

Optical Coherence Microrheology using Spherical and Rod-like Microrheological Probes

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Abstract: Processing of light scattering signals from spherical and rod-like probes in optical coherence tomography provides frequency-dependent viscoelasticity of complex fluids, constituting a novel optical coherence microrheological tool.

OCIS codes: (110.4500) Optical coherence tomography; (290.1990) Diffusion; (110.6150) Speckle imaging; (070.4550) Correlators; (290.5850) Scattering, particles; (290.5855) Scattering, polarization

1. Introduction

In the field of microrheology, there has been considerable interest in techniques that resolve thermal diffusion of probes within a heterogeneous sample. Passive microrheological techniques quantify the Brownian diffusion of the probes whereas active microrheological techniques quantify the diffusion of the probes in response to an external force. The generalized Stokes-Einstein relation then relates diffusion measured by these techniques to the linear viscoelastic properties of the medium, provided that the probe is inert and the medium behaves as a near-equilibrium, homogeneous, isotropic and incompressible continuum [1]. Current passive microrheological techniques rely mostly on single-particle tracking via microscopy, or light scattering-based techniques such as dynamic light scattering (DLS), and diffusing wave spectroscopy (DWS). Single-particle tracking using microscopy provides excellent spatial resolution but limited depth penetration, and is also tedious since individual particles need to be tracked. Light scattering-based techniques such as DLS and DWS provide ensemble-averaged probe response and offer rapid quantification of viscoelastic properties. However, DLS and DWS lack the ability to provide local resolution of viscoelastic properties in a heterogeneous media. Optical Coherence Tomography (OCT), also based on light scattering, due to its ability to path-length resolve dynamic light scattering [2,3], can potentially mitigate these limitations by providing local microrheological properties in turbid media, dubbed “optical coherence microrheology” (OCMR) [4].

2. Methods

We apply OCMR using thermally diffusing spherical and rod-like probes to quantify the viscoelasticity of Newtonian and complex fluids. For spherical probes, the microrheological property is derived from the thermally-driven mean-squared displacement (MSD) of the probes, whereas for rod-like probes, these properties are derived from the thermally-driven mean-squared angular displacement (MSAD) of the probes, as discussed below.

a. Translational diffusion of spherical probes

Spherical probes diffused in the medium undergo Brownian motion, and their translational diffusion is indirectly evident from the intensity fluctuation of backscattered light. The MSD, $\langle \Delta r^2(t) \rangle$, of these spherical probes can be related to first order field autocorrelation, $g^1(t)$, via $\langle \Delta r^2(t) \rangle = -(6/q^2) \log_e[g^1(t)]$, where q is the scattering vector. The frequency-dependent MSD, $\langle \Delta r^2(\omega) \rangle$, of the spherical probe (radius a) undergoing thermal ($k_B T$) motion can then be related to the frequency-dependent complex shear modulus, $G^*(\omega)$ using the generalized Stokes-Einstein relation (Eq. 1) [5], the real and imaginary parts of which are elastic modulus, $G'(\omega)$, and viscous modulus, $G''(\omega)$, respectively.

$$G^*(\omega) = \frac{k_B T}{i\pi a \omega \langle \Delta r^2(\omega) \rangle} \quad (1)$$

Experimentally, an OCT axial scan can be repeated over sufficiently long time, and the autocorrelation can be computed at each depth. The viscoelastic properties can then be evaluated using the formalism described above. Figure 1 shows the elastic and viscous modulus of polyethylene glycol (PEO, molecular weight 10^6 gms/mol) in water measured using 120 nm microspheres as probes with OCMR. As expected, 2.5% PEO has overall higher modulus than 1.25% PEO over most of the frequency range. Moreover, at low frequency (long timescale) the viscous modulus, $G''(\omega)$, dominates, whereas at high frequency (short timescale) the elastic modulus, $G'(\omega)$, is seen to dominate. The 2.5% PEO sample is observed to make the elastic to viscous transition at lower frequency compared to the 1.25% PEO sample, which suggests that the sample with higher concentration of PEO makes a delayed elastic-to-viscous transition, as expected.

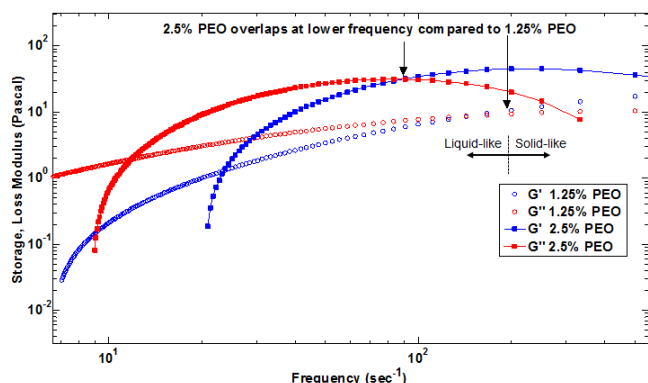


Figure 1. Elastic and viscous modulus of 1.25% PEO and 2.5% PEO measured with OCMR using 120 nm microspheres as probes.

orientation of its long axis [8]. Exploiting the polarization-dependent scattering from GNRs, it was recently shown that GNRs can be used as rotational diffusion sensors and that OCT allows for the rotational diffusion to be depth-resolved [9]. The Brownian rotation of GNRs results in a fluctuating cross-polarized scattered electric field. As described previously [9], the cross-polarized (HV) OCT signal is related to the first-order field autocorrelation of the field, $g_{HV}^1(t)$. The MSAD, $\langle \Delta\theta^2(t) \rangle$, of the GNRs can be related to the first order field autocorrelation via $\langle \Delta\theta^2(t) \rangle = -(2/3) \log_e [g_{HV}^1(t)]$. The frequency-dependent MSAD, $\langle \Delta\theta^2(\omega) \rangle$, of GNRs (length L , width d) undergoing thermal ($k_B T$) motion can then be related to the frequency-dependent complex shear modulus, $G^*(\omega)$ using the generalized Stokes-Einstein relation as follows:

$$G^*(\omega) = \frac{f(L,d)k_B T}{i\pi L^3 \omega \langle \Delta r^2(\omega) \rangle} \quad (2)$$

where $f(L,d)$ is the geometric factor in the Stokes frictional term for rod-like cylinders [10].

Rotational diffusion-based OCMR using GNRs as probes depicts anomalous crossover between $G'(\omega)$ and $G''(\omega)$ compared to microspheres, which is the topic of current investigation. In conclusion, the comparison between spherical and rod-like probes shows advantages and disadvantages of each for microrheological applications.

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b. Rotational diffusion of rod-like probes

Rod-like probes present an appealing alternative to traditional spherical probes due to the ability to monitor their rotational motion resulting from their shape anisotropy. Gold nanorods (GNRs) are a class of such probes, and are particularly appealing as they depict surface plasmon resonance, which allows the scattering response to be tuned based on their aspect ratio [6]. Thus, they can be easily tuned to the near-infrared region where optical absorption by the tissue is minimal. In addition, due to the surface plasmon resonance, GNRs depict strong optical scattering cross-sections despite their small size [7]; this small size is more amenable to diffusion in tissue imaging applications. More importantly, light scattering from a plasmon resonant GNR is strongly polarized along the