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What determines the drop size in sprays of polymer solutions?

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1. Introduction

Spraying is a widely used technique that involves the fragmentation of a liquid sheet or jet into droplets. This process usually involves multiple hydrodynamic instabilities [1,2]. Sprays are important in a wide range of application areas such as agriculture (crop spraying), industry (inkjet printing, spray painting) and medicine (drug delivery) [3,4]. In many applications, understanding how the size distribution of spray droplets depends on external control parameters is of paramount importance. This is particularly true for agricultural pesticide spraying [5] where droplets typically smaller than 100 micrometers may become aerosols which can be blown away by the wind (spray drift), causing environmental pollution [6–8]. Addition of small amounts of polymer molecules to the sprayed liquid has been shown to lead to the formation of larger droplets, hence reducing the volume of spray drift produced by agricultural nozzles [9–12], while also improving droplet deposition on plants by inhibiting the bouncing effect observed for water droplets [13,14]. Polymers hence offer advantages over surfactant additives, which have been shown to only improve the deposition process [15] and not the droplet size distribution [16]. Understanding the effect of viscoelasticity in liquid atomization is also relevant for jet fuel and fire-fighting applications [17,18], as well as for understanding virus propagation through coughing and sneezing [19,20]. However, viscoelastic effects in sprays are not yet fully understood [21].

In this paper, we focus on the fragmentation of liquid sheets generated by flat fan nozzles. For dilute solutions, the mechanism leading to the formation of spray droplets is qualitatively similar to that of Newtonian liquids [11,22]. This is illustrated in Fig. 1 which compares front-view and side-view images of spray sheets of water (a) and of a dilute solution of high-molecular-weight polyethylene oxide (PEO) (b). The fragmentation of the spray sheet into droplets is initiated by the growth of out-of-plane oscillations along the sheet caused by friction with the surrounding air via the Squire instability [23], as can be observed in the side-view images of Fig. 1. Thickness modulations in the sheet caused by this flapping motion [24] result in the rupture of the sheet into fragments of size proportional to the Squire wavelength λ [25], as can be observed in the front-view images of Fig. 1. These sheet fragments retract into ligaments which ultimately breakup into droplets due to surface tension via the Rayleigh–Plateau instability [26].

Kooij et al. [27] derived a model for low-viscosity Newtonian liquids where the median diameter $D_{50}$ of spray droplets is set by the Squire wavelength and by the thickness of the sheet at the point of breakup. This model leads to the following universal expression

$$D_{50}/b = c_1 a^{-1/6} W e^{-1/3}$$

where $a$ is the air-to-liquid-density ratio, $c_1$ is a numerical constant and the Weber number is $W e = \rho U^2 b/\sigma$ with $b$ a characteristic size of the nozzle opening, $U$ the liquid velocity, $\rho$ the liquid density and $\sigma$ the surface tension. This expression, which does not account for viscous...
effects, was found to be in agreement with experimental measurements for liquids of viscosities up to 30 times larger than that of water.

Many studies have shown that increasing the extensional viscosity of the sprayed liquid by adding polymers to a Newtonian solvent leads to the formation of larger droplets, for both flat fan nozzles and pressure swirl atomizers producing flat and conical sheets, respectively [12,28–31]. The first attempt to propose a correlation between the characteristic droplet size and pertinent non-dimensional numbers is found in the work of Stelter et al. [11], who performed measurements of the droplet size distribution for different nozzles, spraying dilute aqueous solutions of both flexible and rigid polymers. Viscoelasticity is quantified by an extensional relaxation time $\tau$ measured using a Capillary Breakup Extensional Rheometer (CaBER). Their empirical expression predicts that the (Sauter) mean diameter, expressed in terms of Weber and Reynolds numbers, scales as $\tau^{0.425}$ for viscoelastic liquids and scales as $\eta^{0.213}$ for Newtonian liquids of dynamic shear viscosity $\eta$. However, these predictions are inconsistent with the absence of viscous effects measured by Kooij et al. [27] for low-viscosity Newtonian liquids. A more recent attempt is found in the work of Brenn & Plohl [32], who used a linear analysis of the instability of viscoelastic liquid sheets [33] and jets [34] to derive an expression in which the increase in droplet size after polymer addition is mainly attributed to the increase of shear viscosity of the liquid, while an increase in polymer relaxation time for a given shear viscosity leads to a decrease in droplet size. These predictions are inconsistent with the increase in droplet size observed by Mun et al. [30] for polymer solutions exhibiting the same shear viscosity. In fact, these predictions are based on a linearized viscoelastic model which do not account for the strain hardening properties associated with large deformations of polymer molecules in spray flows.

Hence, to the best of our knowledge, no satisfactory expression has been proposed for the characteristic droplet size in sprays of viscoelastic liquids produced by flat fan nozzles. For air-assisted jet atomization nozzles on the other hand, Keshavarz et al. [35] showed a logarithmic increase of the mean droplet size with the relaxation time of the polymer solution, consistent with earlier measurements by Christanti & Walker [36]. Keshavarz et al. [35] also proposed a phenomenological model that accounts for their observations.

For flat fan nozzles, the physical origin of the increase in droplet size with increasing viscoelasticity is also an open question. Stelter et al. [11] proposed that it is a consequence of the retardation of ligament breakup caused by strain hardening. The authors also proposed that the ligament size is not affected since the sheet thickness at breakup was estimated to be unchanged after polymer addition. Later, Hartranft & Settles [22] proposed that spray droplets are larger because of an increase in ligament size caused by (i) an increase in the Squire wavelength and (ii) a decrease in sheet length (while Stelter et al. [11] reported an increase in the sheet length in their experiments). However, these studies did not provide quantitative comparisons of the drop size against relevant physical quantities to validate their hypothesis.

In this study, we propose a universal empirical expression of the median diameter of spray droplets of polymer solutions generated from flat fan nozzles in terms of the non-dimensional Weber and Deborah numbers. The physical origin of the increasing drop size with increasing viscoelasticity is also investigated via measurements of different quantities involved in the sheet fragmentation mechanism, revealing a significant role of the increasing Squire wavelength. Finally, the effect of viscoelasticity on the droplet size distribution is discussed.

Section 2 describes the experimental setup and the measurement techniques as well as the rheology of the test liquids. It also includes...
a discussion of the mechanical degradation of the polymer solutions. The experimental results are presented in Section 3 and discussed in Section 4.

2. Materials and methods

2.1. Nozzles

Sprays are generated using two flat fan nozzles provided by TeeJet® Technologies, producing flat liquid sheets. The dimensions of the oval opening of each nozzle, measured from high-resolution images, are reported in Table 1, where $A$ is the nozzle area and $b$ and $a$ are the nozzle thickness and width corresponding to the minimum and maximum lengths of the cross section area, respectively. The spray angle $\theta_s$ measured for water from front pictures of the sheet is observed to increase in a range reported in Table 1 when increasing the operating pressure from 1 to 8 bar.

2.2. Test liquids and injection system

Spray experiments are performed using aqueous solutions of polyethylene oxide with molecular weight $M_W = 4 \cdot 10^6$ g/mol provided by Alroko®, henceforth referred to as PEO-4M. Solutions are obtained from dilution of 10 000 ppm (weight parts per million) stock solutions mixed for 16 h by a mechanical stirrer to ensure homogeneity. To minimize aggregation, a mass of isopropanol equal to the mass of polymer is added to the polymer powder before it is added to water. Rheological measurements reveal that stock solutions prepared via this protocol are highly reproducible.

The polymer solutions fed to the nozzle are generated by a pressure powered polymer injection system from Dosatron® (ref. D25WL2 IE PO + Mixer). Tap water is pumped into a water inlet, generating suction of a concentrated polymer solution through a polymer inlet, which is injected into the water flow at a controlled volume ratio. Polymer injection is followed by passage through a mixing chamber delivering a homogeneous diluted polymer solution. The concentration of these diluted solutions is varied between 10 and 100 ppm. The mixer outlet is connected to the nozzle inlet through a rigid pipe equipped with a pressure and flow rate meter provided by Krohne® (ref. OPTIBAR PM 3050 and H250 RR M40) and time measurements of the flow rate and pressure are recorded on a computer. The Dosatron injection system causes periodic pressure variations of the order of 0.2 bar on the operating pressure are recorded on a computer. The Dosatron injection system causes periodic pressure variations of the order of 0.2 bar on the operating pressure is 0.2 bar on a time scale of about 20 s. Hence, the experimental data reported in this work correspond to averages calculated over 2 min of experiments performed at a constant average pressure $P$ and volumetric flow rate $Q$.

The flow rate varies with pressure according to $Q = C_p A \sqrt{2P/\rho}$, where $\rho$ is the liquid density and $C_p$ a discharge coefficient, which is found to not vary significantly with polymer concentration and is close to 1, as also reported by Kooijj et al. [27]. The operating pressure is varied from 1 to 8 bar, yielding liquid velocities $U_0 = Q/A$ in the nozzle ranging between 10 and 40 m/s for both nozzles.

2.3. Droplet size measurements

The size distribution of the spray droplets is measured by a laser diffraction instrument from Malvern Spraytech®. This method has been shown to give results comparable to other measurements techniques [37]. The laser beam is placed vertically below the nozzle and passes through the spray in a direction perpendicular to its plane. The diffraction angle being inversely proportional to the droplet size, the light diffraction pattern allows, assuming a spherical shape of the droplets, to obtain the droplet size distribution [38,39]. The laser beam is placed at a fixed distance 50 cm below the nozzle where high-speed photography reveals that droplets are reasonably spherical and are no longer connected by filaments, see Christanti & Walker [36] for calculations of the effect of filaments on drop size measurements.

2.4. Shear rheology

In Sections 2.4 and 2.5, we present shear and extensional rheology measurements performed on PEO-4M solutions prepared by dilutions from a 10 000 ppm stock solution. These measurements include concentrations above the maximum concentration 100 ppm used in spray experiments for a more general characterization of aqueous PEO solutions. The rheology of solutions used in spray experiments is discussed in Section 2.6. Rheological measurements are performed at the temperature of spray experiments (18.5°C).

Shear rheology measurements are performed using a MRC-302 rheometer from Anton Paar equipped with a cone plate geometry (diameter 50 mm, angle 1° and truncation gap 53 μm). The apparent shear viscosity $\eta$ and first normal stress difference $N_1$ are plotted in Fig. 2(a) against the shear rate $\dot{\gamma}$ for water and PEO-4M solutions. To measure $N_1$, we follow a step-by-step protocol similar to Casanellas et al. [40] in order to circumvent the instrumental drift of the normal force. This protocol consists in applying steps of constant shear rate followed by steps of zero shear and subtracting the two raw $N_1$ plateau values. The contribution of inertia to the normal force is corrected for by the rheometer [41].

We fit the apparent shear viscosity in Fig. 2(a) using the Carreau–Yasuda formula:

$$\eta(\dot{\gamma}) = \eta_0 (1 + (\dot{\gamma}/\dot{\gamma}_c)^n)^{\frac{n-1}{n}},$$

where $\eta_0$ is the zero shear viscosity, $\dot{\gamma}_c$ is the shear rate beyond which shear thinning is observed, $n$ is the shear thinning exponent and $\dot{\gamma}_c$ encodes the sharpness of the transition towards the shear thinning regime. The first normal stress difference $N_1$ in Fig. 2(a) can be described by a power law:

$$N_1 = \Psi_1 \dot{\gamma}^{n_1},$$

where $\Psi_1$ is the first normal stress coefficient. All fitting parameters are reported in Table 2. The table also includes the surface tension $\sigma$ of the solutions, which is measured by a pendant drop method. The liquid density is $\rho = 997$ kg/m$^3$ for all solutions.

The polymer viscosity is defined as $\eta_g = \eta_0 - \eta_1$, where $\eta_1 = 1.036$ mPa s is the viscosity of the water solvent. We find that $\eta_g$ increases linearly at low polymer concentrations (dilute regime), i.e. $\eta_g = \eta_g[\eta]c$ with $c$ the polymer mass concentration. We find an intrinsic viscosity $[\eta] = 2.61$ m$^2$/kg. Following the expression of Graessley [43], the critical overlap concentration is $c^* = 0.77/[\eta] = 0.295$ kg/m$^3$ (equivalently 296 ppm). Hence, PEO solutions used in spray experiments with concentrations up to 100 ppm are dilute solutions with a quasi constant shear viscosity $\eta_0$ $(n \approx 1)$ and quadratic first normal stress difference ($Q_1 = 2$), see Table 2. Note that our value of $[\eta]$ is 1.85 times larger than the value expected from the Mark–Houwink prediction $[\eta] = 0.072 M^6$ from Tirtaatmadja et al. [44] for PEO. A possible explanation is a difference in PEO provider leading to a different polydispersity.

<table>
<thead>
<tr>
<th>Type</th>
<th>Nozzle</th>
<th>A (mm$^2$)</th>
<th>b (mm)</th>
<th>a (mm)</th>
<th>$a/b$</th>
<th>$\theta_s$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flat fan</td>
<td>XR Teejet 11 004 VS</td>
<td>1.15</td>
<td>0.601</td>
<td>2.25</td>
<td>3.74</td>
<td>105–120</td>
</tr>
<tr>
<td>Flat fan</td>
<td>XR Teejet 11 002 VK</td>
<td>0.545</td>
<td>0.458</td>
<td>1.52</td>
<td>3.31</td>
<td>93–114</td>
</tr>
</tbody>
</table>

Table 1: Dimensions of the oval nozzle opening of the two flat fan nozzles: area $A$, thickness $b$, width $a$, aspect ratio $a/b$. The range of spray angles $\theta_s$ measured for water corresponds to operating pressures from 1 to 8 bar.
\[ \eta_{\text{app}} = -\sigma/(dD_{\min}/dt) \] saturates to a value \( \eta_E \) called the terminal extensional viscosity. The FENE-P model predicts \( \eta_E = 3\eta_0 + 2\eta_0 L^2 \) [51] where \( L^2 \) is the finite extensibility parameter and \( L \) is the ratio of the polymer size at full extension to its size in the coiled state at equilibrium. Values of \( \eta_E \) are obtained by plotting \( \eta_{\text{app}} \) as a function of the Hencky strain \( \varepsilon = -2\ln(D_{\min}/D_0) \), where \( D_0 \) is the minimum bridge diameter slightly before onset of the elastic regime when polymer molecule start unrolling, see inset of Fig. 2(b). Values of \( \eta_E \) and \( L^2 = (\eta_E - 3\eta_0)/(2\eta_0) \) are reported in Table 2. We find \( \tau \propto e^{0.76} \), consistent with Tirtaatmadja et al. [44] and Zell et al. [52] and \( \eta_E \propto \tau \), consistent with Stelter et al. [53].

2.6. Mechanical degradation

The rheology of the polymer solutions used in spray experiments is investigated by collecting samples of the sprayed liquid. In Fig. 3, we plot the extensional relaxation time \( \tau \) measured on samples of different polymer concentrations, sprayed at different flow rates from both nozzles, against the liquid velocity \( U_0 \) in the nozzle. A significant reduction of the relaxation time is observed when increasing the flow rate, meaning that polymer solutions undergo severe mechanical degradation, as also reported by Stelter et al. [11] and Hartranft & Settles [22]. Note that we disregard samples for which no clear exponential regime could be observed in CaBER experiments, as advised by Campo-Deano & Clasen [45]. Since the spray formation is governed by the properties of the liquid in the spray sheet, it would be incorrect to correlate

Table 2

Rheological parameters and surface tension \( \sigma \) of PEO-4M solutions used for rheological characterization. Shear parameters: \( \eta_0, n, \gamma, \eta_s \) and \( \Psi_1 \) are such that the shear viscosity \( \eta(\gamma) \) and the first normal stress difference \( \Psi_1(\gamma) \) are captured by Eqs. (2) and (3) \( (N_s) \) was not measurable at 10 ppm. Values of \( c'/c \) are indicated, where \( c \) is the polymer mass fraction and \( c' = 0.259 \text{ kg/m}^3 \) (equivalently 296 ppm) is the critical overlap concentration. \( \eta_s = \eta_0 - \eta_e \), where \( \eta_e = 1.036 \text{ mPa s} \) is the water solvent viscosity. We use \( \eta_0 = \eta_0 \) to determine \( \eta_{\text{app}} \) at 10 ppm since the difference between \( \eta_0 \) and \( \eta_e \) is too small to be measured directly, where \( |\eta| = 2.61 \text{ mA}^2/\text{kg} \) is the intrinsic viscosity. Extensional parameters (CaBER): \( \tau \) is the extensional relaxation time and \( \eta_{\text{app}} \) is the terminal extensional viscosity. \( L^2 = (\eta_0 - 3\eta_e)/(2\eta_0) \) is the effective value of the finite extensibility parameter.

<table>
<thead>
<tr>
<th>[PEO] (ppm)</th>
<th>( c'/c )</th>
<th>( \sigma ) (mN/m)</th>
<th>( \eta_0 ) (mPa s)</th>
<th>( \eta_s ) (mPa s)</th>
<th>( n )</th>
<th>( \phi_1/\psi_1 )</th>
<th>( \eta_{\text{app}} ) (Pa s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>0.034</td>
<td>67.1</td>
<td>1.037</td>
<td>0.027</td>
<td>1.00</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>25</td>
<td>0.084</td>
<td>64.1</td>
<td>1.098</td>
<td>0.062</td>
<td>1.00</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>50</td>
<td>0.17</td>
<td>63.0</td>
<td>1.17</td>
<td>0.134</td>
<td>0.98</td>
<td>0.0258</td>
<td>2.5 \times 10^{-4}</td>
</tr>
<tr>
<td>100</td>
<td>0.34</td>
<td>62.8</td>
<td>1.28</td>
<td>0.244</td>
<td>0.98</td>
<td>0.0335</td>
<td>6.54 \times 10^{-4}</td>
</tr>
<tr>
<td>150</td>
<td>0.51</td>
<td>62.5</td>
<td>1.44</td>
<td>0.404</td>
<td>0.97</td>
<td>0.0445</td>
<td>13.8 \times 10^{-4}</td>
</tr>
<tr>
<td>250</td>
<td>0.84</td>
<td>62.5</td>
<td>1.72</td>
<td>0.684</td>
<td>0.96</td>
<td>0.0975</td>
<td>2.0 \times 10^{-4}</td>
</tr>
<tr>
<td>500</td>
<td>1.7</td>
<td>62.5</td>
<td>2.95</td>
<td>1.91</td>
<td>0.93</td>
<td>0.130</td>
<td>5.5 \times 10^{-5}</td>
</tr>
<tr>
<td>1000</td>
<td>3.4</td>
<td>62.5</td>
<td>6.3</td>
<td>5.26</td>
<td>0.86</td>
<td>0.152</td>
<td>7.0 \times 10^{-5}</td>
</tr>
<tr>
<td>5000</td>
<td>8.4</td>
<td>62.1</td>
<td>38</td>
<td>37.0</td>
<td>0.71</td>
<td>0.282</td>
<td>1.47 \times 10^{-4}</td>
</tr>
<tr>
<td>10000</td>
<td>34</td>
<td>61.5</td>
<td>15000</td>
<td>15000</td>
<td>0.42</td>
<td>23.8</td>
<td>2.2 \times 10^{-5}</td>
</tr>
</tbody>
</table>

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Fig. 2. (a) Apparent shear viscosity \( \eta(\gamma) \) and first normal stress difference \( N_s \) as a function of shear rate \( \gamma \) for water and PEO-4M polymer solutions of various concentrations. Data are fitted respectively by Eqs. (2) and (3). (b) Minimum filament diameter \( D_{\min} \) as a function of the time \( t-\tau \) elapsed since the beginning of the elastic regime for the same solutions, with the same color coding as panel (a). Data are fitted by the empirical law proposed by Anna & McKinley [42]: \( D_{\min} = \sigma c' = \alpha + \gamma \), where \( a, b, c \) and \( d \) are fitting parameters. The inset figure shows the apparent extensional viscosity \( \eta_{\text{app}} = -\sigma/(dD_{\min}/dt) \) as a function of the Hencky strain \( \varepsilon = -2\ln(D_{\min}/D_0) \), where we choose \( D_1 = 1 \text{ mm} \). Inset snapshots labeled 1 to 4 show a filament of a 2500 ppm solution at different stages of thinning indicated by the same labels on the corresponding \( D_{\min} \) and \( \eta_{\text{app}} \) curves.

2.5. Extensional rheology

Extensional rheology measurements are performed using a non-commercial Capillary Breakup Extensional Rheometer (CaBER) based on the filament thinning technique documented by Anna & McKinley [42]. A drop of polymer solution is placed between two horizontal circular plates of diameter 5 mm which are separated until the liquid bridge connecting the two end drops becomes unstable, transiently forming a filament in which polymer molecules are stretched by an extensional flow. Following Campo-Deano & Clasen [45], the plates are separated at a low velocity (200 \( \mu \text{m/s} \)) to minimize oscillations of the two end drops inherent to the classical step-strain plate separation protocols [46]. The minimum filament diameter \( D_{\min} \) is calculated from images recorded by a high-speed camera equipped with a high-magnification objective.

Time evolution of the minimum filament diameter is shown in Fig. 2(b) for PEO-4M solutions of various concentrations, showing an exponential decay in the elastic regime. This is consistent with the multimode Oldroyd-B model which, for the elastic regime starting at \( t = \tau_0 \), predicts [47–49] that \( D_{\min} \propto \exp(-\tau/\tau_0)/\tau_0 \) where \( \tau \) is the longest relaxation time of the spectrum. Values of \( \tau \) are obtained from fitting the experimental exponential regime to this expression. In this regime, the extension rate \( \varepsilon \equiv -2(dD_{\min}/dt)/D_{\min} \) gives \( \varepsilon' = 2/\tau \) which is above the coil-stretch transition value 1/2 [50]. Hence, polymer chains progressively unravel, ultimately reaching full extension, at which point the apparent extensional viscosity defined as

\[ \eta_{\text{app}} = -\sigma/(dD_{\min}/dt), \]
spray properties to the rheology of the initial fresh solution before degradation. Fig. 3 shows that the decrease in \( r(U_0) \) is well fitted by an exponential decay for every polymer concentration. The parameters of the fits are hence used to calculate the relaxation time in the spray experiments reported in Section 3.

We note that values of \( r \) in Fig. 3 are the same for both nozzles at a given liquid velocity and polymer concentration. This is surprising since a reduction of the nozzle area is expected to lead to more degradation, owing to the increase in strain rate associated to the contraction flow at the nozzle inlet, as reported by Stelter et al. [11]. We therefore suspect that degradation might not occur only at the nozzle opening in our experiments.

3. Results

We start by a qualitative description of the sheet fragmentation process. We find that, similarly to Newtonian fluids, the fragmentation of the viscoelastic spray sheet is initiated by the growth of Squire waves along the sheet, leading to the formation of sheet fragments retracting into ligaments which ultimately breakup into droplets. This is shown in the high-speed images of Fig. 1, which compares a spray of water (a) and of 25 ppm PEO-4M solution (b) for the same nozzle and operating pressure. However, significant qualitative differences are observed in these images.

First, the liquid sheet close to the nozzle is much smoother in the viscoelastic case, reminiscent of the reduction of small-scale surface disturbances in jets of polymer solutions reported by Hoyt et al. [18]. The rim of the sheet is also stabilized, as also observed by Thompson & Rothstein [54] for flat fan sprays of wormlike micelle solutions. We also observe that polymers delay the breakup of the sheet to a larger distance away from the nozzle, consistent with observations by Stelter et al. [11], suggesting a stabilizing effect of viscoelasticity. This is reminiscent of the work of Karim et al. [55], who found that polymer addition may prevent the breakup of a liquid curtain subjected to a localized perturbation created by an air jet. The authors concluded that the growth rate of disturbances that may lead to a perforation of the sheet is delayed by elastic stresses. Following sheet fragmentation, we observe that the breakup of ligaments into droplets is delayed, resulting in the formation of a complex filamentous network below the sheet, see insets labeled ‘zoom’ in Fig. 1(b). This is also reported by various studies of a wide range of jet and sheet fragmentation processes [31,36,54,56-60] and is attributed to the strain hardening properties of polymer solutions [51].

3.1. Drop size

We now turn to the size of spray droplets as measured by laser diffraction. In Fig. 4(a), we compare the drop size distribution measured for water and PEO-4M solutions of various concentrations for a given nozzle and operating pressure. The flow rate is the same to within 6% for all these measurements since its dependence on operating pressure is only marginally influenced by polymer addition for flat fan nozzles. The figure shows that spray droplets of polymer solutions are overall bigger than for the water solvent alone and feature a broader size distribution. We first focus on the characteristic drop size, leaving discussions of the distribution to Section 3.8.

We choose the volume median diameter \( D_{50} \) as a characteristic drop size. It is compared with the mean drop size \((D)\) and with the Sauter mean diameter in appendix Appendix A. In Fig. 4(b), we plot \( D_{50} \) rescaled by the thickness \( b \) of the nozzle opening against the Weber number

\[
We = \frac{\rho U_0^2 b}{\sigma}
\]

for sprays of water and PEO-4M solutions of various concentrations and for all nozzles. The data in Fig. 4(b) is obtained by varying the liquid flow rate for each polymer concentration and nozzle. In the case of water sprays, the decrease with increasing Weber number is well captured by Eq. (1), corresponding to the scaling of Kooij et al. [27] for Newtonian liquids. Polymer addition to the water solvent results in larger values of \( D_{50} \). Fig. 4(b) suggests that the median drop size converges asymptotically towards the Newtonian scaling at high liquid velocity \( U_0 \). However, this can be partially due to mechanical degradation since, for a given polymer concentration and nozzle, the extensional relaxation time \( r \) decreases as \( U_0 \) increases, see Section 2.6.

3.2. Universal scaling of the median drop size

The zero-shear viscosity of polymer solutions used in spray experiments reaches 1.28 mPa s at 100 ppm, which corresponds to Ohnesorge numbers \( Oh = \eta_0/\sqrt{\rho \sigma} \) up to \( 7 \cdot 10^{-3} \) in our experiments. Hence, the increase in shear viscosity cannot account for the significant increase in median drop size observed in Fig. 4(b) since we know from Kooij et al. [27] that sprays of Newtonian liquids of viscosity up to 32.3 mPa s exhibit median drop sizes nearly identical to that of water sprays. The increase in \( D_{50} \) can hence be attributed to elastic effects, which we choose to quantify using the extensional relaxation time \( r \) measured from filament thinning (CaBER) measurements. Viscoelastic effects are expected to be significant when \( r \) is comparable to the inertio-capillary Rayleigh time scale \( \sqrt{\rho b^3/\sigma} \), i.e. when the Deborah number defined as

\[
De = \frac{\tau}{\sqrt{\rho b^3/\sigma}}
\]

is of order unity. Note that this definition is different from the local Deborah number considered by Keshavarz et al. [35] in air-assisted jet atomization, where the size of the ligaments was chosen as the characteristic length scale.

In order to bypass the polymer degradation problem discussed in Section 2.6 and obtain the true physical dependence of \( D_{50} \) on the Weber and Deborah numbers, we select data points from Fig. 4(b) corresponding to the same Weber number and plot the associated non-dimensional median drop size \( D_{50}/b \) against the Deborah number \( De \). We account for mechanical degradation by choosing the relaxation time of the degraded solution samples collected from the spray. We observe a linear dependence of \( D_{50}/b \) on \( De \) where the value at \( De = 0 \) corresponds to the Newtonian value. The slope of this linear dependence decreases with increasing Weber number. The inset in Fig. 5(a) shows that it follows a power law \( We^k \) with an exponent \( k \).
between −0.6 and −0.5. We can therefore write the following empirical formula for the median drop size:

$$D_{50}/b = c_1 a^{-1/6} (We)^{−1/3} + c_2 De We^{-k}$$

(7)

where we find $c_2 = 33$ for $k = 0.6$ and $c_2 = 14$ for $k = −0.5$. Note that $c_2$ is not necessarily a purely numerical prefactor as it might depend, for example, on the air–liquid density ratio $\alpha$. This expression reduces to Eq. (1) for Newtonian liquids ($De = 0$). We show in Fig. 5(b) that this empirical expression leads to a collapse of the data of Fig. 4(b) on a single master curve for $k = −0.5$. An equally good collapse is found for $k = 0.6$ and $c_2 = 33$, for all data of Fig. 4(b). The dashed line corresponds to Eq. (7) with $c_1 = 1.65$.

3.3. General expression of the median drop size

We now address the question of the physical origin of the increasing size of spray droplets with increasing liquid elasticity. To this end, we first derive a general expression of the characteristic drop diameter $D_{50}$ in terms of quantities that can be easily measured.

Fig. 6 provides a schematic representation of the sheet fragmentation mechanism in front view (a) and side view (b). We recall that the sheet fragmentation is initiated by the growth of Squire waves along the sheet. This flapping motion causes the sheet to break up at a radial distance $l$ from the nozzle, henceforth referred to as the breakup length, leaving sheet fragments which then retract into ligaments. The size of these sheet fragments is of the order of the wavelength $\lambda$ of the Squire instability. Hence, using mass conservation, the characteristic diameter $d_l$ of the ligaments should scale as

$$d_l \sim \sqrt{h l},$$

(9)

where $h(r)$ is the sheet thickness at a radial distance $r$ from the nozzle. If the liquid velocity is constant along the sheet, flow rate conservation gives

$$h = \frac{Q}{\pi w},$$

(10)

where $\xi = U_0/U_1$ is the ratio between the liquid velocity $U_0 = Q/A$ in the nozzle of area $A$ and the liquid velocity $U_1$ in the sheet.
where \( w \) is the length of the arc of a circle of radius \( r \) stopping at the sheet edges (see Fig. 6). We expect \( w = \theta r \) for a sheet expanding with a constant angle \( \theta \). This relation neglects the presence of rims at the edges of the sheet, which are observable in the front images of Fig. 1. Finally, the characteristic drop diameter \( D_{50} \) can be deduced from the characteristic ligament diameter using mass conservation, which gives

\[
D_{50} = d_i \left( \lambda_i / d_i \right)^{1/3},
\]

where \( \lambda_i \) is the characteristic distance between successive drops along the ligament. If drops are formed regularly along the ligament, \( \lambda_i \) is the wavelength of the Rayleigh–Plateau instability, which is \( \lambda_i / d_i \approx 4.55 \) for Newtonian liquids in the absence of viscous effects.

Eqs. (9)–(11) do not depend on the rheology of the liquid being sprayed. Therefore, we can measure how different quantities such as \( \lambda, l, \xi \) and \( w \) are affected by polymer addition and hence identify which effects play an important role in explaining the increase in drop size with increasing polymer concentration. Measurements of these quantities are reported in Sections 3.4–3.6.

### 3.4. Die swell

Viscoelastic liquids extruded from orifices are known to potentially expand in air due to the well known die swell effect [61,62]. In our experiments, this would cause spray sheets of polymer solutions to be thicker than water sheets, potentially leading to the formation of larger spray droplets. If die swell occurs, we expect to observe a reduction of the liquid velocity \( U_l \) in the sheet compared to the liquid velocity \( U_0 \) in the nozzle, as shown schematically in Fig. 6(b). Therefore, in order to verify if die swell occurs, we measure the radial liquid velocity \( U_l \) in the spray sheet by a particle tracking method, using silver-coated hollow glass spheres of diameter 100 \( \mu \)m as tracers.

We find that \( U_l \) is constant along the unbroken part of the sheet, as expected if gravitational acceleration is negligible, which is the case since \( U_l / \sqrt{g} > 10 \) in our experiments. \( U_l \) is plotted in Fig. 7 as a function of \( U_0 \) for sprays of water and of 25 and 100 ppm PEO-4M solutions formed using both nozzles. We find that \( U_l = U_0 \) for both water and the 25 ppm solution, which means that there is no swelling. For the 100 ppm solution however, we observe that \( U_l \) is smaller than \( U_0 \) with a constant shift of about 6 m/s, meaning that the velocity ratio \( \xi = U_0 / U_l \) decreases as \( U_0 \) increases. This is opposite to the trend expected from the elastic die-swell theory of Tanner [61,62]. This is most likely due to the reduction of the liquid elasticity with increasing flow rate caused by mechanical degradation, as discussed in Section 2.6.

We conclude from these measurements that die swell cannot be the main cause of the increasing spray droplet size since droplets of 25 ppm PEO-4M solutions are significantly larger than water droplets while no die swell is observed at this concentration, see Fig. 4(b).

The breakup length \( l \) also plays a role in setting the median drop size according to Section 3.3. In our experiments, values of \( l \) are estimated from front-view images of the sheet similar to Fig. 1 obtained from high-speed photography. Average values are obtained from 30 pictures for each set of parameters. Results are presented in Fig. 8(a) where \( l/b \) is plotted against the Weber number \( W_e = \rho U_l^2 b/\sigma \) based on the liquid velocity in the sheet, for sprays of water and of 25 and 100 ppm PEO-4M solutions formed using both nozzles. We observe that \( l \) increases significantly with polymer concentration.

For water, we note that experimental values of \( l \) in Fig. 8(a) are consistent with the inviscid Newtonian theory of Kooij et al. [27], which predicts

\[
l/b \sim a^{-2/3} W_e^{-1/3}.
\]

This is similar to the scaling of the radius of a circular flapping liquid sheet [63]. Surprisingly, the dependence on the Weber number is also close to a power law with exponent \(-1/3\) for the two polymer concentrations in Fig. 8(a), at least at high Weber numbers. However, this may not be the true physical \( W_e \) exponent. Indeed, in this case, the decrease of the sheet length with liquid velocity may be partly attributed to the reduction of the liquid elasticity caused by mechanical degradation, see Section 2.6. The evolution of \( l/b \) with the Deborah number is presented in appendix Appendix B.

According to Eqs. (9)–(11), an increase in breakup length alone, while other parameters remain constant, would lead to smaller droplets. A possible compensating effect is the narrowing of the sheet in the orthoradial direction observed for viscoelastic spray sheets, i.e. smaller values of \( w \) according to Eq. (10). This effect is illustrated in Fig. 8(b) in which we juxtapose 30 images of a spray of water (left half) and of 25 ppm PEO-4M solution (right half), corresponding to the same nozzle and flow rate, to obtain a better visualization of the edges of the spray sheet. This figure reveals that the water sheet expands linearly with a constant angle \( \theta \), i.e. \( w(r) = \theta r \), while the viscoelastic sheet exhibits a concave profile with \( w(r) < \theta r \). This effect is most likely a consequence of the elastic stresses arising from the stretching of polymer molecules caused by the sheet expansion. However, measurements of the \( w(r) \) profile reveal that the value of \( w(r = l) \) actually increases with polymer concentration because of the significant increase in breakup length.

Using the measured values of \( \xi \) and of \( w(l) \), we find that the sheet thickness \( b(l) \) at breakup, calculated using Eq. (10), does not increase significantly with increasing polymer concentration. Hence, none of the effects discussed so far can explain the increase in spray droplet size.
with Eq. (13), we find that experimental values are underpredicted by a factor up to 2 at high liquid velocities.

3.7. Physical origin of the increase in drop size

We can now check if the increase in Squire wavelength accounts for the increase in droplet size with increasing viscoelasticity. As Eq. (9) predicts that the ligament diameter scales with $\sqrt{\lambda h(l)}$, in Fig. 10(a), we plot the median drop size $D_{50}$ against $\sqrt{\lambda h(l)}$, where $\lambda$ is the measured Squire wavelength and where the sheet thickness $h(l)$ at breakup is calculated from Eq. (10) with measured values of $\xi$ and $\omega(l)$. We obtain a relatively good collapse of data points for the different liquids and nozzles, which would not have been possible if the increase of $\lambda$ with polymer concentration had been neglected. Hence, the increase in Squire wavelength is a key effect for explaining the increase in droplet size. For comparison, we find that the effect of elasticity on $h(l)$ is less important since assuming $h(l) \sim A/l$ has less impact on the quality of the collapse.

However, we also find that $D_{50}$ is only proportional to $\sqrt{\lambda h(l)}$ for water in Fig. 10(a), while the linear fit obtained for water underestimates the droplet size by a factor of up to 2 for polymer solutions exhibiting the largest relaxation times. To investigate this further, we measure the mean diameter $\langle d_l \rangle$ of the ligaments formed in the sheet breakup region from high-speed images of the spray. In Fig. 10(b) we plot $\langle d_l \rangle$ against $\sqrt{\lambda h(l)}$ for water and polymer solutions. The data reveal that $\langle d_l \rangle$ is proportional to $\sqrt{\lambda h(l)}$ with a numerical prefactor 0.48, consistent with Eq. (9). Therefore, according to Fig. 10(a), the ratio $D_{50}/\langle d_l \rangle$ increases with increasing viscoelasticity.

We conclude that the increase in droplet size in ligament-mediated fragmentation of a viscoelastic sheet is partially explained by an increase in ligament diameter, caused by an increase of the Squire wavelength, which sets the size of the sheet fragments detached from the sheet. The discrepancy between droplet and ligament diameters observed for increasing viscoelasticity is likely caused by an increase in the distance $\lambda_l$ between droplets along a ligament, resulting in the formation of fewer, bigger droplets according to Eq. (11). This idea is supported by the inset pictures of Fig. 10(b) where we measure $\lambda_l/d_l \approx 7$ for the 25 ppm PEO-4M solution (bottom) while we measure $\lambda_l/d_l \approx 3$ for water (top).

3.8. Droplet size distribution

After having discussed the increase in median droplet size with increasing viscoelasticity, we now turn our attention to the distribution of droplet sizes in the spray. Droplet size distributions are presented in Fig. 11(a) in terms of normalized probability density functions (PDFs)
Eq. (14) reduces to the gamma distribution respectively. In the limit $m$, $n$ formed) and the width of the ligament diameter distribution, respectively. The parameters $\psi$ where $\gamma$ is the modified Bessel function of second kind and $\Gamma$ is the gamma function. The parameters $m$ and $n$ correspond to the corrugation of the ligaments formed during sheet breakup (from which droplets are generated) and the width of the ligament diameter distribution, respectively. In the limit $m \to \infty$, where all ligaments have the same diameter, Eq. (14) reduces to the gamma distribution

$$
\Gamma(n, x = \frac{D}{\langle D \rangle}) = \frac{\Gamma(n)}{\Gamma(m)} x^{\frac{m-n}{2}} e^{-\frac{x^2}{2}}.
$$

which corresponds to a fragmentation–coalescence scenario [1]. Large values of $n$ correspond to smooth ligaments producing droplets of the same size.

We find that the PDFs in Fig. 11(a) are best fitted by the compound gamma distribution of Eq. (14) with values $m = 15$ and $n = 6$ for water and with lower values $m = 4$ and $n = 4$ for the highest PEO concentrations. This means that the presence of polymers results in the formation of more corrugated ligaments with a broader ligament diameter distribution. The latter statement is confirmed by direct measurements of the ligament diameter distribution which are in good agreements with the values of $m$ extracted from the drop size distributions, see appendix Appendix C.

The same low values of $m = 4$ and $n = 4$ are also found to yield the best fit of the PDFs in Fig. 11(b), corresponding to different operating pressures for a fixed polymer concentration (50 ppm) for both flat fan nozzles. The data in this figure cover Weber numbers $We$ ranging from 3000 to 9000 and a range of Deborah numbers $De$ from 0.15 to 0.5. The solid line corresponds to the best linear fit with slope 0.48. The sprays are made of water and PEO-4M solutions of various concentrations obtained from the Teejet 110-02 nozzle at an operating pressure of 3 bar. The dashed and solid lines correspond to the best fits of the compound gamma distribution (Eq. (14)) for water ($n = 4$, $m = 15$), respectively. (b) Droplet size distributions for sprays of a 50 ppm PEO-4M solution generated at different operating pressures from both nozzles, covering a range of Weber numbers $We$ from 3000 to 9000 and a range of Deborah numbers $De$ from 0.15 to 0.5. The solid line corresponds to the best linear fit for water with slope 0.70. (b) Mean diameter $\langle d_l \rangle$ of the ligaments formed during sheet breakup (from which droplets are generated) at different operating pressures from both nozzles. Insets in figure (b) show ligaments and droplets for a spray of water (top) and 25 ppm PEO-4M solution (bottom) extruded at pressures 1 and 2 bars respectively.
produced from various fragmentation processes, saturates with increasing viscoelasticity to a low value $n = 4$. The authors show that this value corresponds to a geometrical limit which cannot be exceeded in ligament-mediated fragmentation phenomena. The authors report experimental PDFs which are well fitted by the gamma distribution \((15)\) with \(n = 4\) for three different fragmentation processes: air-assisted jet atomization, oblique jet impact atomization and droplet impact on a small target (no flat fan nozzles). This means that ligaments were monodisperse in their experiments \((n \to \infty)\). However, this is not the case in our measurements using flat fan nozzles, as illustrated in Fig. 11(b) where a gamma distribution with \(n = 4\) is shown to significantly underpredict the width of droplet size distribution. This discrepancy shows that the droplet size distribution obtained from the fragmentation of viscoelastic liquids depends on the details of the fragmentation process: different spraying techniques may lead to different widths of ligament size distributions, i.e. different values of \(m\).

4. Conclusion and discussion

The impact of viscoelasticity on sprays produced by agricultural flat fan nozzles was investigated experimentally using dilute aqueous solutions of high-molecular-weight polyethylene oxide. The size distribution of spray droplets was measured by laser diffraction at a distance from the nozzle where ligaments had fully broken up into independent droplets. As the polymer concentration was increased, we observed the formation of droplets with a higher median size and a broader size distribution. The median droplet size \(D_{50}\) was found to increase linearly with the extensional relaxation time \(\tau\) of the polymer solution and to be well captured by a simple empirical expression in terms of non-dimensional Weber and Deborah numbers. The decrease of the relaxation time with increasing liquid velocity for a given polymer concentration, which was due to increasing mechanical degradation, was taken into account.

Measurements of different quantities involved in the sheet fragmentation mechanism revealed that the increase in droplet size with increasing polymer concentration is partly due to the increase of the wavelength of the Squire waves responsible for the sheet breakup; the thickness of the sheet calculated at the point of breakup was not significantly altered by polymer addition. The larger wavelength leads to the formation of larger ligaments from the sheet breakup. However, our results show that this mechanism does not fully explain the measured increase in droplet size, suggesting the existence of a second effect of viscoelasticity in the breakup of the ligaments themselves.

The increase in the Squire wavelength \(\lambda\) may come from an additional elastic tension \(T\) in the sheet arising from the stretching of polymer molecules, which would imply rewriting equation \((13)\) as \(\lambda \sim (\sigma + T)/\rho \sqrt{\gamma h}\). The growth rate \(\omega \sim U_1 \sqrt{\gamma / \sqrt{\lambda h}}\) of the Squire instability would decrease accordingly, explaining the increase in the breakup length \(l \sim U_1/\omega\). This elastic tension can in principle be calculated by integrating the normal component \(\sigma_1\) of the stress tensor over the sheet thickness \(h\), i.e. \(T = \int \sigma_1\) (Villermoux [21] suggested using an expression from the theoretical work of Rallison & Hinch [66] (see also Azizie & Homys [67] (appendix by J. Hinch)) on the instability of a high-speed submerged viscoelastic sheet of constant thickness \(h = b\) with a Poiseuille base flow and \(\sigma_1 = 2G \gamma^2 \gamma^2\), where \(G\) and \(\gamma\) are the elastic modulus and relaxation time in the Oldroyd-B model, respectively, and where \(\gamma\) is the local shear rate. In this work, the authors find that the instability's growth rate and wavenumber decrease with increasing elasticity number \(EI = A\gamma b/\rho \sqrt{U_1}\) which, in our experiments, is up to a value of 0.01 for which the theory predicts almost no difference with the case \(EI = 0\). However, in our experiments, this shear-dominated tension is most likely not relevant since we expect to have a plug flow in the sheet. Indeed, the thickness \(\sqrt{\rho \gamma U_1}\) of the momentum diffusion layer at the liquid–air interface reaches half of the sheet thickness \(h \approx A/\sqrt{\gamma}r\) at a distance \(r\) from the nozzle which is 3 to 10 times shorter than the breakup length \(l\). Besides, the flow in the nozzle of thickness \(b\) is not a Poiseuille flow since the length over which the liquid is sheared is only about 0.3 mm \(\ll \rho U_0 b^2/\eta\). The elastic stress responsible for the increase of the Squire wavelength is hence more likely to arise from the contraction flow in the nozzle and from the radial flow in the sheet, two extensions flows with extension rates \(\dot{v}\) scaling as \(U_0/b\) and \(U_1/r\), respectively. Note that the contribution of the contraction flow is expected to partially relax in the sheet since \(r U_1/\dot{v} < 0.4\) in our experiments. The elastic tension \(T\) is then expected to increase with liquid velocity, meaning that the elastic limit \(T \gg \sigma\) would correspond to large Weber numbers. This is in contradiction with our empirical formula \((8)\) which suggests that the elastic limit \(D_{50} > D_{50,N}\) corresponds to low Weber numbers since \(k + 1/3 < 0\). A theory for the increase of droplet sizes is sprays of polymer solutions is hence still missing.

When rescaled by the average drop size, the measured droplet size distributions are well described by a compound gamma distribution where the parameters \(n\) and \(m\), encoding for the ligament corrugation and the width of the ligament size distribution, respectively, saturate to values \(n = 4\) and \(m = 4\) at sufficiently high polymer concentrations. We can therefore now relate the changes in drop size and its distribution to the presence of polymers, and notably the polymer relaxation time. This then gives a handle for controlling sprays using polymer additives; one important application of this is spray drift reduction in e.g. shipyard or agricultural spraying, where highly toxic anti-fouling or pesticide molecules in small drops are prone to drift away with the wind.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. \(D_{50}\), \((D)\) And SMD

Three characteristic droplet sizes are usually reported in the literature: the median \((D_{50})\) and mean \((\langle D \rangle)\) diameters as well as the Sauter mean diameter \(SMD = \langle D^3 \rangle/\langle D \rangle^2\). These three quantities are extracted from the experimental droplet size distributions and are compared in Fig. A.12 for sprays of water and of polymer solutions. Fig. A.12(a) shows that \(D_{50}\) is fairly proportional to \((D)\). This is not surprising since, as we show in Section 3.8, droplet size distributions are fairly well captured by compound gamma distributions with values of \((n, m)\) ranging between \((6, 15)\) (water) and \((4, 4)\) (polymer solutions), for which \(D_{50}/(D)\) ranges between two close values \(0.91\) and \(0.81\). Experimentally, we find a lower value \(D_{50}/(D) \approx 0.77\), indicating a deviation from Eq. \((14)\). As shown in Fig. A.12(b), values of SMD/(D) increase with polymer concentration for any given operating pressure, consistent with a broadening of the distribution, in agreement with the values \(1.51\) to \(2.24\) expected for compound gamma distributions with values of \((n, m)\) going from \((6, 15)\) to \((4, 4)\).

Appendix B. Sheet length and square wavelength vs. \(De\)

In Fig. A.13, we show the evolution of the non-dimensional sheet length \(l/b\) (a) and of the Squire wavelength \(\lambda\) (b) with the Deborah number for fixed values of the Weber number. Both quantities increase with \(De\), clearly non linearly for \(l/b\). More data points would be needed to extract satisfying empirical expressions in terms of Weber and Deborah numbers.
Fig. A.12. (a) Median drop size $D_{50}$ against the mean droplet size $⟨D⟩$ and (b) Sauter mean diameter (SMD) rescaled by $⟨D⟩$ against the polymer concentration for sprays of water and of PEO solutions obtained from both nozzles (only Teejet 110-02 in (b)) at different operating pressures. Values of SMD$/⟨D⟩$ are compared with the values 1.51 and 2.24 expected from compound gamma distributions (Eq. (14)) with $(n, m) = (6, 15)$ and $(n, m) = (4, 4)$ respectively.

Fig. A.13. (a) Non-dimensional sheet length $l/b$ and (b) Squire wavelength $\lambda$ against the Deborah number $De$ for four sets of data points. Within each set, the Weber number $We_1 = \rho U_1^2 b/\sigma$ is the same to within up to 7%.

Fig. A.14. Distributions of ligament diameters $d_l$ rescaled by the mean ligament diameter $⟨d_l⟩$ for a spray of water and of 100 ppm PEO-4M solution obtained from the Teejet 110-02 nozzle at an operating pressure of 3 bar. The dashed and dashed–dotted lines correspond to gamma distributions (Eq. (15)) with $n = 15$ for water and $n = 4$ for the PEO solution, respectively.

Appendix C. Ligament size distribution

In Fig. A.14, we show two probability density functions (PDF) for the distribution of ligament diameters $d_l$ which were measured from high resolution images of the spray. These two PDFs correspond to a spray of water and of 100 ppm PEO solution obtained from the same nozzle and operating pressure as in Fig. 11(a). We find that the distribution of ligament sizes is broader for the PEO solution and that both PDFs are well described by gamma distributions (Eq. (15)) with $n = 15$ for water and $n = 4$ for the PEO solution, which are the values of $m$ found in Fig. 11(a).

References
