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Preface

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MOBILITY OF HIGHLY DEFORMABLE NANOFILAMENTS

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Summary Mobility of hydrogel nanofilaments suspended in liquid is investigated to gain basic knowledge on hydrodynamic interactions biased by Brownian fluctuations. Typical for long macromolecules effects like spontaneous conformational changes and cross-flow migration are observed and evaluated. The collected experimental data can be used to validate assumptions present in numerical models describing intercellular transport of long biomolecules.

INTRODUCTION

Cross-flow migration of long micro objects is one of the fundamental mechanisms of lab-on-chip bio-processors. Moreover, behavior of long macromolecules is important in a variety of biological processes responsible for transport, aggregation and structure formation at the cellular level. Mobility of long deformable objects suspended in liquid depends on large number of parameters. Some of them like local flexibility, 3-D orientation, coiling - uncoiling are essential for resulting dynamics, however difficult to be accurately incorporated in practical models. Most common hydrodynamic models are based on several variants of classical *worm-like chain* (WLC) approach, describing flexible object as systems of interconnected identical spheres [1]. Its flexibility is usually defined by two parameters of stretching and bending. Experimental validation of such models using polymer solutions or DNA [2] is difficult and mainly limited to very general observations. In the following we propose more robust experimental study, based on optical observations of hydrogel filaments. Their characteristic size (dia. ~100 nm) allows for accurate evaluation of shape and orientation, but still remains small enough to reproduce characteristics of the flow regime influenced by Brownian fluctuations.

EXPERIMENT

We have developed new method allowing for fabrication of highly deformable microscopic filaments with typical diameter of 100 nm and contour length ranging from single micrometers to millimeters [3]. The nanofilaments are obtain by extracting hydrogel core of electrospun polymeric nanofibres. The composition of materials used allows for *in flight* modification of the hydrogel polymerization, tuning its mechanical properties to desired values. The surface topography of filaments is obtained by AFM equipped with a closed liquid cell and a rectangular silicon cantilever. Despite relatively large diameters, high deformability of our hydrogel filaments permits to obtain objects with mechanical properties resembling that of long biomolecules (comp. Table 1).

Table 1. Typical experimental parameters: mean flow velocity (Vm), contour length (L), radius (R), persistence length (Lp), and Young modulus (E) of the analyzed filament. Flow Reynolds number (Re) is based on the channel width (200 μm).

No	Vm [μm/s]	L [μm]	R [nm]	Re	Lp [μm]	E [kPa]
1	59.02	71.91	81	1.51E-02	17.62	2.17
2	77.78	72.87	81	1.98E-02	12.38	1.53
3	39.65	34.49	45	1.01E-02	3.44	4.45
4	68.87	54.31	45	1.76E-02	7.51	9.72

The dilute suspension of hydrogel nanofilaments is pumped through the 500 μm long PDMS microchannel and imaged by a fluorescent microscope coupled with high-gain EM-CCD camera. The experiment is based on observation of position and instantaneous shape of nanofilament conveyed by steady or pulsating flow. Some hundreds of long sequences of filament images are processed to extract their shape with specially designed Matlab software. The velocity profile in the channel is obtained from small amount of fluorescent tracers. For each analyzed filament its flexibility is determined by calculating Young modulus. For this purpose we analyze their Brownian shape fluctuations, and then used cosine correlation method to determine the persistence length [3]. Evaluation of geometry for three dimensional objects using 2-D projections obtained from the microscope is overwhelmed by unavoidable inaccuracy, it additionally complicates if parts of the filament tend to overlap. Hence, only clearly distinguishable objects with in plane shape deformations are taken into account. For this purpose constancy of apparent contour length of visualized filament is used as a control parameter. Channel depth is 60 μm. Hence, additional efforts (e.g. flow focusing) are undertaken to keep filaments far from the upper and bottom wall of the channel.

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RESULTS

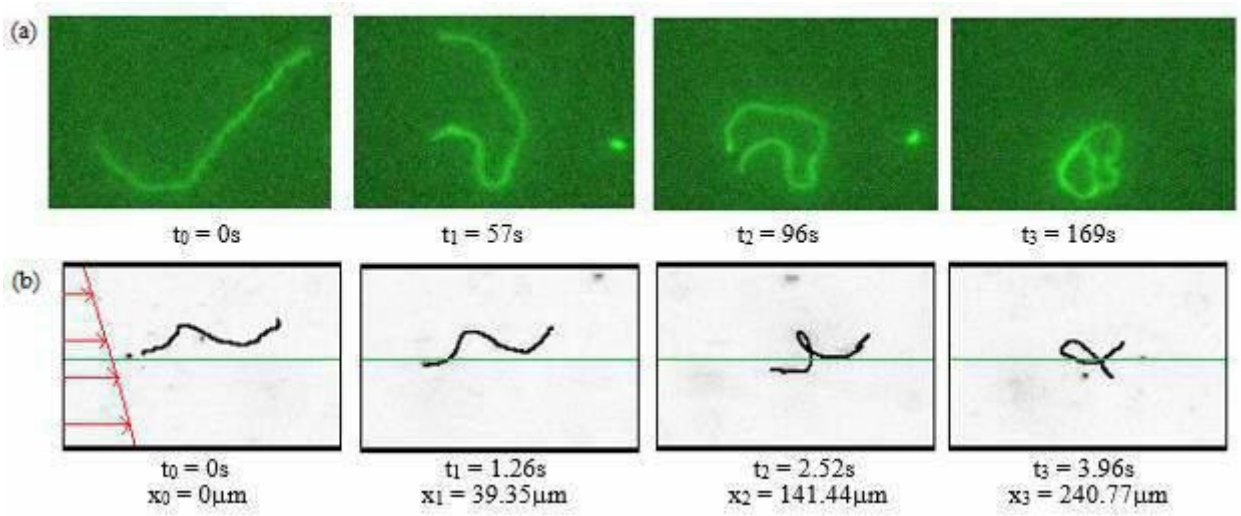

Figure 1. Sequence of images for a single flexible nanofilament suspended in liquid. (a) – Brownian motion observed for very long filament (R= 90 nm, L= 60 μm), image height 35 μm; (b) - filament conveyed by Poiseuille flow (no. 4 in Table 1), image centerline located 38 μm from the wall, t – time stamp, x – distance traveled from the channel entry (picture frame moves with the object), image height 66 μm.

Mobility of our filaments strongly depends on their length. For short pieces (10 – 20 μm) translational and rotational diffusion coefficients could be evaluated from Brownian motion. They are typically of the same order of magnitude as those obtained for rigid rods. For longer filaments their bending flexibility is well exhibited, especially pronounced for millimeters long objects (Fig. 1a). Interesting to note that despite negligible ionic interactions bending following characteristic coiling and knots formation is observed. This effect remains to dominate for objects conveyed by Poiseuille flow (Fig. 1b). The approximate center of mass for the conveyed filaments slightly lags behind the local flow field (Fig. 2), whereas cross-flow migration into the center of channel can be detected. Applying periodic flow we may notice evidence of broken reversibility of the Stokesian approximation. This is obvious for motion of deformable objects like droplets, bubbles and flexible fibers. In the present study it helps us to perform long time study for the same filament remaining in the field of observation.

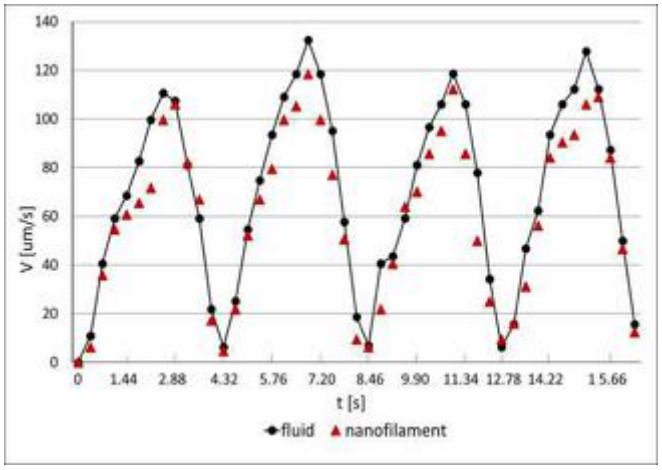

Figure 2. Velocity of fluid and for center of mass of the filament conveyed by pulsatile flow.

CONCLUSIONS

We have demonstrated possibility to create useful microscale experimental model to study hydrodynamic interactions for highly flexible long objects formed as hydrogel nanofilaments. Such objects can be used as possible carriers or indicators in biomedical applications. Our study gives evidence of difficulties to describe mobility of long flexible nanofilaments in terms of coarse parametric description, like hydrodynamic diameter and constant friction tensor. At each time step variable conformation of the observed objects strongly modifies hydrodynamic interactions, effects of wall and shear field forces, leading to more or less predictable evolution of their transport properties. Probably statistical analysis performed for large number of experiments is the only way to obtain plausible average mobility values convenient to predict longitudinal and cross-flow transport properties of such objects.

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