Simulation methodology

2.1. Simulation system

In this work, we employ GROMACS package [35,36] in canonical ensemble to obtain the thermodynamic properties of hydrocarbon-water interfaces. During each simulation, the number of molecules (N), system volume (V), and temperature (T) are fixed. The simulation box

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