Supplementary Information

"Drop Dynamics of Viscoelastic Filament"

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In this Supplemental Material, we present the experimental setup (§1), the sample solutions (§2), the solutions properties (§3), the evolution of D_{CaBER} (§4), additional space-time diagrams (§5), and the description of the supplementary videos (§6).

I. EXPERIMENTAL SETUP

In the capillary breakup extensional rheometer (CaBER), the sample solution is placed between two plates and stretched. The laser micrometer has a resolution of 0.01 mm and measures around the mid-plane diameter of thinning filament. In addition, an imaging system consists of a CaBER, a high-speed camera (Miro series from Phantom) fitted with a (Questar) lens allowing shadowgraph images enabling a resolution up to ≈ 3 μm, a 2 W continuous laser (RayPower) and anti-speckle optic, as shown schematically in Fig. 1. The bottom plate of CaBER is fixed and the top plate can be moved at a desired speed with the help of a linear motor having resolution of 0.02 mm. The experimental setup is kept in a temperature-controlled room. Moreover, a thermal bath (from ThermoFisher scientific) is used to ensure CaBER remains at 20°C. With the help of a pipette (from Eppendorf research plus, range of 10–100 μL), samples are placed between two steel plates.

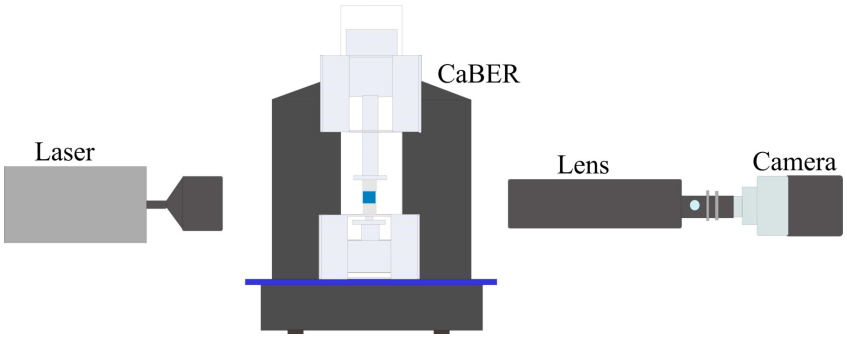

FIG. 1. Schematic of the experimental setup (drawn to scale). The small blue rectangle represents the sample polymer solution.

II. SAMPLE SOLUTIONS

Three different types of aqueous solutions: polyethylene glycol (PEG), polyethylene oxyde (PEO) and PEG+PEO solutions, as shown in Table I, are prepared as per the desired viscosity, as well as, elasticity in degassed deionised water. In the first type of solution, to make it highly viscous, PEG is added to the water; specifically 20% wt. and is labelled as "PEG20". In the second type, to have highly elastic solutions, PEO is mixed with the water in various concentrations. In the third type of solutions, hereafter "PEG+PEO" solutions, both PEO and PEG are added separately to the water and then mixed together to create viscoelastic solutions. In the mixtures of PEG+PEO, the concentration of PEG is 20% wt. and is kept constant. For any solutions containing PEO, 0.5% wt. iso-propyl alchohol (IPA) is added for easy dispersion and stabilisation [1] of PEO molecules in solvent. Preparation and storage of all solutions are carried out at room temperature, 20°C. The solutions are kept for at least 72 hours to ensure complete mixing of the polymers. As PEG and IPA are used along with PEO, it is difficult to calculate the critical concentration, c^*, from a dilute to semi-concentrated solution. In addition, there is no exact formula for high molecular weight of PEO at 20°C. Hence, to have approximate estimate of c^*, the Mark-Houwink-Sakurada formula, which agrees well for PEO in water over the range of molecular weight, $M_w = 8 \times 10^3$ to 5×10^6 g/mol as found by Tirtaatmadja *et al.* [2], is used to calculate the intrinsic viscosity: $[\eta] = 0.072 \times M_w^{0.65}$. For PEO solution at 20°C, $[\eta] \simeq 2210$ mL/g and

$c^* = 0.77/[\eta] \simeq 348$ ppm. For PEG solution at 20°C, $[\eta] \simeq 45$ mL/g and $c^* \approx 1.71\%$. Hence, it is assumed, for PEG+PEO solutions with 20% wt. PEG, that interactions between PEG molecules are significant.

III. SOLUTIONS PROPERTIES

Our solutions are thus prepared above, as well as, below 348 ppm of PEO, as shown in Table I, to cover a wide range of concentrations. To measure the zero shear viscosity, η_0, the solutions are tested using (TA Instrument Discovery HR-3) rheometers with double wall concentric cylinder geometry: inside cup diameter 30.2 mm, outside cup diameter 37 mm, inside bob diameter 32 mm, outside bob diameter 35 mm and inner cylinder height 55 mm. For the obtained results, as shown is Fig. 2, the Carreau fit is used to calculate the zero shear viscosity. The results are in agreement with Crumeyrolle *et al.* [3]. Densities of the solutions are measured with a portable density-meter from Anton Paar (resolution 0.1 mg/cm^3). Finally, the surface tensions, σ, of solutions are measured with (Kruss) Drop Shape Analyser (series 100) with a resolution 0.01 mN/m.

TABLE I. Properties of the solutions

Abbreviation	c_{PEG} % wt.[a]	c_{PEO} ppm[a]	ρ kg/m^3	σ mN/m	η_0 mPa.s	λ ms	De
PEG20	20	0	1033	57	136		
PEO100	0	100	997	59	1	41	1.9
PEO250	0	250	997	59	2	106	4.9
PEO1000	0	1000	997	59	6	344	16.1
PEO2000	0	2000	998	59	60	696	32.7
PEG20PEO100	20	100	1033	56	156	535	24.0
PEG20PEO250	20	250	1033	56	169	1350	60.8
PEG20PEO400	20	400	1034	56	185[b]	1367	61.7
PEG20PEO800	20	800	1034	56	281[b]	2396	108.0
PEG20PEO1000	20	1000	1034	56	333	2952	132.8
PEG20PEO2000	20	2000	1034	56	576	3580	161.3

[a] 1 g/mL $\doteq 10^6$ ppm = 0.01% wt.
[b] These data were obtained by interpolation

IV. EVOLUTION OF D_{CaBER}

Fig. 3 presents the diameter of the filament measured by CaBER's micrometer for all solutions. It is interesting to note that PEG20 shows Newtonian behaviour. Moreover, with increase in concentration of PEO, the filament breakup time increases.

V. SPACE-TIME DIAGRAMS

Additional diameter-space-time diagrams (DST) and the Hencky strain-space-time diagrams (HSST) are depicted in Fig. 4 for PEO100 and PEO2000, respectively. The DST and HSST diagrams for PEG20PEO100, PEG20PEO1000 and PEG20PEO2000 are shown in Fig. 5. The influence of concentration of PEO, as well as, PEG on the filament breakup time and the number of drops can be clearly observed.

VI. VIDEOS

Two experimental videos for PEG20PEO1000 and PEG20PEO2000 are provided:

- PEG20PEO1000.gif shows the complete filament thinning until breakup.
- PEG20PEO2000.avi shows a zoom and slow-motion of the pinching and the coalescence of drops.

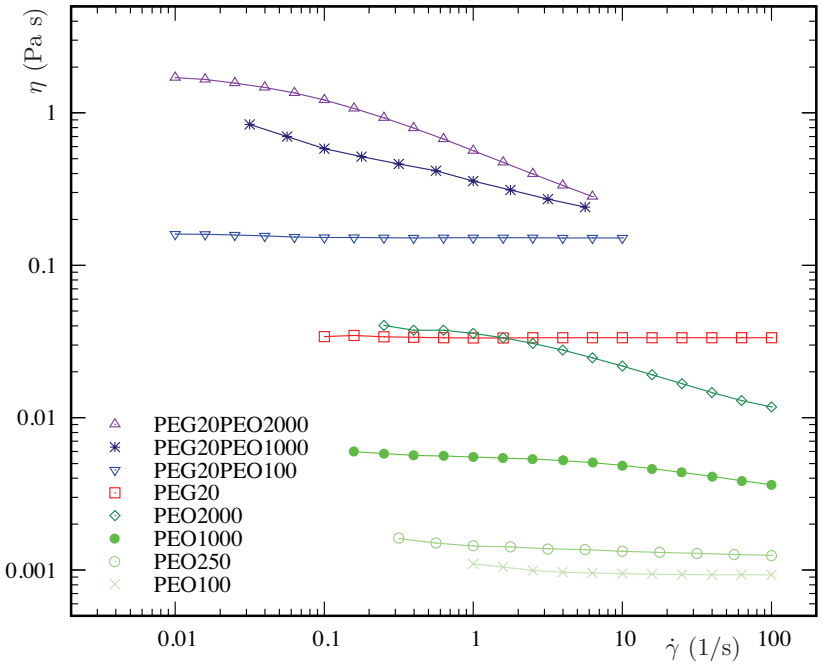

FIG. 2. Shear viscosity, η, versus shear rate, $\dot{\gamma}$, measurements for all tested solutions

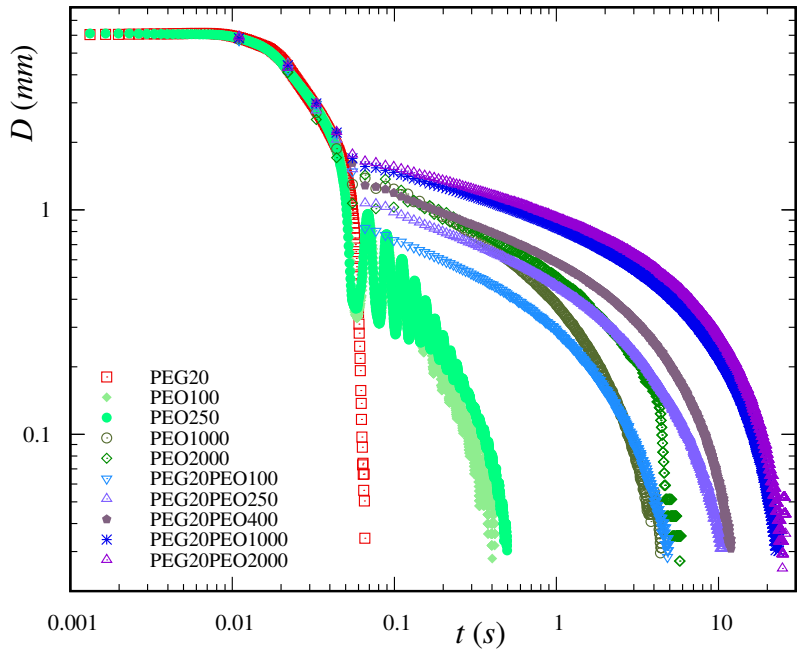

FIG. 3. Evolution of the CaBER diameter, D_{CaBER}, versus time for all measured solutions

[1] Y. Layec and M.-N. Layec-Raphalen, "Instability of dilute poly(ethylene-oxide) solutions," J. Physique Lett. **44**, 121–128 (1983).

[2] V. Tirtaatmadja, G. H. McKinley, and J. J. Cooper-White, "Drop formation and breakup of low viscosity elastic fluids: Effects of molecular weight and concentration," Phys. Fluids **18**, 043101 (2006).

[3] O. Crumeyrolle, N. Latrache, A. Ezersky, and I. Mutabazi, "Instability modes observed in a viscoelastic couettetaylor flow," Mechanics & Industries **4**, 397–409 (2003).

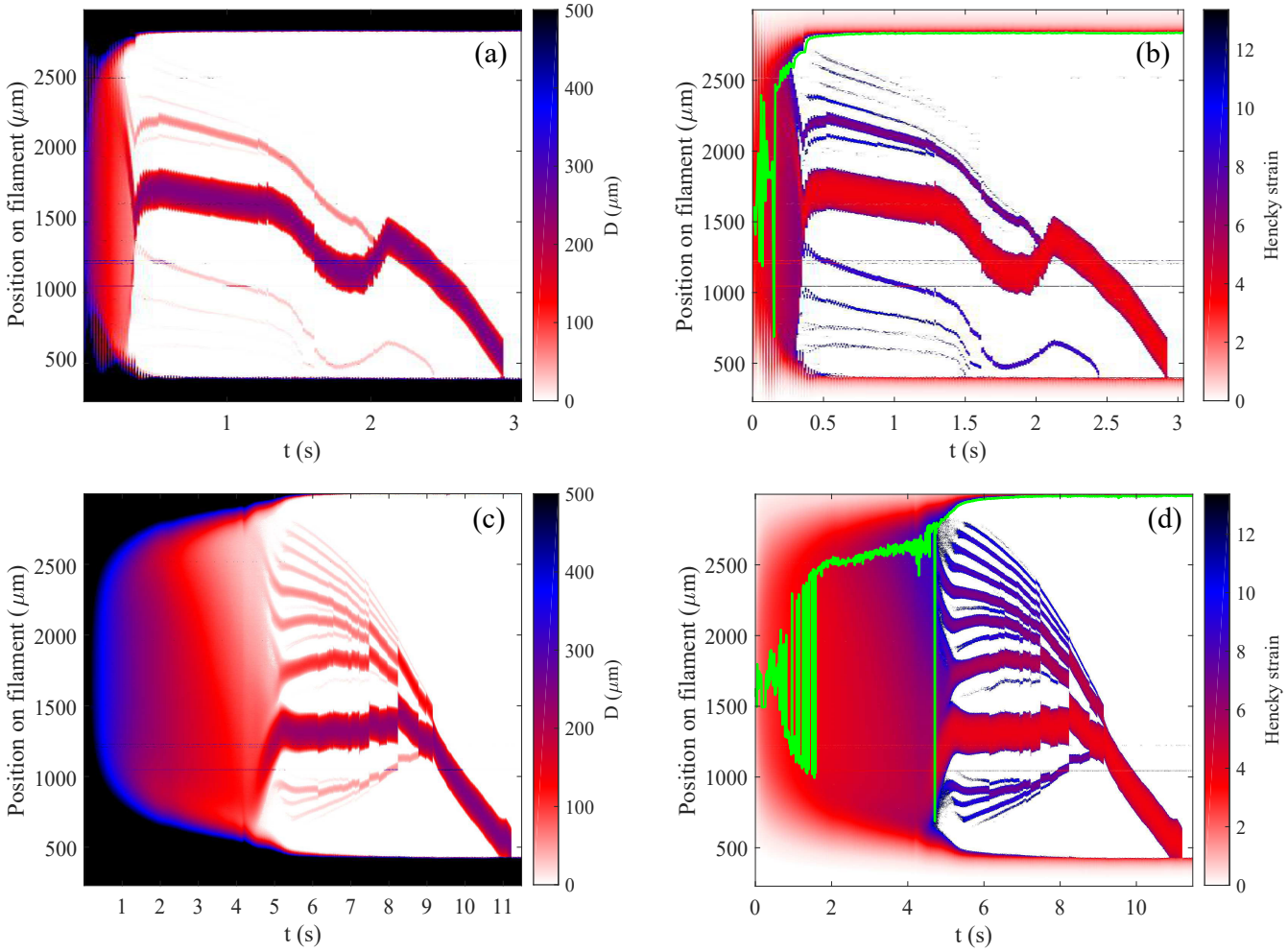

FIG. 4. (a) DST and (b) HST for PEO100, (c) DST and (d) HSST for PEO2000

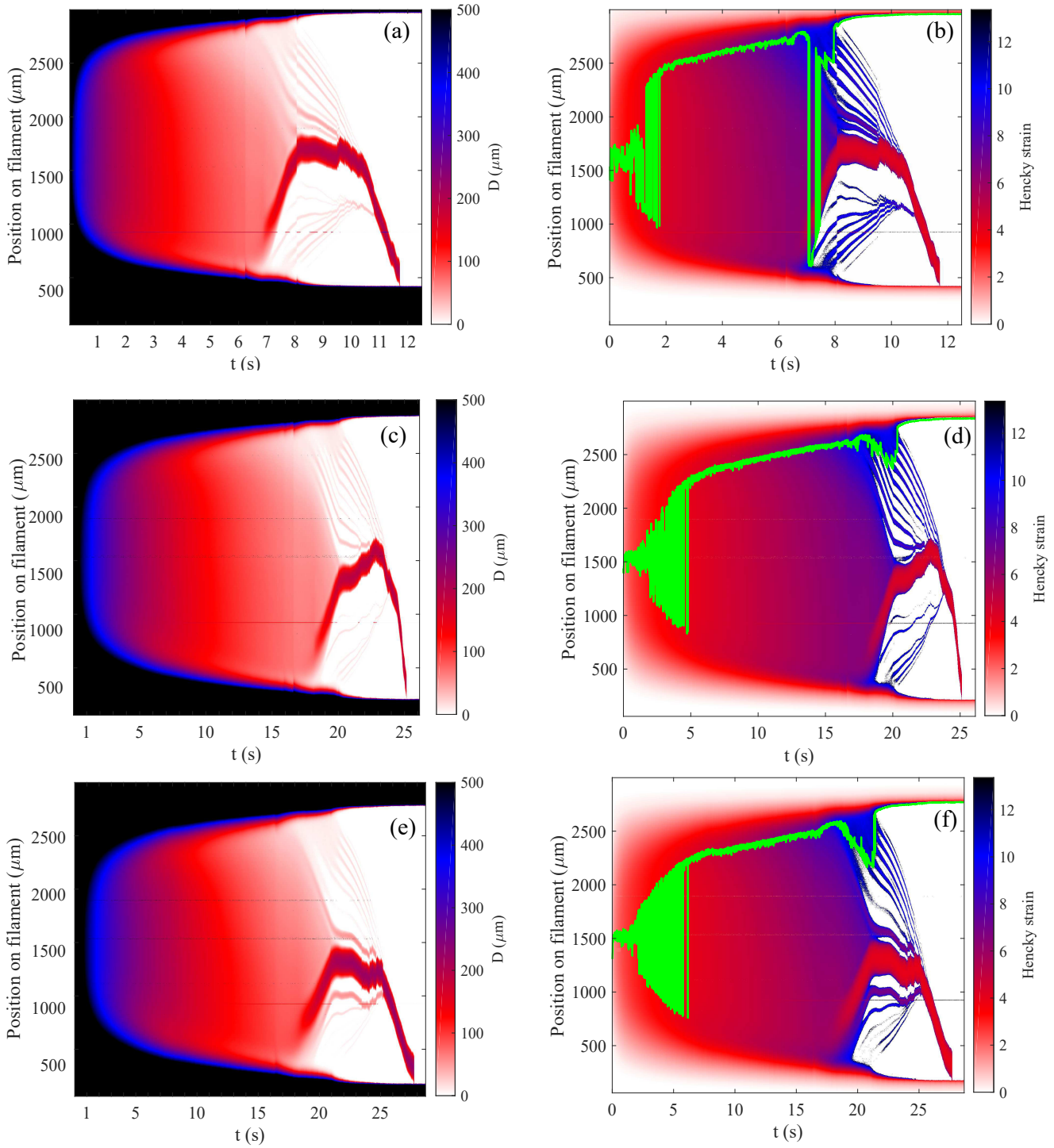

FIG. 5. (a) DST and (b) HSST for PEG20PEO100, (c) DST and (d) for PEG20PEO1000, (e) DST and (f) HSST for PEG20PEO2000