Mean squared displacement of a probe particle in a viscoelastic fluid

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We consider the characteristic behavior (mainly mean squared displacements) of a probe particle in several types of viscoelastic fluids. In this study, the probe particle undergoing Brownian motion in viscoelastic fluids is assumed to be described by the generalized Langevin equation (GLE) [1]:

$$m\frac{d\mathbf{v}(t)}{dt} = -\int_0^t \zeta(t-t')\mathbf{v}(t')dt' + \mathbf{f}_{\mathrm{R}}(t), \quad (1)$$

where m is the mass of a probe particle and $\mathbf{v}(t)$ is its velocity. Viscoelastic fluids can store energy and have a finite relaxation time to dissipate it, so that the embedded probe particle confined initially in elastic surroundings undergoes finally dissipative motion. This feature can be described by a memory kernel $\zeta(t)$ in the GLE [1]. The random Gaussian force $\mathbf{f}_R(t)$ and the time inhomogeneous friction force $\zeta(t)$ are related each other by the fluctuation-dissipation theorem: $\langle \mathbf{f}_{\mathrm{R}}(t)\cdot\mathbf{f}_{\mathrm{R}}(t')\rangle = k_{\mathrm{B}}T\zeta(t-t')$. Here, k_{B} is the Boltzmann constant and T is the temperature of the fluid, and the bracket indicates the ensemble average. In order to investigate the dynamics of a probe particle described by Eq.(1), we have to determine time dependency of unknown memory kernel $\zeta(t)$. Here we adopt the phenomenological procedure to obtain an explicit form of memory kernel, in other words, the memory kernel is determined from the bulk complex modulus of a viscoelastic fluid.

In this study we first reconsider the case of a single-relaxation time Maxwell fluid, that is an ideal model fluid in rheology. Many years ago several authors [2] have shown from non-Newtonian fluid mechanics that the memory kernel for a particle embedded in a single-relaxation time Maxwell fluid should take a simple exponential form. Since a class of polymer solutions can be classified as a two-fluid system composed of a continuum matrix and a solvent, we include the effect of solvent with the Newtonian viscosity as well (Jeffreys fluid model). The memory kernel for a probe particle is consequently expressed by

$$\zeta(t) = \zeta_0\delta(t) + (\zeta_{\mathrm{p}}/\tau)\exp(-t/\tau), \quad (2)$$

for a single-relaxation time Jeffreys fluid. Here, ζ_0, ζ_{p} represent friction coefficients of a Newtonian solvent and a polymer matrix having Maxwellian viscoelasticity, respectively. τ is a characteristic relaxation time. The dynamics of a Brownian particle in polymer solutions, which have the bulk complex modulus of the single-relaxation time Jeffreys fluid, is well described by the GLE with the memory kernel given by (2) [3, 4] although real materials showing perfect single-relaxation time Maxwell (as well as Jeffreys) behavior are very few. It has been also shown [3] that an exponential form of memory function correctly leads to the complex modulus (or complex viscosity) of a single-relaxation time Maxwell fluid by using the extended Stokes' friction law introduced by Mason and Weitz [1]. By combining these facts, it can be seen that the complex modulus and the memory kernel are in one-to-one correspondence for a single-relaxation time Maxwell fluid. Therefore we can apply a relaxation-time spectrum of a rheological material, which uses conventionally a Maxwell element as a basis, to a memory kernel for a probe particle.

As a particular example, we use a relaxation-time spectrum having the same form as the chi-squared distribution in order to explore how the whole time behavior of characteristic quantities of a Brownian particle is affected by continuous modification of the memory kernel from an exponential form by studying, e.g., mean squared displacements of probe particle. We also discuss our results in a possible connection with a couple of topics: the probe particle size effect, and the existence of anomalous diffusion.

References

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