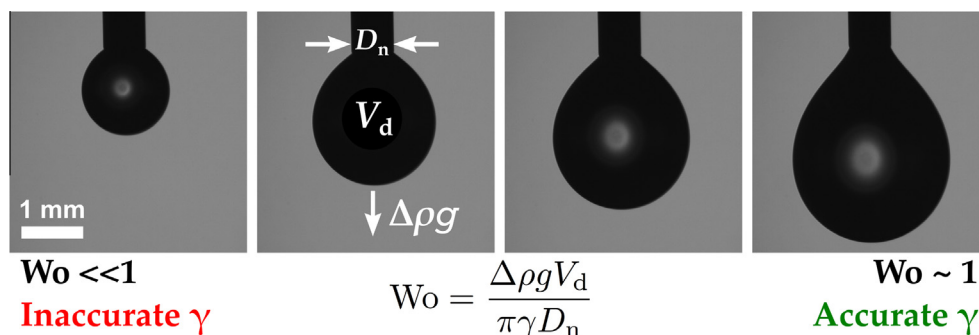


Feature Article

Measurement of surface and interfacial tension using pendant drop tensiometry

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GRAPHICAL ABSTRACT



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ABSTRACT

Pendant drop tensiometry offers a simple and elegant solution to determining surface and interfacial tension – a central parameter in many colloidal systems including emulsions, foams and wetting phenomena. The technique involves the acquisition of a silhouette of an axisymmetric fluid droplet, and iterative fitting of the Young–Laplace equation that balances gravitational deformation of the drop with the restorative interfacial tension. Since the advent of high-quality digital cameras and desktop computers, this process has been automated with high speed and precision. However, despite its beguiling simplicity, there are complications and limitations that accompany pendant drop tensiometry connected with both Bond number (the balance between interfacial tension and gravitational forces) and drop volume. Here, we discuss the process involved with going from a captured experimental image to a fitted interfacial tension value, highlighting pertinent features and limitations along the way. We introduce a new parameter, the Worthington number, Wo , to characterise the measurement precision. A fully functional, open-source acquisition and fitting software is provided to enable the reader to test and develop the technique further.

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Abbreviations: IFT, interfacial tension; Bo, Bond number; Wo , Worthington number; LMF-AO, Levenberg–Marquardt–Fletcher – automated optimisation; CMC, critical micelle concentration; CTAB, cetyltrimethylammonium bromide; SDS, sodium dodecylsulfate; RSD, relative standard deviation.

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Contents

1. Introduction	227
2. Experiment	228
3. Theory	229
4. Method overview	230
5. Results and limitations	231
5.1. Equilibrium and dynamic interfacial tension	231
5.2. Dynamic effects	231
5.3. Physical limitations	232
5.3.1. Low Bond number systems	232
5.3.2. Drop volume effects	232
6. Current research and future directions	235
7. Conclusion	235
8. Experimental	235
Supplementary Information	235
Acknowledgments	235
Appendix A. Fletcher's criteria for setting λ	236
Appendix B. The residual vector and Jacobian matrix	236
Appendix C. Calculating the arc lengths	237
References	237

1. Introduction

Interfacial tension is a phenomenon that, at the molecular level, results from the difference in energy between molecules at a fluid interface when compared to their bulk counterparts. It is equally correctly described as a measure of how much energy is required to make a unit area of interface between two immiscible liquids, thus taking units of Joules per square metre – equivalent to the more commonly used Newtons per metre. Interfacial tension is a concept of fundamental importance in colloid science, describing phenomena as diverse as the formation, shape and stability of liquid drops [1,2], the surface energy cost in forming an emulsion and the force applied by a capillary liquid bridge. Determination of the interfacial tension allows deductions to be made regarding the chemical composition of fluid interfaces and the adsorption and desorption of surface active solutes. Further, interfacial tension is the dominant force in microfluidic flows that are increasingly used in advanced liquid handling. It follows directly that precise measurement of interfacial tension is of critical importance to both science and industry. Many techniques have been proposed to measure interfacial tension (Fig. 1), and their features and qualities are described in detail by Drelich et al. [3]. Arguably the simplest (in terms of instrumentation), most robust, and most versatile of these methods is pendant drop tensiometry, where the measurement consists simply of a fluid droplet suspended from a needle.²

The ability to determine the interfacial tension from the shape of a pendant liquid drop deformed by gravity was first proposed over a century ago by Worthington [4–6], who evaluated the pressure drop across a portion of the curved interface of a drop hanging from a ground glass tube. In 1883, Bashforth and Adams [7] formed comprehensive numerical tables of approximate solutions to the axisymmetric Young–Laplace equation, and these are still in use today. Using an appropriate scaling, they showed that the shape of an axisymmetric pendant drop depended on a single dimensionless quantity, which they termed β , defined as $\beta \equiv \Delta\rho g R_0^2 / \gamma$, where the symbols represent density difference $\Delta\rho$, gravitational

acceleration g , drop dimension R_0 and interfacial tension γ . Bashforth and Adams [7] described β as an “abstract number”, when in fact it has significant physical meaning as a measure of the relative importance of gravitational forces to interfacial forces. This quantity was later termed the ‘Bond number’³ by Merrington and Richardson [9] in 1947, named after the British physicist Wilfrid Bond (1897–1937) who introduced the quantity in relation to the terminal velocity of drops and bubbles in 1928 [10].

If one can accurately quantify the Bond number from the drop shape, the interfacial tension readily follows from the known quantities of density, gravity and drop size. However, determining the Bond number for a given system proved difficult. In the 1940s, Andreas et al. [11] devised a simple approach for determining this quantity by taking the ratio of two easily measured experimental quantities: the maximum drop diameter d_e , and the drop diameter d_s measured at a distance d_e from the apex. The ratio $S = d_s/d_e$ could then be compared to tables to determine the Bond number, thereby obtaining the interfacial tension. Andreas et al. calculated these tables experimentally, however these were later improved through numerical integration of the Young–Laplace equation [12,13].

This approach offered a simple method for calculating the interfacial tension; however a large portion of data relating to the actual drop profile was discarded. In 1983, two transformational articles were published that developed computational routines to utilise all the available data, which greatly increased the precision of the method [14,15]. These methods compared the entire drop profile to the theoretical drop profile by considering the sum of the squared residuals between each experimental data point and the theoretical drop profile. While the methods share many similarities, the approach of Huh and Reed [15] implements an approximate expression compared to the exact expression presented in

² We use ‘droplet’ throughout for simplicity, although the technique can be equally applied to study gas bubbles in surrounding liquids, and the droplet may be pendant or sessile with respect to the needle, provided that the sign of the density difference is adjusted accordingly. Any combination of immiscible fluids and orientations can be handled theoretically, although some systems may be experimentally challenging to realise.

³ The Bond number is also known in the literature as the Eötvös number, named after the Hungarian physicist Lóránd Eötvös (1848–1919), who invented the reflection method for the measurement of surface tension and also discovered the linear relationship between the surface tension of a liquid and its temperature (known as the Eötvös law). Eötvös spent the first part of his career studying the effects of capillarity, and then moved on to make important observations on gravity. It is not evident that the Bond number appears in any of the publications of Eötvös. Indeed, it appears to have been named after Eötvös by Harmathy [8] in 1960 firstly to pay homage to a fellow Hungarian, and secondly as a subtle reference to the trajectory of Eötvös’ research career. Interestingly, and completely irrelevantly, after coining the Eötvös number Harmathy went on to invent a one-handed toilet paper dispenser.

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