



Measurement and modelling of interfacial tension in methane/water and methane/brine systems at reservoir conditions



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ABSTRACT

It is well established that the presence of salt in the aqueous phase raises the interfacial tension (IFT) of gas–water systems when compared to the case of pure water under the same pressure and temperature conditions. However, experimental data for gas–brine systems is still scarce, in particular at high pressure and high temperature (HPHT) conditions. In this communication, IFT data for methane with distilled water were experimentally determined using the pendant drop and bubble rise methods for temperatures ranging from (311–473) K and pressures up to 92 MPa and the results compared against literature data. Moreover, for the first time, the effect of NaCl on the IFT of the methane–water system was investigated in solutions with a maximum salinity of 10 wt% at HPHT conditions.

The Cubic-Plus-Association equation of state (CPA EoS) along with the Debye–Hückel activity model was used to describe the effect of salt in the phase behaviour of the methane–water system and bulk properties used to compute the IFT of the studied systems with the Density Gradient Theory (DGT). The modelling results showed that by using only bulk phase properties and one temperature independent binary interaction parameter adjusted to the methane–water system, the DGT was able to predict the impact of NaCl on the IFT with remarkably low deviations from measured values. Furthermore, the impact of the fluids microstructure and the distribution of molecules across the interface on the IFT were evaluated with the DGT.

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1. Introduction

The IFT of gas–water and gas–brine systems is one of the most important parameters in petroleum and process industries. IFT can affect water-gas contact movement and distribution of hydrocarbons in a reservoir, water alternating gas drive, gas-injected enhanced oil recovery processes and multiphase flow calculations [1]. Injecting and storing acid gases like carbon dioxide and hydrogen sulphide separated from sour natural gas into reservoir has grown interest in the past decade [2,3] and the accurate knowledge of this property is also of key importance for the efficient and economical design of potential processes.

The most commonly used techniques for the experimental determination of the IFT are the Pendant Drop or Bubble Rise methods. These approaches involve measuring the dimensions of a droplet of liquid in equilibrium vapour (*i.e.* Pendant Drop) or a gas bubble in equilibrium liquid (*i.e.* Bubble Rise), which when

combined with density data, can be used to estimate the IFT of the equilibrated phases. An examination of the literature shows that several studies have investigated the interfacial tension between pure water and mixtures containing common gases such as carbon dioxide, nitrogen and methane in a broad range of pressures and temperatures. Reviewed data can be found in the works of Schmidt et al. [4], Rushing et al. [5], Shariat et al. [6] and Pereira et al. [7] and references therein. However, hydrocarbon reservoirs fluids are often in equilibrium with saline formation water.

The presence of salts can affect the physical properties of the fluid and in particular the IFT of aqueous interfaces. The addition of salt(s) to water is known to increase the interfacial tension of water-rich systems [5,8–19]. This effect has been attributed to the change in the structure of the interface due to the negative adsorption (*i.e.* depletion) of cations in the interface and therefore, to their tendency to accumulate in the aqueous phase [20–23]. This change is to some extent related with the disruption of hydrogen bonding forces due to the high concentration of molecules from another phase near the interfacial region. As a result, cations affinity towards the interface is reduced. Moreover, the addition of

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salts increases the average molecular weight of the aqueous phase which, under the assumption that the excess volume of mixing is zero, results in an overall density increase and in turn of the IFT values.

Several authors have reported the IFT for different brine and liquid hydrocarbon systems. Cai et al. [9] reported IFT data on brine and pure liquid hydrocarbons such as n -C₆, n -C₈, n -C₁₀, n -C₁₂, n -C₁₄ and n -C₁₆ and their mixtures for pressures and temperatures up to 30 MPa and 353 K, respectively. Badakhshan and Bake [24] presented data on the IFT of n -C₆, cyclohexane and toluene with brine. IFT data on n -hexane and brine systems were also reported by Ikeda et al. [25]. However, IFT experimental data on gas–brine systems at hydrocarbon reservoir conditions (HPHT) are scarce in the literature and, to best of our knowledge, still limited to the IFT of brine–natural gas [5] and brine–carbon dioxide [8,10–19].

As happens with any new or already existing processes, the design and/or operation optimization of petroleum engineering processes requires the deployment of theoretical tools for accurately estimating this property. Numerous approaches have been proposed and many empirical or semi-empirical methods are being used as standard models in commercial simulators for the oil and gas industry. Among all, the Parachor method [26,27] and the scaling law [28] gained the most attention due to its simplicity and accuracy for describing the IFT of vapour–liquid interfaces in hydrocarbon systems. Even though a Parachor value has been reported for water, this method is not recommended for the prediction of IFT of aqueous interfaces [1,29]. Instead, a more accurate and reliable model for estimating the interfacial hydrocarbon–water systems was proposed by Firoozabadi and Ramey [29]. In their work, the authors successfully correlated the IFT between methane and hydrocarbons with water against the density of the equilibrated phases over a broad range of conditions. Later, Argaud [30] and Sutton [31] developed new versions of the original correlation from Firoozabadi and Ramey [29] by readjusting the model to a broader class of compounds and larger database. Although good results were obtained with these approaches, they are limited on their transferability and predictive capabilities.

Other methods, based on statistical thermodynamics, take into account the density gradients in the interface like the perturbation theory [32], integral and density functional theories [32–35], or the density gradient theory (DGT) [36,37]. In particular, the latter has been thoroughly applied in the prediction of IFT values of a wide class of systems and interfaces. The reader is referred to the study of Pereira et al. [7] and references within for further details.

Of relevance for this study, Yan et al. [38] and Schmitt et al. [4] were able to describe the IFT of aqueous systems containing methane and common gases over a broad range of conditions by coupling the simplified version of the DGT, originally introduced by Zuo and Stenby [39] (known as Linear Gradient Theory or LGT), with classical cubic equations of state. Khosharay and co-workers [40,41] extended the work to association equations of state (EoSs) and successfully described the IFT of aqueous systems with the LGT. However, the computation of IFT values in a predictive manner with the LGT is very limited as in general there is a necessity to use a considerable number of binary interaction parameters, some of them temperature dependent, regressed against experimental IFT data. On the other hand, Miqueu et al. [42] and Nino-Amézquita and Enders [43] showed that the original DGT when coupled with an appropriate phase behaviour model was capable of predicting the IFT of the methane–water system using bulk phases properties and a single, temperature independent, binary interaction parameter. The computed values were not only in very good agreement with data from the literature over a broad range of thermodynamic conditions but the model was also able to capture the interfacial tension minimum observed by Wiegand and Franck [44] at high

pressures. Moreover, other properties such as species distribution across the interfacial region and interface thickness computed by the DGT revealed to be in very good agreement with molecular simulations [42].

The accurate estimation of IFT values with the DGT relies therefore on a correct description of both composition and density of the bulk phases. For systems containing polar and/or associating compounds, the DGT has been commonly coupled with theoretical based EoSs such as the Statistical Associating Fluid Theory (SAFT), and its variants, or the Cubic-Plus-Association (CPA) to successfully describe the IFT of aqueous interfaces [7,42,43,45–49].

In this communication new experimental IFT data are reported for methane with pure water and aqueous solutions composed of 5 and 10 wt% of sodium chloride for temperatures ranging from (311–473) K and pressures up 92 MPa. The IFT results for methane–water were found to be in good agreement with data from the literature, validating both the method and experimental procedure followed. In addition, the good capabilities shown by the DGT approach for describing the IFT of the methane–water systems was here extended to the methane–brine system by coupling the DGT model with the CPA EoS [50,51] together with the electrostatic term from the Debye–Hückel activity model [52] for accounting the effect of salt on the properties of the bulk phases. All in all, as it will be shown, by accurately describing the effect of pressure, temperature and salt on the density and composition of the equilibrated phases, the DGT was capable of describing the IFT of the methane–water system over a broad range of experimental conditions and of predicting the IFT increase due to the presence of electrolytes.

2. Experimental section

2.1. Materials

The specification and sources of the chemicals used in this work are summarized in Table 1. Aqueous solutions with a concentration of (5.01 ± 0.05) and (10.02 ± 0.11) wt% of NaCl were prepared in this work gravimetrically by adding the salt to degassed and deionized water.

2.2. HPHT PVT cell

The HPHT rig used in this work was configured to examine the phase properties of fluids at representative pressures and temperatures of deep hydrocarbon reservoirs. The experimental equipment can be used for PVT tests on the phase behaviour and properties of pure and multi-component reservoir fluid systems with and without water and in the presence of salt. Dew point, bubble point, phase volumes, interfacial tension, density, and viscosity measurements are the current capabilities of the mentioned apparatus. The physical properties of a wide range of reservoir fluids have been measured and reported so far with this setup [53–62]. The PVT facility is designed to operate from ambient conditions to a maximum pressure of 135 MPa at 523.15 K or 200 MPa at 473.15 K.

The experimental setup (Fig. 1) consists of a high pressure cylindrical equilibrium cell closed at both ends by two sapphire

Table 1
Suppliers and specification of the materials used in this study.

Chemical name	Supplier	Mass fraction purity	Purification method
Methane	BOC	0.99995	None
Water	–	–	Deionized
Sodium chloride	Sigma–Aldrich	>99%	None

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