Colloid Polym Sci 277:993–996 (1999)

SHORT COMMUNICATION

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Direct measurement of the viscous force between two spherical particles trapped in a thin wetting film

Received: 19 March 1999
Accepted in revised form: 11 May 1999

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Abstract Here we present the first direct measurement of the viscous drag force between two spherical particles of millimeter size trapped in a thin wetting film. Each particle is constrained by the liquid/air interface and the solid substrate. The viscous force is counterbalanced by another known force, the attractive capillary immersion force between identical particles protruding from the film surface. The results of the measurements provide evidence for an increased hydrodynamic force due to a non-Stokesian resistance to the particle motion. Our findings can be applied to the self-assembly of colloidal particles in a two-dimensional array for coating and to the friction between small species and a solid.

Key words Drag force · Drag coefficient · Viscous flow · Capillary force · Friction coefficient

Introduction

The constrained motion of a particle trapped in a thin wetting film and the interaction between such particles in an array is of interest because of its relation to the following processes.

1. The formation of a 2D particle array on a solid substrate (for a recent review on dry 2D crystals of submicron- and micron-sized latex particles see Ref. [1]). The observed defects in the crystalline structure could result from increased resistance to the particle motion in the course of layer assembly [2]. The origin of this resistance for monolayer formation can be the restricted liquid flow near the solid substrate and near the three-phase contact line between a particle, liquid and air. If the particle is in contact with the solid, a frictional force, also unknown for such small species, will additionally take place.

2. Industrial coating [3]. The motion of fine particles composing the coating suspension should be rather restricted at the boundary between the liquid and the drying deposit.

3. Friction. The sliding between two solid surfaces in contact is the subject of frictional rheology and tribology applied to micro- and nanospecies [4].

The particles participating in the above processes are so tiny that their motion in the film is hardly sufficient for direct detection, for which reason the operating forces still remain unknown. To the best of our knowledge, we present here the first experimental study on this matter. It became possible to mimic the practical case using two spherical particles of millimeter size trapped in a film of corresponding thickness. The particles are brought toward each other by the attractive capillary immersion force [5]. Hydrodynamic resistance due to the film confinement and the particle–particle interaction opposes the particle motion. The unknown resistance force (viscous drag force) is found from the balance with the capillary force. There are important differences between our problem and the motion of a single particle floating on a free liquid interface [6]. These are the existence of a thin wetting film and the interaction between two particles trapped in this film. Due to these peculiarities the particle motion considered by us is additionally restricted.

Experimental

Two spherical glass beads of 1-mm diameter (radius $R = 0.05$ cm) were immersed in the wetting film formed in a circular cell

composed of a glass plate and a surrounding solid wall of 2-cm diameter (Fig. 1). The glass plate and beads were precleaned with chromic acid to assure good wetting by water. The liquids used were pure water or a water mixture with 40 wt% glycerol. The film thickness was controlled by a micro-syringe. The motion of the particles was observed using an inverted optical microscope from the substrate side and the positions of the particles were recorded over a period of time (Fig. 2). From the video frames one can estimate the center-to-center separation, $X(t)$, i.e. the motion law, and can reconstruct from here the unknown viscous drag force in balance with the known capillary force.

Calculation

Decreasing the film thickness below the particle diameter (Fig. 1) introduces an attractive force, the capillary immersion force $F_c = 2\pi\gamma q Q^2 K_1(qX)$ [5]. Here $q = \sqrt{\rho g/\gamma}$ is the reverse capillary length, where g is the gravitational acceleration, ρ is the liquid density and γ is the surface tension. K_1 is the modified Bessel function of the second kind. $Q = r_c \sin\psi$ is the so-called capillary charge, where ψ is the meniscus slope angle at the contact line of radius r_c. In fact, the equation for F_c is derived for two interacting cylinders of radii r_c immersed in the liquid. In the range of parameters of our experiment this equation does not differ so much from the more complicated expression for two interacting spheres [7]. Moreover, using the experimental values, $\psi \cong 90^\circ$ and $r_c \approx R$ ($Q \approx R$), and the fact that $qX < 1$, i.e. $K_1(qX) \approx 1/qX$, an approximate expression for the capillary force can be written [8]

$$F_c \cong 2\pi\gamma R^2/X \quad . \tag{1}$$

At small r_c (small sphere protrusion from the interface), the capillary force is counterbalanced by the force of friction between the particle and the substrate, $F_f = -\mu(mg + 2\pi\gamma Q)$. Here m is the particle mass and μ is the friction coefficient. The first term in the parentheses accounts for the normal load due to the particle weight. The second term is a contribution from the vertical projection of the surface tension. In our experiment the first term is negligible: it is about 20 times smaller than the second one at $m = 1.3 \times 10^{-3}$ g (glass

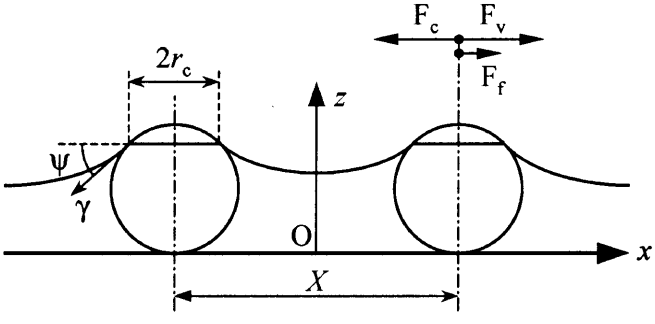

Fig. 1 Two particles trapped in a thin wetting film and moving in front of each other due to the applied forces

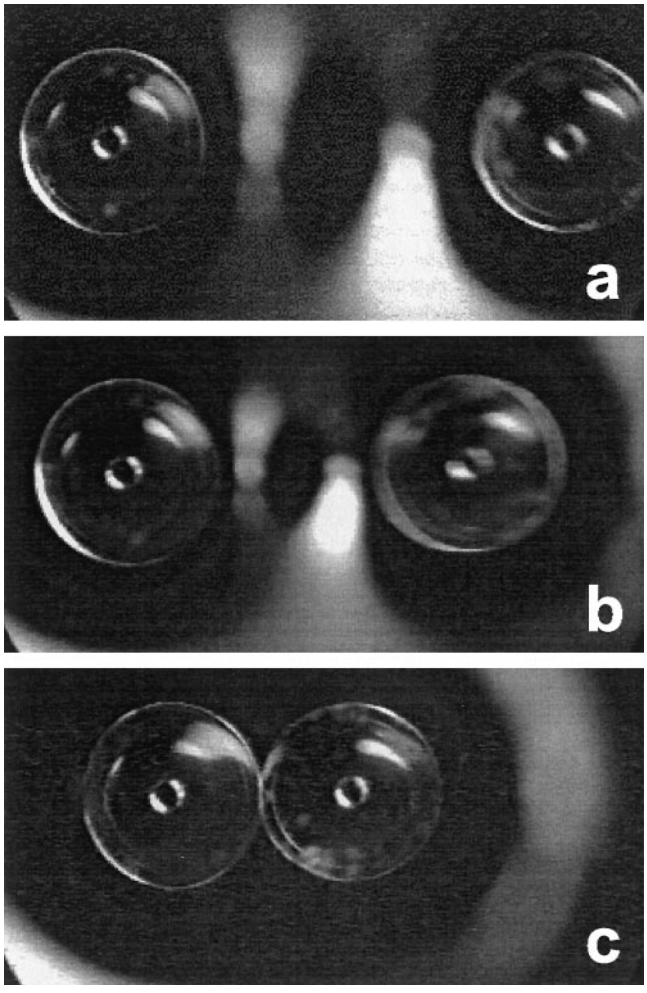

Fig. 2a–c Successive photographs of two glass beads of 1-mm diameter moving in a thin water/glycerol film. **a** Time $t = 0.1$ s from the beginning of motion at a separation distance $X = 2.5$ mm giving rise to a force $F_c = 4.52$ dyn. **b** $t = 0.27$ s, $X = 1.9$ mm and $F_c = 5.95$ dyn. **c** End of motion at $X = 2R = 1$ mm

density 2.5 g/cm^3) and $\gamma = 72$ dyn/cm. This simplifies the expression for the frictional force

$$F_f \cong 2\pi\gamma R\mu \quad . \tag{2}$$

At a certain film thickness the particles start moving toward each other. Equating the two forces, Eqs. (1) and (2), at the beginning of the motion one obtains the static friction coefficient $\mu_0 = R/X_0$, where X_0 is the initial separation distance. For a typical value of $X_0 \cong 2.8$ mm one obtains $\mu_0 \cong 0.18$. This is much smaller than the value for two glass surfaces sliding in dry air, $\mu_0 = 0.94$ [9] (the latter is larger than the respective dynamic friction coefficient $\mu = 0.4$). Obviously, the liquid serves as a lubricant diminishing the friction between the particle and the substrate.

During its motion a particle is also subjected to a viscous drag force

$$F_v = -\beta \dot{x} \ , \tag{3}$$

where β is the unknown drag coefficient, $\dot{x}$ is the particle velocity and $x = X/2$ is the abscissa.

In view of Eqs. (1)–(3), the equation of particle motion $-m\ddot{x} = F_c - F_v - F_f$ becomes

$$\frac{m}{2}\frac{d^2X}{dt^2} + \frac{\beta}{2}\frac{dX}{dt} + 2\pi\gamma R^2\frac{1}{X} - 2\pi\gamma R\mu = 0 \ . \tag{4}$$

Since there are parameters in Eq. (4), which cannot be controlled during our experiment, we simplify it by neglecting the small terms. To compare them with the leading term of expression for the capillary force, one should choose appropriate scales: τ for the time t and R for the length X (Fig. 2). Dividing Eq. (4) by $2\pi\gamma R$ introduces dimensionless coefficients for each term (unity for the capillary-force term). To have a viscous-drag-force term of the same order as the capillary-force term one should set to unity the respective coefficient $\beta/4\pi\gamma\tau$. Hence, for the characteristic time of motion one gets

$$\tau = \beta/4\pi\gamma \ . \tag{5}$$

For the parameters in Table 1, τ is 0.005 s for water and 0.03 s for water/glycerol (Fig. 3). At these values the coefficient of the inertia term in Eq. (4) is $m/4\pi\gamma\tau^2 =$ 0.057–0.0016 and can always be neglected. Similarly, one can also neglect the constant friction term since the dynamic friction coefficient, μ, is expected to be even smaller than the static one, $\mu_0 = 0.18$.

Hence, Eq. (4) takes the simple form $X\mathrm{d}X = -(R^2/\tau)\mathrm{d}t$, whose solution is

$$X/X_0 = \sqrt{1 - 2a^2 t/\tau} \ , \tag{6}$$

where $X(0) = X_0$ and $a = R/X_0$. Equation (6) was used to process the experimental data for $X(t)$ in Fig. 3 with very good accuracy.

Discussion

Our measurements with two spherical particles moving in a thin liquid film showed that the viscous drag force obeys the general law (Eq. 3) with a constant drag coefficient β. However, β calculated from the experimental data in Fig. 3 is substantially larger than the respective value for a particle moving in an infinite liquid given by Stokes, $\beta_S = 6\pi\eta R$, where η is the usual viscosity (Table 1). This additional viscous friction seems to have a different origin for a large or small distance between the particles.

At large X, it is most probably due to the restricted hydrodynamic flow in the narrow gaps between the particle and the solid substrate and between the particle and the liquid interface. These two cases are considered separately in the literature [10, 11], but with each of them pointing out an increased resistance to the particle motion. An additional restriction comes from the three-phase contact line [11] and the dynamic surface tension [12]. However, the complicated numerical simulations of the respective hydrodynamic problems [10–12] could not be directly implemented in our system.

At small X, the hydrodynamic interaction between the two particles should dominate. Since this mathematical problem is even more sophisticated than the motion of a single particle, here one can give only an idea for the increased resistance to flow. It is based on the approximate equation of Taylor for the hydrodynamic force between two spheres approaching each other in an infinite liquid (see e.g. Ref. [13]). This approximation assumes a variable drag coefficient, $\beta(X)$, which is valid at $X \ll 3R$. Indeed, there is a substantial increase in the calculated force at $X < 0.15$ cm as illustrated in Fig. 4.

Table 1 Experimental parameters

Liquid	ρ (g/cm^3)	q (cm^{-1})	η [14] (dyn s/cm^2)	β (g/s)	β_S (g/s)
Water	1.0	3.69	0.01	4.52	0.009
Water/glycerol	1.09[a]	3.86	0.032	27.14	0.03

[a] Calculated from the density of pure water and pure glycerol ($\rho = 1.26$ g/cm^3 [15])

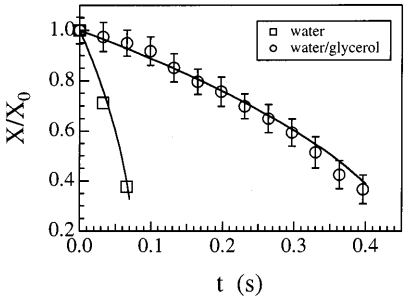

Fig. 3 The time-dependence of the distance between two particles, which are moving in films of different liquids. The *solid lines* are predicted using Eq. (6)

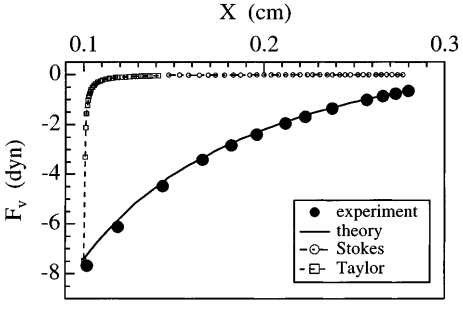

Fig. 4 Force of viscous interaction between the particles moving in a water/glycerol film. The *solid line* is calculated using Eq. (3) with β from Table 1. The rest of the calculations were made using Stokes' or Taylor's drag coefficients

To eliminate other possible effects, we also accounted for contribution from the inertia term. The numerical integration of Eq. (4) [2] does not appreciably affect the results presented here. Considering the friction term defined by Eq. (2) leads to another simple solution for $X(t)$:

$$t = (\tau/\mu a)\{-1 + X/X_0 + (a/\mu)\ln[(X/X_0 - a)/(\mu - a)]\}$$

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This equation represents a worse fit to the experimental data, especially at large t. The characteristic time is almost the same as the one obtained using Eq. (6), while the friction coefficient, $\mu = 0.15$, is indeed smaller than μ_0. Nevertheless, the corresponding frictional force, $F_f = 3.39$ dyn, is appreciable and is, therefore, subtracted from the viscous drag force plotted in Fig. 4.

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