

Highly accurate liquid–liquid interfacial tension measurement by a convenient capillary apparatus



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ARTICLE INFO

Article history:

Received 7 January 2014

Received in revised form 2 June 2014

Accepted 23 June 2014

Available online 1 July 2014

Keywords:

Alkane–water interface

Capillary constant

Capillary rise method

Hexanoic acid and heptanol–water interface

Liquid–liquid interfacial tension measurement

ABSTRACT

A very useful capillary apparatus is pioneered for the experimental measurement of liquid–liquid interfacial tension of two immiscible liquids, with highly accurate results. The method features integration of two coexisting capillary actions. The results of measurement for alkanes–water, heptanol–water and hexanoic acid–water systems (in the range of 298–333 K) demonstrate that this method of liquid–liquid interfacial tension measurement is highly accurate, as known to be the case for liquid–vapor system. The coexisting capillarity actions involving liquid–liquid system is studied by the capillary constant, and importantly is practiced to be the augmentation of the two neat liquid capillary constants less a length scale proportional to the extent of effective dispersion interaction at the liquid–liquid interface.

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1. Introduction

Capillarity is one of the most interesting phenomena in the nature with great impact, controlling plants sap dynamics, contributing to biological activities, explaining liquid adsorption on porous solid and many more. In supporting, explaining and interpreting these phenomena, the capillary rise method is one of the most accurate techniques for the measurement of liquid surface tension in lab. This method has found many applications due to rather practical surface tension measurement of pure liquids in the range of freezing to critical point [1,2]. Alternative methods with the possibility for automation measurement and efficiency on lab and industrial scale are the so called ring and pendant drop methods [3–5], which require corrections [6], at the level that the nature of measurements demands. However, the capillary rise technique has not been considered inventively for the measurement of tension at interface between two condensed phases like two immiscible liquids. For the present demanding interdisciplinary research works in life science an easy, practical and importantly accurate method of surface tension measurement is highly challenging.

The feature, characterization and measurement of the oil–water interface have been explored widely because of impact in various areas of chemistry, biology, medicine and engineering [7].

In the past decades, various techniques have been used to measure interfacial tension between two immiscible liquid phases [4]. Techniques relevant to drop shape have been implemented and explored rather extensively. Among these, the pendant drop method has been used most frequently. The general success, flexibility and performance of such techniques are drastically sensitive to drop or bubble shape [8]. More generally, the accuracy of the measurements depends mainly on the drop shape and the design of drop experiment [8–10]. Designing and optimizing the drop shape measurement as well as improving the accuracy of the surface tension measurements has been the subject of many investigations [11].

Capillary rise method, in spite of dependence on the contact angle, has been proved to produce highly accurate result for most liquid by using glass capillary. Our easy to use homemade capillary apparatus [12], designed to provide the possibility of surface tension measurement in the absence of atmosphere and under liquid–vapor equilibrium, has recently been recognized to produce highly accurate and correlated surface tension for various liquids and ionic liquids [12–15]. The apparatus also has the feature of easy thermostating, evacuating and pressurizing (i.e., gas injection). In this work we present the method of surface tension measurement at the liquid–liquid interface based on capillary rise method, which is described exclusively here for the first time with some physical impact. The two immiscible liquids are brought into contact, forming interfaces both in pool and in the capillary (Fig. 1). From this new treatment, the capillary apparatus essentially provides the

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List of symbols

T	temperature
r	capillary radius
h	capillary height
a^2	capillary constant
g	gravity acceleration

Symbols

γ	surface tension
ρ	density
θ	contact angle
Δ	difference

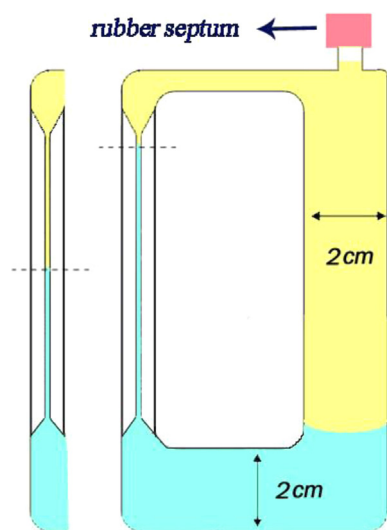


Fig. 1. Capillary apparatus for measurement of liquid–liquid interfacial tension. Capillary rise for system of high (right) and low (left) interfacial tension is demonstrated by dotted lines.

possibility of having two coexisting capillary actions integrated in one. The height between these two interfaces (when the apparatus is mounted precisely in vertical position parallel to the gravitational field) is proportional to the liquid–liquid interfacial tension. The capillary constant of concurrent capillary actions will be discussed. Interfacial measurement under coexisting capillary actions is more realistic (as occurs very often in nature and in industrial application) than the method which uses apparatus modeling, the phenomenon by separate capillary actions with separate measurement of heights [16,17]. These separate measured heights are then scaled by their corresponding liquid densities, which appear in the final expression for the calculation of interfacial tension. Accordingly, the results of present work can be used to conclude that each liquid–liquid system may have a characteristics capillary constant.

2. Experimental

2.1. Materials

Hexane, heptanes, octane, nonane, heptanol and hexanoic acid were used as received (see Table 1). Capillary apparatus was made out of borosilicate glass [12].

2.2. Method

The capillary apparatus was filled with deionized water and the liquid hydrocarbon. Two liquid–liquid interfaces were formed both in pool and in the capillary tube (see Fig. 1). The amount of

Table 1
Samples description.

Chemical name	Source	Purity mass%	Purification method
Hexane	Merck	99	none
Heptane	Merck	99	none
Octane	RDH	99	none
Nonane	Merck	99	none
Heptanol	Merck	99	none
Hexanoic acid	Merck	99	none

each liquid was preset such that the interface comes to equilibrium not more than about 2–3 mm below the top of the capillary tube, providing the condition of ignorable organic phase hydrostatic pressure above the meniscus level. After vigorous shaking, the system was allowed to equilibrate for 2 h and sealed by rubber septum at atmospheric pressure ($639.1\text{--}641.5 \pm 0.1$ mmHg). The capillary apparatus involving the liquids was mounted in an oil bath thermostat and the temperature was controlled within ± 0.5 K. The altitude (h) between bottom of liquid–liquid meniscus in the capillary and in the pool was measured digitally (within ± 0.01 mm) by setting up a digital cathometer [12]. The liquid density at each temperature was measured (where applied) by using a calibrated glass pycnometer and a microbalance (within ± 0.0001 g/cm³). Using the capillary radius (r) [12], and the difference in density of liquids ($\Delta\rho = \rho_{\text{water}} - \rho_{\text{oil}}$) [18–22], the interfacial tension (γ) was calculated by

$$2\gamma \cos \theta = g\Delta\rho r(h + r/3) \quad (1)$$

where g is the acceleration of gravity and θ is the contact angle. The approximation of $\theta \approx 0^\circ$ leads to results with high accuracies. This equation, which is customarily used to treat liquid–vapor surface, is now proved to be applicable to the liquid–liquid interface too.

3. Results and discussion

Alkanes, heptanol and hexanoic acid studied here are prototype systems and understanding of their behavior at interfaces with water are vital in engineering applications such as enhanced crude oil recovery. Other applications are food industry, cosmetic and drug delivery, to mention a few. All these systems are less dense than water, and extension to hydrocarbon systems with densities higher than water will be published latter.

Interfacial tensions measured in the range of $T = 298\text{--}343$ K for a set of liquid hydrocarbon–water systems are illustrated in Figs. 2 and 3. The measurements include water immiscible alkanes (hexane (C₆) to nonane (C₉)) with high interfacial tension as well as for slightly polar liquid hydrocarbon systems with low interfacial tension (heptanol and hexanoic acid). The latter systems are

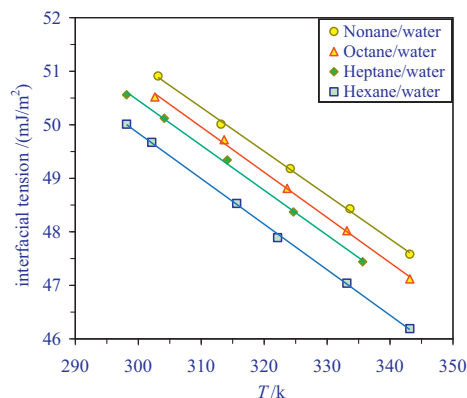


Fig. 2. Measured interfacial tension at alkane–water interface. Lines are trend lines.

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