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Historical perspective

The capillary bridge between two spheres: New closed-form equations in a two century old problem

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ABSTRACT

We discuss progress in obtaining explicit equations for the capillary force between nano and micron sized solid spheres. Early approaches to this two-century old problem adopted approximations to the geometry. With the toroidal approximation, the meridian profile is approximated by an arc, and the approach leads to the capillary force being dependent on the location at which the force is evaluated. The Derjaguin approximation further assumes that the meridian radius is orders of magnitude smaller than the azimuth radius. An explicit expression for the capillary force is obtained, but the equation is limited to sufficiently small liquid volumes and separation distances. Significant progress has been made in recent years in using numerical solutions to derive analytical expressions for capillary bridges. Early numerical investigation established that the maximum separation for stable capillary bridges before rupture scales to the cubic root of the liquid volume. We report new progress in using numerical solutions to obtain more accurate and more general closed-form expressions for capillary bridges. Simple explicit algebraic equations have been observed to fit the numerical results well, leading to a closed-form solution applicable to capillary bridges between equal and unequal spheres and with zero or finite solid–liquid contact angles. The newly derived closed-form equation is more accurate and reduces to the Derjaguin equation when the liquid volume (or half-filling angle) and separation distance are both sufficiently small.

Contents

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1. Introduction

When a quantity of liquid is introduced at the contact between two adjacent micro and nano sized solid particles, a capillary bridge, a form of meniscus, will be formed. Capillary bridges are ubiquitous in nature and engineering systems. They can exist at length scales as low as a few nanometres [1]. For granular materials, capillary bridges are responsible for a number of phenomena, including caking, agglomeration, ageing and mechanical strength [2–5]. In nature, the remarkable switchable adhesion of gecko lizards and insect species is also believed to originate from the capillary force [6,7]. Indeed, capillary forces are of importance in many applications such as nature-inspired switchable adhesive pads [8], surface cleaning [9,10], particle-stabilised foams and emulsions [11–13], capillary suspensions [14,15], fibre wetting [16] as well as the preparation of graphene films [17] and so on.

The capillary bridge has been a scientific topic of research for more than two centuries. The subject attracted interest from various backgrounds. The mathematical foundation of capillary action was laid by Thomas Young and Pierre Simon Marguis de Laplace at the beginning of the 18th century. The so-called Laplace–Young equation relates the capillary pressure difference across an interface of two static fluids to the shape and surface tension of the interface. Capillary bridges between solid spheres normally have the azimuth radius curved inwards and the meridian radius curved outwards. This is the conventional liquid bridge shape, sometimes loosely referred to as "toroidal". Capillary bridges between solid spheres can also have the two principal radii both curved inwards. This is the case when the solid–liquid contact angle is greater than π/2 and sometimes referred to as "convex", as in the case of a liquid drop [18]. In this case, the most favourable shape of the bridge is as part of a sphere as this is the most stable condition of minimised interfacial energy. The capillary force under static condition is essentially zero. Another special case is the cylindrical bridge that also has a simple analytical solution for the mean curvature and the capillary force is πrγ. This paper considers the exact forms of the conventional "toroidal" bridges, which are difficult to solve analytically.

Early theoretical studies considered approximations to the geometry. Fisher [19] approximated the capillary bridge as toroid, the so-called toroidal approximation. The approach leads to a capillary force which is dependent on the location at which the force is evaluated. Derjaguin derived an explicit analytical solution for the capillary force in which the radius of the meridian profile of the bridge is considered to be orders of magnitude smaller than the radius of the neck [20]. Recently, attempts were also reported to redevelop the Derjaguin equation, e.g. [21]. Both the toroidal and Derjaguin solutions are approximations to the geometry of the bridge and the capillary force. They are accurate only for liquid bridges of sufficiently small liquid volumes and separation distances [22,23]. Under more general conditions, both approaches lead to significant errors in the capillary force.

Experimental studies on capillary bridge forces were reported by MacFarlane and Tabor [24], Mason and Clark [25], and Erle et al [26]. Recent advances in AFM enabled more accurate measurement of the capillary force [22,27]. Comparisons of the measured capillary force with theoretical models confirmed the limitation of the Derjaguin equation and toroidal approximation [22,23].

Numerical studies were also reported on capillary bridges [22,23,28]. By solving the Laplace–Young equation numerically, it was observed that the maximum stable separation distance for capillary bridges between solid spheres scales to the cubic root of the liquid volume [23]. Beyond this critical separation distance, capillary bridges are not stable. This scaling relationship was found to apply for a wide range of conditions of solid–liquid contact angle, unequal sized spheres and flat surfaces [22]. Based on the numerical solution, Willett et al [22] also fitted an expression for calculating the capillary force as functions of liquid volume, contact angle and separation distance.

Although the subject of capillary bridges has attracted interest from many directions, it has remained difficult to calculate the capillary force under general conditions. Here, we report new progress in obtaining explicit analytical equations in this two-century old problem. We re-examine the numerical solution for capillary bridges and obtain, for the first time, simple fitted analytical expressions for the force, half-filling angle and radius of the neck of capillary bridges. First, by solving the Laplace–Young equation at zero separation numerically, we observe simple power-law equations for calculating the volume, capillary force and radius of the neck as a function of the half-filling angle. Second, we examine the numerical solution for capillary bridge geometry with increasing separation distance while keeping the volume of liquid constant. Simple analytical relationships are further observed for the force, the radius of the neck and half-filling angle as a function of the separation distance. Third, we examine capillary bridges between unequal sized spheres of finite solid–liquid contact angles. We show that for unequal sized spheres, the capillary force can be calculated from the expression for equal spheres of the same liquid volume, made dimensionless to the harmonic radius. Increasing the size of the large sphere results in an increase of the capillary force on the small sphere. For non-zero contact angle with solid spheres, a more accurate explicit expression for the capillary force at zero separation is obtained. At finite separation distance, the decrease of the capillary force for a finite solid liquid contact angle followed essentially the same trend for zero solid liquid contact angle. Finally, we present an explicit closed-form equation for the capillary force between equal and unequal sized spheres and for zero and finite solid–liquid contact angles. The equation is more accurate than the Derjaguin approximation and reduces to the Derjaguin equation when the half-filling angle and separation distance are both sufficiently small.

2. Problem definition

2.1. Laplace–Young equation

We consider the general problem of a capillary bridge between two unequal spheres with a finite solid liquid contact angle as shown in Fig. 1a. As usual, we introduce the harmonic radius

$$\frac{2}{r_{12}} = \frac{1}{r_1} + \frac{1}{r_2} \tag{1}$$

where r_1 and r_2 are the radii of the two spheres. We generally consider $r_1 \le r_2$. In particular, when $r_2 \to \infty$, the capillary bridge between a sphere and a flat surface is recovered.

The profile of the capillary bridge is described by the Laplace–Young equation, which relates the mean curvature of the bridge to the surface tension and pressure difference across the liquid-air interface. The equation can be written in the following dimensionless form

$$2H = \frac{\ddot{Y}}{\left(1+\dot{Y}^2\right)^{3/2}} - \frac{1}{Y\left(1+\dot{Y}^2\right)^{1/2}} \tag{2}$$

where $H = {}^{r_{12}\Delta p}/_{2\gamma}$ is the dimensionless mean curvature of the capillary bridge, Δp is the pressure difference across the liquid–air interface, and γ is the surface tension. $Y(X)$ denotes the azimuth radius or the axi-symmetrical profile of the capillary bridge in dimensionless form, i.e. $X = x/r_{12}$ and $Y = y/r_{12}$.

The boundary condition for the equation can be formulated at the solid–liquid contact lines with the two spheres as follows

$$Y_{c1} = R_1 \sin\varphi_1, \quad \dot{Y}_{c1} = -\cot(\varphi_1 + \theta) \quad \text{at} \quad X_1 = 0 \tag{3}$$