F-SHELL BLOB MODEL INSPIRED BY SIMULATIONS OF TETHERED POLYMERS

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dynamics simulations of a tethered polymer in uniform flow show that polymers are neither impenetrable nor free-draining. According to these results a f-shell blob model has been introduced which contains the well known non-draining blob model as well as the free-draining blob model as limiting cases. In addition, this model describes the transition from a nearly impenetrable polymer coil at small flow velocities to a nearly free-draining uncoiled polymer at large flow velocities.

1 Introduction

The dynamics of polymers in a solvent and its interaction with the surrounding flow form the basis of non-Newtonian behavior of polymer solutions[1,2]. One of the spectacular non-Newtonian phenomena in polymer solutions is turbulent drag reduction. A thorough understanding of this and other viscoelastic phenomena is still not available and one of the reasons are that most of the experimental techniques could only detect the spatially and temporally averaged behavior. Only in recent experiments using fluorescence microscopy a considerable step forward has been done, because fluorescence microscopy allows a direct visualization of the stretching of tethered polymers in flow held by optical tweezers as well as the deformation of free polymers in elongational or pipe flow[3]. Theoretically three approaches have been used to calculate the shape of tethered polymers in flow. The most coarse grained approach is given by the so-called blob model, where the deformed polymer is replaced by a chain of spherical blobs in the so-called trumpet regime[5]. A more microscopic description is provided by the bead-spring model, which has been studied in Brownian dynamics simulations[6,7,8]. The dumbbell-model can be considered as a limiting case of the bead-spring model, where the polymer is composed of two beads only, which are connected by a nonlinear spring.

One appeal of the blob- and the dumbbell-model is their simple structure allowing for several situations analytical solutions. For instance for the model with impenetrable blobs a simple power law, $L(v) \sim v^2$, for the elongation of a tethered polymer as a function of the velocity of the uniform flow has been derived[5].

However, the validity of a number of assumptions of the dumbbell and the blob model has not been investigated. Recent Brownian dynamics simulations[6,7,8] provide besides the polymer induced deformation also the perturbed flow field and they show that a flow-deformed polymer is not completely impenetrable. Furthermore, a closer look on the numerical data show that the effects of the hydrodynamic interaction (HI) between different parts of the polymer vary along the polymer. Since both effects are not taken into account by former blob models this observation gives rise to a generalization of former models to the so-called f-shell blob model, where each blob is composed of an impentrable inner sphere and a free-draining outer shell as indicated in fig. 1.

2 f-shell blob model

The partial penetration of flow into the polymer coil may be taken into account in blob models by assuming for each blob a free-draining outer shell of thickness d and an impenetrable inner sphere, cf. fig. 1. The previously discussed non-draining and the free-draining blob models are limiting cases of this f-shell blob model. According to the Pincus rule[9], the radius R_k of a blob is determined by a balance between the total force F_k acting on it and the thermal agitation, $R_k = k_B T / F_k$.

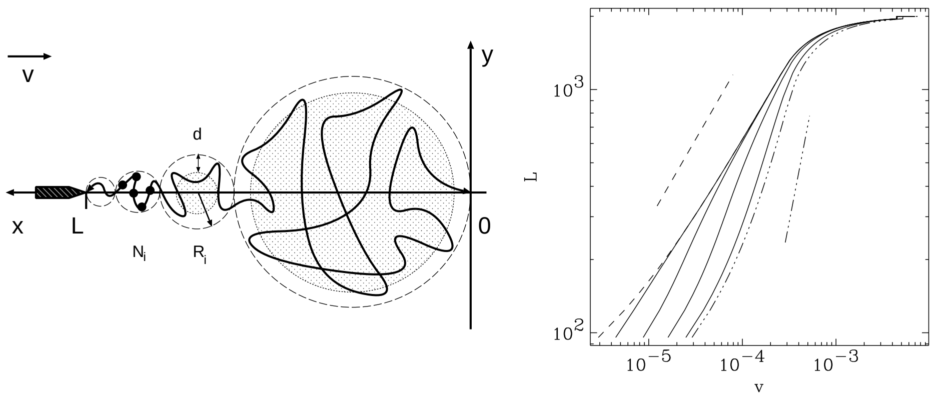

Figure 1. a) Sketch of a polymer tethered at one end and exposed to a uniform flow field with velocity $\vec{v}$. In the blob picture the deformed polymer with N segments (beads) is approximated by a sequence of spheres (blobs) with radius R_i containing N_i segments. In the f-shell blob model each blob consists of a free-draining outer shell of thickness d and an impenetrable inner sphere (shaded). b) Elongation $L(v)$ of the polymer calculated for the f-shell blob model with various values of the penetration depth $d = 1.0, 5.0, 15.0, 40.0$ from right to left. The dashed-dotted curve is for the non-draining limit $d = 0$ while the dashed line corresponds to the free-draining limit $d = R_F$. The straight lines are the power laws obtained for analytical approximations. Parameters are $N = 2000$ and $b = 1.0$ with a corresponding Flory radius $R_F = bN^{3/5} = 95.6$.

The radius of the non-draining inner sphere is $R_k^{\rm non} = R_k - d$. Using the Flory scaling $R_k = bN_k^\nu$ the number of segments $N_k = (R_k/b)^{1/\nu}$ within the k-th blob may be calculated as well as the number $N_k^{\rm non} = [(R_k - d)/b]^{1/\nu}$ in the non-draining part. The outer free-draining shell then contains $N_k^{\rm free} = N_k - N_k^{\rm non}$ segments. The total force on the k-th blob F_k is the sum of drag forces exerted by the $(k-1)$ blobs counted from the free end plus the local Stokes friction acting on the k-th blob caused by both the non-draining inner sphere and by the free-draining shell, *i.e.* $F_k = F_{k-1} + 6\pi\eta v R_k^{\rm non} + 6\pi\eta a v N_k^{\rm free}$. Here a is an effective hydrodynamic bead radius. This together provides the recursion relation for the force F_k

$$F_k = F_{k-1} + 6\pi\eta v \left(\frac{k_B T}{F_k} - d\right) + \frac{6\pi\eta v a}{b^{1/\nu}} \left(\left(\frac{k_B T}{F_k}\right)^{1/\nu} - \left(\frac{k_B T}{F_k} - d\right)^{1/\nu} \right), \quad (1)$$

with the boundary condition $F_0 = 0$. The radii R_k are determined via the Pincus rule, i.e. $R_k = k_B T / F_k$ and their sum $\sum_k R_k$ is the overall extension $L(v)$ of a polymer as a function of the velocity, cf. fig. 1. The bead radius a has been chosen such that the polymer elongation starts already at smaller velocities in the free-draining limit (dashed line in fig. 1) than for the non-draining limit (dashed-dotted line), similar as in our simulations. The intermediate curves in fig. 1 (solid lines) are for different values of the penetration depth d.

In the non-draining and the free-draining limit eq. (1) may be solved analytically with some approximations[5,7]. Then one obtains simple power laws for the polymer extension as $L(v) \sim v^2$ in the non-draining limit[5] and $L(v) \sim v^{2/3}$ in the free-draining limit[7], when $\nu = 3/5$ is chosen in the Flory relation. However, a numerical calculation of $L(v)$ as shown in fig. 1 by the dashed and the dashed-dotted curves, has more structure than the simple power laws obtained by the approximate calculation, cf. straight lines in fig. 1. As mentioned above, the power laws may be considered as upper limits for the slope $L(v)$ for very long chains. A finite penetration depth d gives a smaller slope $l(V)$ than for $d = 0$, as shown in fig. 1. According to both effects one cannot expect any scaling regime with simple power laws for $L(v)$ for molecules having less than $N \sim 1500 - 2000$ Kuhn segments which are used in experiments.

3 Conclusions

Motivated by simulations of tethered polymers in uniform flows[7,8] we introduced the f-shell blob model, which includes both the finite penetration of the external flow into the polymer and the gradual switching-off of HI along the chain of blobs. The calculation of $L(v)$ within this model shows more structure than the scaling relations obtained by analytical approximations for the free- and the non-draining case, which are upper bounds in the limit of a large number of segments N. Furthermore, the scaling laws are restricted to a finite velocity range.

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