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High-frequency viscoelastic measurements of fluids based on microcantilever sensing: New modeling and experimental issues

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ABSTRACT

In general, microrheology is carried out using active or passive particle-tracking techniques. In the present paper, a novel technique based on the out-of-plane bending vibrations of a microcantilever beam immersed into a liquid is proposed for microrheological property measurement. We propose to analytically link the damped beam motion with the rheological properties of the fluid in order to establish a dynamic rheogram which spans at least one decade of the kiloHertz frequency domain. The latest improvements in terms of both analytical modeling and experimental set-up are detailed, along with a complete explanation of the calculation method. Four rheograms of Newtonian and non-Newtonian liquids obtained from the frequency response of three immersed cantilevers of different geometries are presented.

1. Introduction

Exploring in situ microrheology is of fundamental interest and is widely used in many applications, such as viscoelastic monitoring of screen-printing ink and small-volume texture monitoring of commercial foams or gels. This paper presents a method for the improvement of viscoelastic property extraction for complex fluids using a microcantilever as a dynamic rheometer oscillating in the kiloHertz domain [1]. This frequency range is difficult to explore with classical dynamic rheometers due to inertial limitations. MEMS rheometers [2,3] (as well as other mechanical resonators [4–6]) are good candidates as high-frequency rheometers (for determining elastic, G', and viscous, G'', shear moduli vs. frequency) by characterizing complex fluids at the microscale by using very small amounts of liquid. In this context, dynamic rheology is performed by exploring the use of small-scale devices and short characteristic times; this yields viscoelastic property measurements on downscaled specimen sizes not obtainable with traditional instruments, thereby forming the basis of “microrheology”.

The concept of microrheology dates from 1922, when Freundlich and Seifriz used magnetic particles to study the elasticity of gelatin gels [7]. Over the last two decades, rapid developments have occurred in this field [8–11]. Renewed interest in microrheology has been increased by virtue of the technological innovations in colloidal engineering, light scattering, position sensitive detection and video microscopy. Currently there are two main categories of microrheology: active microrheology which involves the active manipulation of the probe particle by external forces (e.g., optical and magnetic tweezers and atomic force microscopy (AFM)) and passive microrheology which relies on the Brownian motion (thermal fluctuation) of the probe particles (e.g. video (VPT) and laser (LPT) particle tracking). Diffusing wave spectroscopy (DWS) is also a common microrheological technique widely used to characterize complex mixtures. As with other optical methods, DWS is based on the mean square displacement measurement which characterizes Brownian motion and permits the calculation of an equivalent microscopic and local shear modulus.

Microcantilevers vibrating in fluids are strongly influenced by the surrounding fluid properties [12]; thus, the out-of-plane transverse deflection response may be mathematically linked to the rheological properties of the surrounding liquid [13]. Based on previous work [14,15], we present here a considerably enhanced analytical model in order to (1) identify highly sensitive parameters in the determination method; (2) eliminate a misleading simplifying hypothesis in the solution; and (3) produce accurate rheograms in the case of non-Newtonian fluids. We also discuss the accuracy of the proposed method in order to examine the validity of the high-frequency rheograms produced and to demonstrate proof of concept. Using this new modeling approach, microcantilevers

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can probe the high-frequency viscoelastic behavior of fluids, especially the viscous characteristics, over a large frequency bandwidth around the resonant frequency. One of the main advantages of our technique is a faster response time than macrorheometers or other microrheological techniques.

2. Modeling of microcantilever behavior in complex fluids

The Euler–Bernoulli differential equation (Eq. (1)) governing the out-of-plane bending vibration of a microcantilever in a liquid medium is used to model the sensing situation:

$$EI\frac{\partial^4 w(x,\omega)}{\partial x^4} + j\omega g_1(x,\omega)w(x,\omega) - \omega^2(g_2(x,\omega)+m_L)w(x,\omega) = F_{\mathrm{ext}}(\omega)\delta(x-L) \quad (1)$$

where E is the Young Modulus, I is the second moment of area of the rectangular cross section, $w(x,\omega)$ is the vibration amplitude at position x along the length axis, depending on the radial frequency ω of the vibration, L is the beam length and the position of the free-end of the cantilever, m_L is the cantilever mass per unit length, $F_{\mathrm{ext}}(\omega)$ is the applied force amplitude at the cantilever free-end, δ is the Dirac function and g_1 and g_2 are, respectively, functions of the dissipative and inertial part of the hydrodynamic force per unit length, F_{hydro}, exerted by the fluid on the cantilever.

Assuming that g_1 and g_2 are independent of x [16], an exact analytical solution of Eq. (1) exists for the free-end cantilever deflection, $w(L,\omega)$. This solution gives a frequency-dependent response partially influenced by the rheological properties of the fluid:

$$w(L,\omega) = \frac{3F_{\mathrm{ext}}}{k_0A^3L^3}\,\frac{\sin h(AL)\cos(AL) - \cos h(AL)\sin(AL)}{1+\cos h(AL)\cos(AL)} \quad (2)$$

where

$$A^4 = \frac{\omega^2}{EI}\left(m_L + g_2(\omega) - \frac{jg_1(\omega)}{\omega}\right), \quad (3)$$

and k_0 is the cantilever stiffness. In Eqs. (2) and (3), the hydrodynamic properties of the surrounding fluid are contained in the terms g_1 and g_2 that are related to the imaginary and real parts of F_{hydro}. However, Eqs. (2) and (3) do not allow the inverse problem to be solved, meaning that g_1 and g_2 cannot be expressed explicitly as functions of the different parameters and measurement data (ω, $w(L,\omega)$, E, I, L, k_0, F_{ext}, m_L). Nevertheless, these analytical equations may be used to fit the experimental mechanical spectra of microcantilevers measured in known fluids. To do so, final rheological properties must be linked to g_1 and g_2, respectively, depending on the viscous and inertial parts of the hydrodynamic force:

$$F_{\mathrm{hydro}} = \omega^2 g_2(\omega)w(x,\omega) - j\omega g_1(\omega)w(x,\omega) \quad (4)$$

The differential equation solution supposes that the viscosity η (hidden in the terms g_1 and g_2) is a real number. Viscoelastic characterization probed by microcantilevers is based on the assumption that the fluid viscosity can have both a real and an imaginary part. Consequently, the constant real term η corresponding to viscosity is in this case replaced by a complex number η^* accounting for both the viscous and elastic behavior of viscoelastic fluids. (This is classic within the context of rheological modeling [17,18].) This notation is equivalent to considering a complex shear modulus G^* for a viscoelastic fluid instead of an imaginary shear modulus (see details in Appendix A). Moreover, a fluid is rheologically defined by a mass density ρ_f, and values G' and G'' which are, respectively, the elastic (real) part and the viscous (imaginary) part of the complex shear modulus G^* at each frequency. A Newtonian fluid such as water has no elasticity ($G' = 0$ Pa) and a viscosity with a frequency-independent real part ($\eta = 10^{-3}$ Pa s = 1 cP, in the case of water). This defines a viscous modulus G'' that linearly increases with frequency ($G'' = \eta\omega$). A fluid is considered as complex if it does not satisfy those two conditions.

In the case of thin rectangular microcantilevers, anexact equation of the hydrodynamic force has been developed, first by Sader [16] and then fitted by Maali et al. [19] in order to express separately the real and imaginary parts of the hydrodynamic force in terms of four constant coefficients (a_1, a_2, b_1, b_2) whose values are dependent on the Reynolds number range. The accuracy of the fitting expressions are very good, provided that the appropriate Reynolds number range restriction has been verified. Based on those works, analytical equations of g_1 and g_2 as functions of the shear modulus were established [20]. Those expressions are recalled in Eqs. (5)–(7) (due to physical considerations detailed in Appendix A, $a_1 = 1$ and $a_2 = b_1$):

$$g_1 = DG'' + B\sqrt{\sqrt{G'^2+G''^2} - G'} \quad (5)$$

$$g_2 = \frac{C + DG' + B\sqrt{\sqrt{G'^2+G''^2}+G'}}{\omega} \quad (6)$$

where

$$B = \frac{\pi b_1}{2\sqrt{2}}b\sqrt{\rho_f} \quad C = \frac{\pi}{4}\rho_f b^2\omega \quad D = \frac{\pi b_2}{2\omega} \quad (7)$$

and b is the cantilever width.

Using Eqs. (2)–(7) and considering a fluid with a known rheological behavior and a microcantilever with known geometry and material, it is possible to calculate the free-end deflection of the microstructure at each frequency and to compare it with experimental measurements. This also means that we are able to simulate experimental data.

3. Inverse problem: method for determination of rheological fluid properties

3.1. From g_1, g_2 to G', G''

With the expression of the hydrodynamic force from Sader [16] it is impossible to dissociate analytically the real part from the imaginary part, whereas it is possible with the approximation developed by Maali et al. [19]. Then, based on Eqs. (5)–(7), the elastic and viscous shear moduli can be expressed. (Details are presented in Appendix B.) The analytical expressions are

$$G'' = \frac{g_1}{D} - \frac{B}{D\sqrt{2D}}\sqrt{\sqrt{\left(\frac{B^2}{D}+2(\omega g_2 - C)\right)^2 + 4g_1^2} - \frac{B^2}{D} - 2(\omega g_2 - C)} \quad (8)$$

$$G' = \frac{1}{D}\left(\omega g_2 - C - \frac{B^2G''}{g_1 - DG''}\right) \quad (9)$$

The calculation of G' and G'' for any radial frequency ω can then be performed provided that g_1 and g_2 can be estimated with enough accuracy from the spectrum measurements. The calculation of the terms g_1 and g_2 as a function of both the amplitude and phase of the cantilever free-end deflection measurements is the purpose of the next section.

3.2. Improvement of the inverse problem: from deflection to g_1, g_2

The calculation of g_1 and g_2 from the amplitude and phase of the cantilever free-end deflection has been previously done by Belmiloud et al. [13]. At that time, the authors have considered that the analytical solution (Eq. (2)) of the differential equation (Eq. (1)) can be approximated by a second order mechanical low-pass filter