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**The matching of polymer solution fast filament stretching, relaxation and break up  experimental results with 1D and 2D numerical viscoelastic simulation.**

**by**

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**Abstract**

This paper is concerned with the comparison of two numerical viscoelastic strategies for predicting the fast filament stretching, relaxation and break up of low viscosity, weakly elastic polymeric fluids. Experimental data on stretch, relaxation and breakup was obtained using a Cambridge Trimaster for a Newtonian solvent (DEP) and three monodisperse polystyrene polymer solutions. Two numerical codes were tested to simulate the flow numerically. One code used a 1D approximation coupled with the Arbitrary Lagrangian Eulerian (ALE) approach and the other, a 2D axisymmetric approximation for the flow. In both cases the same constitutive equations and mono and multimode parameter fitting was used; thereby enabling a direct comparison on both codes and their respective fit to the experimental data. Both simulations fitted the experimental data well and surprisingly the 1D code closely matched that of the 2D. In both cases it was found necessary to utilise a multimode approach to obtain a realistic match to the experimental data. The sensitivity of the simulation to the choice of constitutive equation (Oldroyd-B and FENE-CR) and the magnitude of non linear parameters were also investigated. The results are of particular relevance to ink jet processing and demonstrate that high shear rate, low viscosity viscoelastic polymeric flows can be simulated with reasonable accuracy.

**1.Introduction**

The way in which viscoelastic fluids stretch, thin and break up is of relevance to a number of technologies and these three aspects of the flow have in the past received extensive scientific attention; although generally as three different individual topics. The stretching of polymeric fluids in particular has received detailed experimental and modeling attention in the last decade from amongst others (Anna and McKinley

44　(2001), McKinley and Sridhar (2002), Bach *et al*. (2002), Clasen *et al*. (2006)) where

45　the work has concentrated on determining the transient extensional viscosity of fluids.

46　The thinning of prestretched polymeric fluids has also been investigated

47　experimentally following pioneering experimental work by (Bazilevsky *et al.*, 1997)

48　which was subsequently modelled by (Entov and Hinch (1997)). A review by

49　McKinley (2005a) gives an authoritative account of factors that influence filament

50　thinning behaviour. Filament breakup is a delicate process and is the least well

51　characterized and modelled of the three topics amongst stretching, relaxation and

52　breakup covered by this paper.

53　Ink jet printing can involve all three elements considered above during filament

54　formation and droplet breakup (Dong *et al.* (2006), Hoath *et al.* (2009), Jang *et al.*

55　(2009)). In order to mimic elements of this complex deformation process a

56　"Cambridge Trimaster" geometry apparatus was developed specifically as a device to

57　capture aspects of the process with well-defined boundary conditions (Vadillo *et al.*

58　(2010a)). The Cambridge Trimaster has strong similarity to the single piston Rozhkov

59　filament thinning device (Bazilevsky *et al.* (1990) and the Haake Caber filament

60　thinning apparatus (http://www.thermo.com/com/cda/product/detail/). The twin piston

61　Trimaster was developed specifically for low viscosity fluids with a fast, controlled

62　initial displacement and for use with high speed photography [Vadillo *et al.*(2010a)].

63　Characterisation of low viscosity, linear viscoelasticity with short relaxation times is a

64　challenging area of rheology, however the Pechold, Piezo Axial Vibrator (PAV)

65　(Groß *et al.* (2002), Kirschenmann (2003), Crassous *et al.* (2005), and Vadillo *et al.*

66　(2010b)) is an apparatus that can probe fluid within the range of millisecond

67　relaxation times. Thus by using a combination of the Cambridge Trimaster and the

68　PAV it was possible to probe both the extensional filament break up behaviour of

69  viscoelastic fluids that are well characterized, at least in the Linear Viscoelastic

70  (LVE) regime using the PAV.

71  In a recent work, some authors of this paper have published the matching of

72  experimental and simulation filament stretching and thinning data using the single

73  mode Maxwell description for the viscoelastic contribution of the fluid (Tembely *et*

74  *al.* (2012). The results were promising, although all the elements of the Trimaster data

75  with a single mode 1D simulation of the process of thinning and break up could not be

76  fully captured. A direct comparison between 1D and 2D modeling may be found in

77  the work of Yildirim and Basaran (2001) and more recently by Furlani and Hanchak

78  (2011). The latter authors have used the slender jet 1D approximation and solved the

79  nonlinear partial differential equations using the method-of lines wherein the PDEs

80  are transformed to a system of ordinary differential equations for the nodal values of

81  the jet variables on a uniform staggered grid. The 1D results are impressive with the

82  key advantages being the ease of implementation and the speed of computation albeit

83  in a different configuration than the problem considered in this paper. In the present

84  paper, Trimaster data for polymer solutions are matched to single and multimode

85  viscoelastic simulation data, using both a computationally time efficient 1D

86  simulation and a potentially more rigorous 2D simulation. The paper represents a

87  "state of art" position in matching extensional time dependent results with high level

88  numerical simulation, thereby enabling the effects of constitutive equation and

89  constitutive parameters to be tested.

90  **2.Test fluids, rheological characterisation and Trimaster experimental protocols.**

91  **2a Test fluid preparation and characterisation.**

4

92 The fluids used were a series of mono-disperse polystyrene dissolved in diethyl

93 phthalate (DEP) solvent as previously described in [Vadillo *et al.*, 2010]. Near mono

94 disperse Polystyrene polymer was manufactured specially by Dow, and gel

95 permeation chromatography (GPC) with THF as the solvent enabled determination of

96 mass and number average molecular weights $M_w$ and $M_n$ as 110 kg/mol and

97 105kg/mol respectively. A stock solution of PS dilution series was prepared by adding

98 10wt% of PS to the DEP at ambient temperature. The resulting solution was heated to

99 180°C and stirred for several hours until the polymer was fully dissolved. The

100 dilution series were prepared by subsequent dilution of the respective stock solutions.

101 Sample surface tension remained constant at 37mN/m up to 10wt% PS110

102 concentration and with a critical polymer overlap concentration c* of 2.40wt%

103 [Clasen *et al.* (2006a)]. The zero shear viscosities $\eta_0$ of the solutions were determined

104 from PAV low frequency complex viscosity $\eta^*$ data within the terminal relaxation

105 regime and the measured viscosities are given in Table I.

106 **Table I:** Zero shear rate complex viscosity of the different polymer solutions at 25°C

107 **2b. Rheological characterisation.**

108 The Piezo Axial Vibrator (PAV) has been used to characterise the linear viscoelastic

109 behaviour of samples with viscosity has low as 1mPa.s on a frequency range

110 comprised between 0.1Hz and 10000Hz [Groß *et al.* (2002); Kirschenmann (2003);

111 Crassous *et al.* (2005); Vadillo *et al.* (2010b The PAV measures the complex

112 modulus $G^*$ of the test fluid with $G^* = G' + iG''$ and where $G'$ is the storage modulus

113 and $G''$ is the loss modulus. The complex viscosity $\eta^*$ is related to the complex

114 modulus by $\eta^* = G^*/\omega$ where $\omega$ is the angular frequency. Experimental LVE results

115 are presented in Fig. 1. Loss modulus G'' and elastic modulus G' have been found to

116    increase with the frequency and to vertically shift with the polymer addition. Note, the

117    pure DEP solvent does not show any G'.  Both moduli approach at lower frequencies

118    the terminal relaxation regime with the expected scaling with a power of 1 for the loss

119    modulus (Fig. 1.a), and a power of 2 for the storage modulus (Fig. 1.b), and a constant

120    complex viscosity $\eta^*$ in this regime as shown in Fig. 1.c (except for 5wt% PS110 after

121    2000Hz).  The experimental results are displayed between $10^2$ and $10^4$Hz, the range

122    on which the storage modulus has been captured.  At lower frequency, the fluids have

123    been found essentially to behave as a Newtonian fluid with the presence of a loss

124    modulus only.

## 2c. Cambridge Trimaster experimental protocol

126    The Cambridge Trimaster (CTM) is a Capillary Breakup Extensional Rheometer that

127    has been specifically designed to probe the extensional rheology of weakly

128    viscoelastic fluids.  This apparatus performs a fast stretch of a cylinder of fluid

129    initially located between two identical pistons over a short distance.  This apparatus

130    and its limitation have been presented in details in [Vadillo *et al*. 2010a]. In the

131    present study, the piston diameters are 1.2mm and the experimental filament

132    stretching conditions are an initial gap size $L_0$ of 0.6mm and a stretching distance $L_f$

133    of 0.8mm at a relative piston speed $2V_p$ of 150mm/s.  This corresponds to a filament

134    strain rate $2V_p/L_0 = 250$ s$^{-1}$ and a filament aspect ratio $L_f/L_0$ of 2.3.  The piston

135    velocity and stretching distance have been chosen to ensure that pistons stop their

136    motion before the critical time scale for inertio-capillary break up for the sample with

137    the lower viscosity, here the DEP.  For such a fluid, this time scale has been estimated

138    around 5ms [Tembely *et al.*, 2012].    These conditions have been used in the

139    following for both experiments and simulations.

140  The transient filament profiles were captured using a Photron Fastcam

141  (http://www.photron.com/index.php?cmd=product_general&product_id=1) 1024 PCI

142  high speed camera at 6000 fps, for a picture size of 128 x 256 with a shutter time of

143  3µs. The filament thinning measurement, as well as the filament breakup behaviour,

144  was obtained using automatic image processing based of greyscale variation

145  throughout image for edge detection and the minimum diameter that can be resolved

146  was about ~ 6µm.

**2d Relaxation time and moduli determination.**

148  Relaxation spectrum determination from LVE measurements is an ill-posed problem

149  and has been studied extensively in the literature [see for example Baumgaertel and

150  Winter (1989); Kamath *et al.* (1990), Stadler and Bailly (2009)] and different

151  techniques from linear to non-linear regression have been developed to obtain

152  relaxation spectra from oscillatory LVE data. In the modelling carried out here, a

153  series of equidistant relaxation times spaced on the logarithmic scale was chosen with

154  one mode per decade. This was motivated by the fact that, in experiments, low visco-

155  elastic fluids have shown significant differences between relaxation times in shear and

156  in extension [Clasen *et al.* (2006)] and recent simulations have shown that using a

157  single mode Maxwell description of the fluid was not sufficient [Tembely *et al*.

158  (2012)] to capture those differences. The minimization program for both G' and G''

159  data was solved using Matlab®. The solution involved the use of SQP (Sequential

160  quadratic programming) [Jorge and Wright (2006)] methods which may be considered

161  as a state of the art nonlinear programming optimization technique. This method has

162  been shown to outperform other methods in terms of accuracy, efficiency, and

163  adaptability over a large number of problems [Schittkowski (1985)] and it is an

164 effective method for non-linear optimization with constraints. In each iteration the

165 non-linear problem was approximated using a quadratic which is easy to solve (hence

166 the name SQP).

167 The conversion of the experimental data (G'$_m$, G''$_m$, $\omega_j$) into a relaxation function was

168 performed by expressing G(t) as a discrete relaxation spectrum ($g_i$, $\lambda_i$). The Maxwell

169 model relates the real and imaginary parts of the complex modulus determined in

170 LVE measurement to the discrete relaxation spectrum of $N$ relaxation times $\lambda_i$ and a

171 relaxation strengths $g_i$ through:

172
$$G'(\omega) = \sum_{i=1}^{N} g_i \frac{(\omega\lambda_i)^2}{1+(\omega\lambda_i)^2} \tag{1}$$

173
$$G''(\omega) = \eta_s\omega + \sum_{i=1}^{N} g_i \frac{\omega\lambda_i}{1+(\omega\lambda_i)^2} \tag{1}$$

174 with $\omega$ being the angular frequency of the experiment, and $N$ is the number of

175 relaxation modes. As indicated in (2), G'' accounts for the solvent viscosity.

176 Generally the spectra can be computed by minimizing the "least mean square error"

177 as follows [Armstrong *et al.* (1987); Curtiss *et al.* (1987); Stadler and Bailly (2009)]:

178
$$D = \sum_{j=1}^{M} \left[ \frac{G'(\omega_j)}{G'_m(\omega_j)} - 1 \right]^2 + \left[ \frac{G''(\omega_j)}{G''_m(\omega_j)} - 1 \right]^2 \tag{2}$$

179 where $M$ is the number of measurements.

180 The model was initialized by choosing the relaxation times to be equidistantly spaced

181 on a logarithmic scale such that $\log(\lambda_i / \lambda_{i+1}) = 1/p$. Setting p = 1, i.e, one mode per

182 decade, has been found to provide sufficient accuracy to accurately describe the LVE

183 behavior (Fig. 1). In the numerical simulation, the Maxwell component of the model

184 was fitted with 5 modes. The relaxation times are chosen such that G' and G''

185 measured over the frequency range $\omega_{min} < \omega < \omega_{max}$ recover all the information

186 regarding the relaxation spectrum over the range $1/\omega_{max} < \lambda_i < 1/\omega_{min}$, however the

187 correct range is given by $e^{\pi/2}/\omega_{max} < \lambda_i < e^{-\pi/2}/\omega_{min}$ [Davies and Anderssen (1997)].

188 This spectrum is a generalized form of the Maxwellian dynamics [Ferry (1980)] and

189 shown in Table II.

190 **3**. **General equations and numerical simulations.**

191 Numerical simulations of the Trimaster deformation were performed using both a

192 one-dimensional model and a 2D axisymmetric model. In the following sub-sections

193 the general equations and the numerical techniques used in both cases are detailed.

194 **3a. Flow geometry.**

195 To model the experimental conditions, an initial cylindrical column of fluid was

196 considered bounded by two rigid circular pistons of diameter $D_0$. The fluid and the

197 pistons were initially at rest; subsequently the pistons moved vertically outwards with

198 time-dependent velocities $V_p(t)$ (top piston) and $-V_p(t)$ (bottom piston), which are

199 prescribed functions based on fitting a smooth tanh curves through measurements of

200 the Trimaster piston motion in the experiments. As described in Tembely et al 2011,

201 the form of tanh has been chosen to fit the symmetrical "S" shape experimentally

202 observed for the piston motion with time. In that work, the authors have shown that

203 the use of an accurate representation of the piston dynamic response is of importance

204 in the simulation of fast transient dynamic of low viscosity and/or low viscoelasticity

205 fluids.

206 Using a cylindrical coordinate system $\{r, \theta, z\}$, the flow was constrained to be

207 axisymmetric so that all flow fields are independent of the angular coordinate $\theta$, and

208 the simulation may be restricted to the $rz$-plane.  The coordinate origin is at the axis of

209 the jet, midway between the initial positions of the two pistons.  Fig. 2 shows a

210 schematic diagram of the computational domain at an intermediate stage of the piston

211 motion.

212 Symmetric boundary conditions are required along the $z$-axis to maintain

213 axisymmetry, and conditions of no-slip were applied at each piston surface.  The

214 boundary conditions at the free surface are those of zero shear stress and the

215 interfacial pressure discontinuity due to the surface curvature

216 $$\mathbf{t}.\underline{\underline{\mathbf{T}}}.\mathbf{n} = 0 \quad \text{and} \quad \left[\underline{\underline{\mathbf{T}}}.\mathbf{n}\right]_{air}^{fluid} = -\gamma\kappa ,\tag{3}$$

217 where $\underline{\underline{\mathbf{T}}}$ is the total stress tensor, $\mathbf{n}$ is the unit vector normal to the free surface

218 (directed outward from the fluid), $\mathbf{t}$ is the unit tangent vector to the free surface in the

219 rz-plane, $\gamma$ is the coefficient of surface tension, and $\kappa$ is the curvature of the interface.

220 It is assumed that the external air pressure is a negligible constant.

221 The location of the free surface at each time-step was determined implicitly via a

222 kinematic condition.   In the axisymmetric simulations, this was realized

223 automatically, since the mesh is Lagrangian and the mesh nodes are advected with the

224 local fluid velocity.  The contact lines between the free surface and the pistons were

225 held pinned at the piston edges throughout.

226 The initial conditions are that the fluid is at rest ($\mathbf{v}=\mathbf{0}$) and the polymer is at

227 unstretched equilibrium ($\mathbf{A}_i=\mathbf{I}$).

228 **3b. Governing equations**

229     The governing equations for incompressible isothermal flow of a viscoelastic fluid are

230     the classical Navier-Stokes equations for Newtonian fluids together with an additional

231     viscoelastic term coming from the extra stress tensor $\underline{\underline{\sigma}}$. The momentum conservation

232     then may be expressed as follows in which the 3$^{rd}$ term on right-hand-side accounts

233     for viscoelasticity:

$$\rho \frac{d\mathbf{v}}{dt} + \rho(\mathbf{v}.\nabla)\mathbf{v} = -\nabla p + \eta_s \nabla^2 \mathbf{v} + \nabla.\underline{\underline{\sigma}} + \rho g \mathbf{z} \tag{4}$$

235     and the continuity equation reads:

$$\nabla.\mathbf{v} = 0 \tag{5}$$

237     where $p$ is the fluid pressure, $\rho$ is the fluid density, $\eta_s$ is the solvent viscosity, and $g$ is

238     the acceleration due to gravity.

239     **3c. Constitutive equations**

240     For the viscoelastic fluid models, the polymer contribution was described by a

241     Finitely Extensible Nonlinear Elastic (FENE) dumbbell model which makes use of

242     the conformation tensor **A**, and the stress tensor reads [see for example, Chilcott and

243     Rallison (1988)]:

$$\sigma = Gf(R)(\mathbf{A} - \mathbf{I}) \tag{6}$$

245     where $G$ is the elastic modulus, $f(R)$ is the finite extensibility factor related to the

246     finite extensibility parameter $L$, representing the ratio of a fully extended polymer

247     (dumbbell) to its equilibrium length and R = Tr(A). $L$ can be described in terms of

248     molecular parameters as:

11

$$L = \sqrt{3} \left[ \frac{j\left(\sin{\theta}/{2}\right)^2 M_w}{C_\infty M_u} \right]^{1-\nu}$$  (7)

For a multimode model, the extra stress may be expressed as a sum of contributions from each mode. For the generalized multimode problem with *N* modes, each mode (*i*) with partial viscosity ($\eta_i$) and relaxation time ($\lambda_i$), and the extra-stress tensor of the FENE-CR expresses:

$$\underline{\underline{\sigma}} = \sum_{i=1}^{N} g_i f_i(R_i)(\mathbf{A}_i - \mathbf{I}),$$  (8)

where $f_i(R_i) = 1/\left(1 - R_i/L_i^2\right)$ with $R_i = \mathrm{Tr}\left(\mathbf{A}_i\right)$. For simplicity, it is assumed that the extensibility $L_i = L$ is constant, but other approaches may be used [Lielens *et al.* (1998)]. The dimensionless evolution equation for the $i^{th}$ mode is

$$\frac{d\mathbf{A}_i}{dt} = -\frac{f_i(R_i)}{\mathrm{De}_i}(\mathbf{A}_i - \mathbf{I}),$$  (9)

Where $\overset{\triangledown}{\mathbf{A}}_i = \frac{d\mathbf{A}_i}{dt} - \nabla\mathbf{v}^T_i.\mathbf{A}_i - \mathbf{A}_i.\nabla\mathbf{v}_i$ is the Oldroyd upper-convected time derivative of $A_i$, and $De_i$ is the Deborah number for the $i^{th}$ mode defined as follow

$$\mathrm{De}_i = \lambda_i/\tau$$  (11)

12

269    $g_i$ and $\lambda_i$ are the modulii and relaxation times described by the multimode

270    optimization see sub-section (2d) and where $\tau$ is the characteristic inertio-capillary

271    time scale of the system defined by $\tau = \sqrt{\rho R_0^3 / \gamma}$.

272    Scaling was performed using the piston radius $R_0$ as a length scale, and a

273    characteristic speed U as a velocity scale , where U is the average piston speed in the

274    2D case, and $U = R_0 / \tau$ in the 1D case. The time was scaled by $R_0 / U$ and $\tau$, in the 2D and

275    1D cases respectively; whereas pressures and stresses were scaled by $\rho U^2$. The

276    scalings yielded the dimensionless governing equations:

$$\frac{d\mathbf{v}}{dt} + (\mathbf{v}.\nabla)\mathbf{v} = -\nabla p + \frac{1}{\mathrm{Re}}\left(\nabla^2 \mathbf{v} - \sum_{i=1}^{N} c_i \nabla \overset{\nabla}{A_i}\right) + \frac{1}{\mathrm{Fr}^2}\mathbf{z}, \qquad \nabla \cdot \mathbf{v} = 0, (10)$$

278    where $t$, $\mathbf{v}$, and $p$ are now the dimensionless time, velocity, and pressure

279    respectively. For each viscoelastic mode an additional parameter $c_i = g_i \lambda_i / \eta_s$ has been

280    introduced: it may be interpreted as a measure of the concentration (volume fraction)

281    of dumbbell molecules corresponding to the $i^{\text{th}}$ mode. With the particular scalings

282    used here, the flow is characterized by the dimensionless groups Re We, and Fr,

283    which are respectively the Reynolds, Weber, and Froude numbers

$$\mathrm{Re} = \frac{\rho U R_0}{\eta_S}, \quad \mathrm{We} = \frac{\rho U^2 R_0}{\gamma}, \quad \mathrm{Fr} = \left(\frac{U^2}{gR_0}\right)^{1/2},$$

(13)

285    in addition to the Deborah number $\mathrm{De}_i$ for each mode, defined earlier. ==The Reynolds==

286    ==number represents the competition between inertia and viscosity, the Weber number==

287    ==the competition between the inertia and the surface tension while the Froud number==

288    ==represents the competition between inertia and gravity effects.==

289    Another important dimensionless number is that of Ohnesorge, $\mathrm{Oh} = \eta_S / \sqrt{\rho \gamma R_0}$. With

290    the scalings used here, the Ohnesorge number can be expressed in terms of the Weber

291 and Reynolds numbers: $Oh = \sqrt{We} / Re$. Alternative choices of scaling may result in

292 other different dimensionless groupings [Eggers and Villermaux, (2008)] as for

293 example, the Capillary number (ratio between viscous forces and surface tension) and

294 the Bond number (ratio between gravitational forces and surface tension). The Bond

295 number and the Capillary number have been estimated at ~0.11 and between 0.04 and

296 0.28 respectively indicating that surface tension is the dominating force and the

297 gravitational effects negligible. An extensive discussion of dimensionless number of

298 the problem can be found in [McKinley, 2005b].

**3d. Computational methods**

**1D simulation**

301 The previous equations (4), (5), (6) can be further simplified to retrieve the lubrication

302 equation. The 1D simulation method follows the same approach as in the recently

303 presented published work by Tembely *et al*. (2012) namely considering the radial

304 expansions and taking the lower order results in *r* lead to the nonlinear one-

305 dimensional equations describing the filament dynamics [Eggers and Dupont (1994);

306 Shi *et al.* (1994)]. The result is a system of equations for the local radius *h*(z, t) of the

307 fluid neck, and the average velocity *v*(z, t) in the axial direction:

$$\partial_t h + v h' + v' \frac{h}{2} = 0 \tag{14}$$

309 where prime $(')$ denotes the derivative with respect to *z* coordinates and

$$\partial_t v + v v' = -\kappa' + 3 \tilde{v}_s \frac{(v' h^2)'}{h^2} + \frac{1}{h^2} \left[ h^2 (\sigma_{p,zz} - \sigma_{p,rr}) \right]' \tag{15}$$

14

311    For the multimode one-dimensional model in dimensionless form, the axial and radial

312    stress may be expressed as:

313
$$\sigma_{p,zz} = \sum_{i=1}^{N} g_i f(R_i) A_{zz,i}$$
(16)

314
$$\sigma_{p,rr} = \sum_{i=1}^{N} g_i f(R_i) A_{rr,i}$$
(17)

315    <mark>As previously, the full expression of the curvature given in equation (18) was used</mark> to

316    avoid instability in the solution and to provide <mark>the</mark> capacity to represent a rounded

317    drop:

318
$$\kappa = \frac{1}{h(1+h'^2)^{1/2}} - \frac{h''}{(1+h'^2)^{3/2}}$$
(11)

319    To close the one-dimensional model, the following boundary conditions are imposed,

320    the no-slip conditions at the piston surfaces,

321
$$h(z = -L/2, t) = h(z = L/2, t) = R_0$$
(12)

322
$$v(z = -L/2, t) = -V_p, \, v(z = L/2, t) = V_p$$
(13)

323    and a kinematic condition for the radius $h(z,t)$ of the jet may be expressed as

324
$$\frac{dh}{dt} = \frac{\partial h}{\partial t} + v_z \frac{\partial h}{\partial z} = v_r (r = h, t)$$
(14)

325    The governing equations in 1D simulation were solved with COMSOL,

326    <mark>(http://www.uk.comsol.com/) using the Arbitrary Lagrangian-Eulerian (ALE)</mark>

327    <mark>technique. The ALE technique is such that the computational mesh can move</mark>

328    <mark>arbitrarily to optimize the shape of the elements, whilst the mesh on the boundaries</mark>

follows the pistons motion. This ALE capacity implemented in the Comsol code combined with the choice of very fine meshes enables to track the relevant physics as shown in (Tembely *et al.* 2012). Due to the piston motion the computational domain changes with time (see Fig. 3). With the ALE approach, the time derivative of any quantity is defined as $\frac{d}{dt} = \frac{\partial}{\partial t} + (\vec{v} - \vec{v}_m).\nabla$

where $\vec{v}_m$ is the mesh velocity imposed by the piston velocity.

The stress boundaries are ignored in the 1D approach due to the weakly viscoelastic character of the samples and the initial filament aspect ratio being close to 1 [Yao and McKinley, 1998]. The 2D axisymmetric approach includes *per se* that effect.

Fig. 4 presents the evolution of the simulated mid-filament as a function of time for 1D and 2D simulation using different number of mesh elements. The 1D numerical results with between 240 and 3840 mesh elements do not show any difference. The results thus seem to be insensitive to mesh size as shown in the figure below. Similar observation is made for the 2D simulation results regardless of the initial number of mesh elements. The 2D simulation approach mesh is adaptive and evolves with time throughout the simulation resulting a very large number of elements (see insert in Fig. 4.a).

**2D simulation**

An extended version of the split Lagrangian-Eulerian method of Harlen *et al* [Harlen *et al.* (1995)] was used. The nature of the extension was twofold: in the problems for which the method was originally developed there were no free surface boundaries, and the inertial terms were neglected (Re = 0). The method has since been adapted and extended to deal with inertial flows and has been used to model the breakup of

354  Newtonian and viscoelastic jets [Morrison and Harlen (2010); Castrejon-Pita *et al.*

355  (2011)].

356  The velocity and pressure fields are discretized over an irregular triangular mesh of

357  $P_1$--P1 Galerkin elements; each component of the conformation tensor **A** is assigned

358  a value for each element. An artificial stabilization was employed in order to prevent

359  spurious numerical pressure oscillations [Brezzi and Pitkaranta (1984)]. The value of

360  the stabilization parameter was optimized with respect to the spectral properties of the

361  discrete coefficient matrix [Wathen and Silvester (1993)]. A theta-scheme was used

362  for the discrete time-stepping, and the discrete governing equations were linearized

363  via Picard iteration. For each iteration, the linear system was solved numerically using

364  the minimal residual (MINRES) method [Paige and Saunders (1975)]. Adaptive time-

365  stepping was controlled by a CFL [Courant *et al.* (1928)] condition. The position of

366  each mesh node was updated after each time-step using the converged velocity

367  solution.

368  The numerical integration of the evolution equation for the conformation tensor was

369  conducted separately for each element between time-steps, by transforming to a co-

370  deforming frame with local coordinates in each triangle. In such a frame, the upper

371  convected derivative $\overset{\triangledown}{\mathbf{A}}$ becomes the ordinary time derivative dA/dt. Similarly the

372  Lagrangian derivative D**u**/Dt becomes d**u**/dt. The interfacial boundary condition is

373  handled similarly to the treatment by [Westborg and Hassager (1989)].

374  To maintain element shape quality throughout the simulations, local mesh

375  reconnections were made between time-steps in regions where significant element

376  distortion had occurred. The criteria for reconnection were based on the geometric

377  optimality of the Delaunay triangulation [Edelsbrunner (2000)]. The local mesh

17

378      resolution was also maintained by the addition of new nodes in depleted regions, and

379      the removal of nodes in congested regions.

380      In order to represent the capillary breakup of thin fluid filaments, the fluid domain

381      was subdivided artificially when the filament radius falls below a certain threshold.

382      This threshold has been taken as $< 0.5\%$ of the piston diameter to match the smallest

383      diameter that can be experimentally resolved (~6µm). Below this value, the filament

384      is not experimentally visible and is therefore considered broken. A more detailed

385      discussion of the capability of the simulations to capture pinch-off dynamics on a

386      finer scale is given in [Castrejon-Pita *et al.* (2011)].

387

388      **4. Results and discussion**

389      **4.a Experimental results**

390      Examples of the base experimental data are shown in Fig. 5 where photographs of

391      Trimaster experiments for different polymer loading are shown as a function of time.

392      The pure DEP solvent, shown as series 5a, indicates a filament stretch followed by

393      end pinching during relaxation to give a single central drop. The other extreme is

394      shown by series 5d for the 5% polymer loading, where stretching is followed by a

395      progressive filament thinning with a very much longer break up time. The whole time

396      evolution of the full profile along the thread is of general interest and importance;

397      however the detailed behaviour of the centre line diameter will be considered

398      beforehand.

399      **4.b Numerical results**

400      **Mid filament evolution**

401 The experimental time evolution of the mid-point of the filament is given in Fig. 6

402 and the figure displays the characteristic feature of an increased filament life time

403 with a progressive increase of polymer loading. It is this experimental mid filament

404 time evolution that has been used as the basis for comparison with the 1D and 2D

405 numerical simulations. Fig. 7 shows that both the 1D and 2D numerical simulations

406 are in close agreement with the base case Newtonian experimental results. Both the

407 decay profile and final 7.5 ms break point are accurately described by the simulations.

408 Figures 7 to 15 present the evolution of the mid-filament and not the minimum

409 filament or the breakup point which position might vary from one case to another.

410 The simulation breakup diameter has been set at 6µm but might occur at the top and

411 bottom of the filament, as experimentally observed in the case of DEP. In such case,

412 a droplet is formed in the middle of the filament explaining the large diameter

413 observed experimentally and in simulations at breakup time (Fig. 5 and 7).

414 Single mode simulations are shown in Fig. 8, 9 and 10 for 1, 2.5 and 5%

415 concentration solutions respectively. The simulations were carried out using the

416 FENE-CR constitutive equation with the extensibility parameter L = 30. The

417 extensibility value of L = 30 adopted in this paper has been found to provide a better

418 match with the experimental results than the theoretical value of 15. The possible

419 existence of higher molecular mass chains, albeit in small quantities, may justify this

420 choice. Moreover, for an indication of the choice of L, the comparative plot depicted

421 in Fig 13.b of the squared extensibility $L^2$ and $R_i = \mathrm{Tr}\left(\mathbf{A}_i\right)$, which represents the

422 average length per mode i.e. of the polymer chain, shows that an extensibility value of

423 around 30 is an appropriate choice. The 5th mode seems to capture the polymer global

424 chain unravelling mechanism which takes place at larger length scales. On the other

hand, the others modes (1, 2, 3) with negligible values of $R_i$ involves local changes of the molecular conformation; the $R_i$ axial evolution confirms that higher stretching occurs in the middle of the filament.

The capillary thinning of viscoelastic fluid is controlled by the longest relaxation time with a mid-filament diameter decreasing in the form of $D(t) \sim \alpha.\exp(-t/3\lambda)$ [Bazilevsky et al. (1990)). Fitting this exponential decay to the experimental data presented in Fig. 6 yields extensional relaxation times $\lambda_{ext}$ of 0.425ms, 1.19ms and 3.2ms for 1, 2.5 and 5wt% respectively. The extensional relaxation $\lambda_{ext}$ increased with polymer loading as expected. Whilst both the 1D and 2D simulations match the 1% solution data shown in Fig. 8, there is a progressive mismatch in both decay and pinch off with increasing concentration shown in Fig. 9 and 10. In particular the decay immediately after piston cessation is over predicted by both 1D and 2D simulations. Perhaps surprisingly, both the 1D and 2D simulations give a similar response. It was speculated that differences may appear between single mode and multimode models because of the existence of shorter and longer modes and of their interactions close to capillary pinch-off in the vicinity of both pistons [Matallah et al. (2007)].

In the 1D paper, (Tembely et al., JOR 2012) single mode modelling only was used; however both a short mode obtained from the PAV data and a long mode obtained from matching with experiment were used. In that paper it was shown that the smallest relaxation time as input in a non-linear model was unable to correctly predict filament thinning whilst the longest relaxation time gave reasonable filament thinning results but a large discrepancy with the experimental G' and G" data. In this paper, incorporation of multi modes has been carried out in order to fit with greater accuracy the filament thinning experimental results whilst also capturing the PAV data too. We

449     have chosen 5 modes in order to have one mode per decade over the range of interest

450     covered experimentally. The exact choice of the number of modes is a matter of taste.

451     Two would be too few and eight probably too many.

452     In this paper, we have used the same non-linear constitutive equation as in the

453     previous paper and the the oscillatory linear viscoelastic data was then fitted to a

454     multimode model with five modes spaced by a decade between modes and the fitted

455     parameters are given in Table II. These multimode parameters were then used in both

456     the 1D and 2D simulations using the multimode FENE-CR constitutive equation (eq.

457     9 and 10). The results are shown in Fig. 11, 12 and 14 for the 1, 2.5 and 5% solutions

458     respectively. The fit at all concentrations is now greatly improved from the single

459     mode simulations over the whole decay and again there appears to be little difference

460     between the 1D and 2D simulations.

461     Using a multimode Maxwell model approach allows better accounting for the

462     transition between visco-capillary thinning and elasto-capillary thinning as shown by

463     the large reduction of the filament diameter at times between 7 and 10ms. This was

464     one of the main limitations for the single mode Maxwell approach as shown in the

465     previous section and recently reported results by some authors of this paper (Tembely

466     *et al.* (2012)). The results shown I this current paper clearly demonstrate that a

467     multimode description of the fluid is necessary and that, perhaps surprisingly, the 1D

468     simulation appears to give a closer match to the experimental results. The multimode

469     approach also captures the results for potential non-linear elongation behavior and

470     relaxation time changes with the help of using the linear time spectrum and the non

471     linear  constitutive equation.

472     The sensitivity of the filament thinning and breakup to constitutive equation and non

473     linear parameters is shown in Fig. 14 and 15. In Fig. 14 it can be seen that using the

474  1D simulation, there is little difference between the multimode FENE-CR and

475  Oldroyd model predictions. Any differences that may appear were essentially masked

476  by the use of multi modes. Simulation using the theoretically predicted value for the

477  limiting extensibility L of PS110 (L = 15), the "best fit" obtained (L = 30) and a

478  significantly larger value, here L = 100, have been chosen to investigate the effect L

479  of the FENE-CR model. Fig. 14 shows that L does effect the simulation slightly in

480  the transition zone for the short time modes and particularly in the final stages of

481  decay with a pinch off time that decreases with decreasing limiting extensibility

482  parameter L.

483  **Transient profiles**

484  Figure 16 and 17 present the1D and 2D multi modes FENE-CR and Oldroyd-B full

485  simulated transient profiles for the case of 5wt% PS110 diluted in DEP. A generally

486  good match between simulations is observed with differences only appearing towards

487  the end of the filament thinning mechanism, ie, near to break up. Figure 16 shows

488  that the 1D simulation predicts a final thread like decay, whereas the 2D simulation

489  still has a pinch off component. The multi mode Oldroyd-B simulations shown in

490  Figure 17 also shows a similar trend, with the 1D having a more thread like final

491  decay. Despite the improvement provided by the use of multi modes approach instead

492  of the single mode approach, these results clearly highlight the need for investigating

493  other constitutive equations for the modelling of fast stretching and filament thinning

494  of low viscoelastic fluids.

495  Detailed full profile comparison between experimental transient profiles of PS110 at

496  5wt% in DEP with FENE-CR multi modes 1D and 2D simulation transient profiles is

497  presented in Fig. 18. Both simulation approaches provide a good match with the

498  experimental profiles for the overall mechanism with again the main discrepancies

499  appearing at the late stage of the filament thinning mechanism. Close examination of

500  the experimental and simulated profiles show that the fluid regions attached to the top

501  and bottom pistons are smaller experimentally than for both simulations. This results

502  in a larger length of the thinning filament in the experimental case and may explain

503  the differences observed between 1D and 2D simulations. The filament aspect ratio is

504  usually defined by the variation between initial and final position of the piston but it

505  can be seen here that despite using similar piston motions for the simulations and the

506  experiments, differences in the filament length arise. Such filament length variations

507  are expected to significantly affect the filament break up profile especially in the case

508  of low viscosity low viscoelastic fluids. The investigation of the full velocity field, in

509  terms of simulation and using Particle Image Velocimetry (PIV) experiments, within

510  both the filament and the piston region would help the understanding of the

511  differences observed in the filament shape especially toward the break up time.

512

513  **Weissenberg number $W_i$ and apparent extensional viscosity $\eta_{e,app}$**

514  Figure 19 presents the evolution of the Weissenberg number $W_i$ as a function of the

515  filament thinning Hencky strain $\varepsilon$ in the case of multi mode FENE-CR simulations.

516  Weissenberg number and filament thinning Hencky strain may be defined as follows:

517
$$W_i = \lambda_{ext}.\dot{\varepsilon} \tag{22}$$

518
$$\varepsilon = 2.\ln\left(\frac{D_0}{D(t)}\right) \tag{23}$$

519
$$\dot{\varepsilon} = \frac{2}{D(t)}\frac{dD(t)}{dt} \tag{22}$$

520  The simulated data of the mid filament evolution have been used to estimate the

521  longest extensional relaxation time and value of 2.98ms and 5.1ms were obtained for

522    the 1D approach and the 2D simulation respectively, in the case of PS110 at 5wt% in

523    DEP.

524    In the case of the multimode FENE-CR approach, the 1D simulation approach

525    predicts reasonably well the overall mechanism with; in particular the double curved

526    behaviour experimentally observed in the transition between visco-capillary and

527    elasto-capillary regimes ($W_i$ = 0.5) whereas the 2D approach provides a good match

528    on the long time scale but does not capture the double curvature.  The behaviour at

529    high Hencky strain is correctly represented for both types of simulations.

530    The use of the multimode approach does significantly improve the match with

531    experimental data in comparison to that of the single mode and, even if all the

532    subtleties of the complex filament thinning mechanism seem not to be fully

533    represented, it provides good agreement with experimental data. The description of a

534    Weissenberg number, when using a multimode approach, has difficulties in relation to

535    a suitable choice of relaxation time used in the definition of the Weissenberg number.

536    It is also very sensitive to noise (simulation or experimental) due to the fact that it is

537    based on the derivative of the mid filament evolution.

538    Finally, Fig. 20 presents the transient apparent extensional viscosity $\eta_{e,app}$, with

539    $\eta_{e,app} = -\sigma.\dfrac{dt}{dD_{mid(t)}}$, as a function of Hencky strain for multimode FENE-CR. The

540    comparison is particularly good in view of the approximations which have been made

541    for the calculation of the phenomenological Maxwell times. Notably, the complex

542    behaviour of the extensional viscosity is qualitatively correctly predicted at

543    intermediate times by both the 1D and 2D simulations with the prediction of the

544    sudden increase in $\eta_{ext}$ after the pistons have stopped.  Close attention shows  that the

545    1D simulation approach produces a surprisingly good agreement with experimental

546 results, while the 2D simulation approach fails to represent the long term extensional

547 viscosity behaviour.

548

549 **5. Conclusions**

550 Results described in this paper have shown that a multimode constitutive equation

551 approach is necessary to describe the detailed viscoelastic extensional flow behaviour

552 of dilute or semi dilute polymer solutions. The result is consistent with the findings of

553 Entov and Hinch (1997) who also found it necessary to resort to a multimode mode

554 approach for higher viscosity viscoelastic polymer solutions. However, simulations

555 for different polymer concentrations indicate that the improvement due to the use of

556 multimodes instead of single mode is reduced with increase of the solution diluteness.

557 Results presented in this work indicate great potential for the simulation of very fast

558 break up dynamic of more dilute polymer solution using multimode Maxwell

559 approach with important application potential in areas such as ink jet printing.

560 The FENE-CR constitutive equation appears to be an effective suitable constitutive

561 equation to use for the fluids examined in this paper, although the Oldroyd model was

562 found to give an equivalent response when used with multimodes. It appears that

563 multimode modelling can disguise certain limiting features of different constitutive

564 models, but however remains necessary even for the monodisperse polymer systems

565 which have been tested.

566 An initially surprising result of the paper is the fact that the 1D modelling gives

567 apparently improved results over the more rigorous 2D modelling in some limited

568 cases described above. This indicates that the 1D approximation is valid enough for

the initial and boundary conditions used and in particular for the mid filament

diameter evolution. It is probable that when details of highly non-linear behaviour, i.e.

pinch off position, number of beads, etc. are considered differences will emerge from

the two techniques. The pinch off position and the number of small drops is an

essential parameter in ink-jet printing since the satellite drops may merge or not

following the type of detachment.

Further comparison would be to follow the filament transients following breakup.

Such a work has been done for Newtonian liquid (Castrejon Pita et al. 2012) but this

work does not include non-Newtonian fluids., The non-linear evolution of main drop

and satellites do influence printability criterion taking into account the Ohnesorge and

the Deborah numbers as described in preliminary work by Tembely et al. 2011.

589

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695    models," Chem. Eng. Sci. 56(1), 211–233 (2001).

696

697

| Solvent | $M_w$ (g/mol) | C (wt%) | $\eta^*$ (mPa.s) |
|---|---|---|---|
| DEP | 110000 | 0 | 10 |
| DEP | 110000 | 1 | 15.2 |
| DEP | 110000 | 2.5 | 31.5 |
| DEP | 110000 | 5 | 69 |

Table I: Zero shear rate complex viscosity of the different polymer solutions at 25°C

| $l_i(\mu s)$ | 1%PS | 2.5%PS | 5%PS | 10%PS |
|---|---|---|---|---|
| | $g_i$(Pa) | $g_i$(Pa) | $g_i$(Pa) | $g_i$(Pa) |
| 1 | 7.789 | 83.8229 | 397.9015 | 1086.4419 |
| 10 | 428.76 | 1450.8952 | 4680.9517 | 9126.8723 |
| 100 | 1.6435 | 10.5177 | 93.1172 | 2012.6511 |
| 1000 | 0 | 0 | 0 | 16.4133 |
| 10000 | 0.0342 | 0.1855 | 0.4288 | 0.4291 |

Table II: Relaxation time and shear modulus obtained from Maxwell model fit of the

PAV data for the different samples

705

706

a



b



c

707
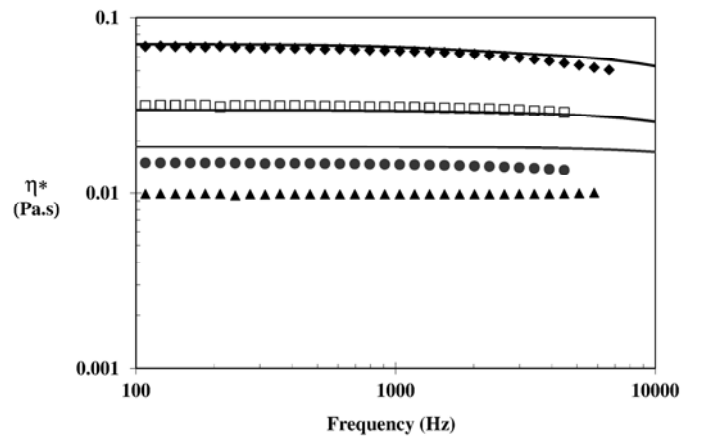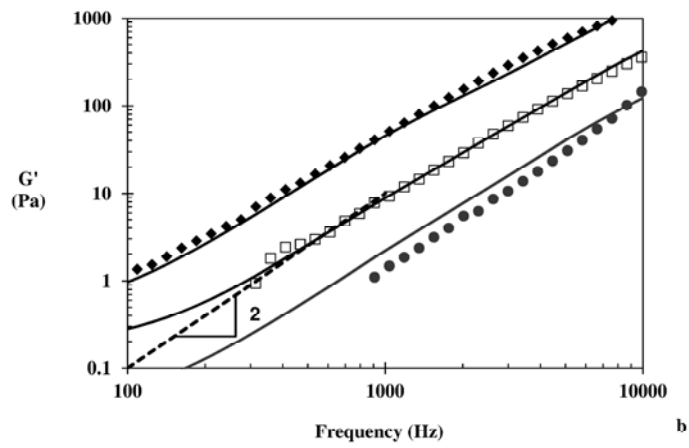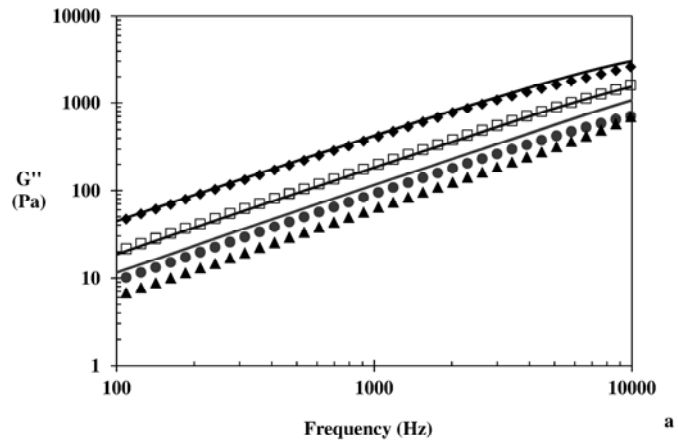
708 **Figure 1:** Evolution of (a) Loss modulus G'', (b) elastic modulus G' and (c) complex

709 viscosity h* as a function frequency for DEP-PS 110 000 solutions at different

710 concentrations. (▲) DEP, (●)DEP-1wt% PS110, (□) DEP-2.5wt% PS110, and (♦)

711 DEP-5wt% PS110. Solid line represents the multimode optimization results while the

712 dashed line on G' graph corresponds to a power law function of index 2.

713

**Free surface**

$D_{mid}(t)$

$L(t) = L_0 + 2V_p t$

$L_0$

$D_0$

$t_0$

**Symmetry axis**   $t > t_0$

$-V_p$

$V_p$
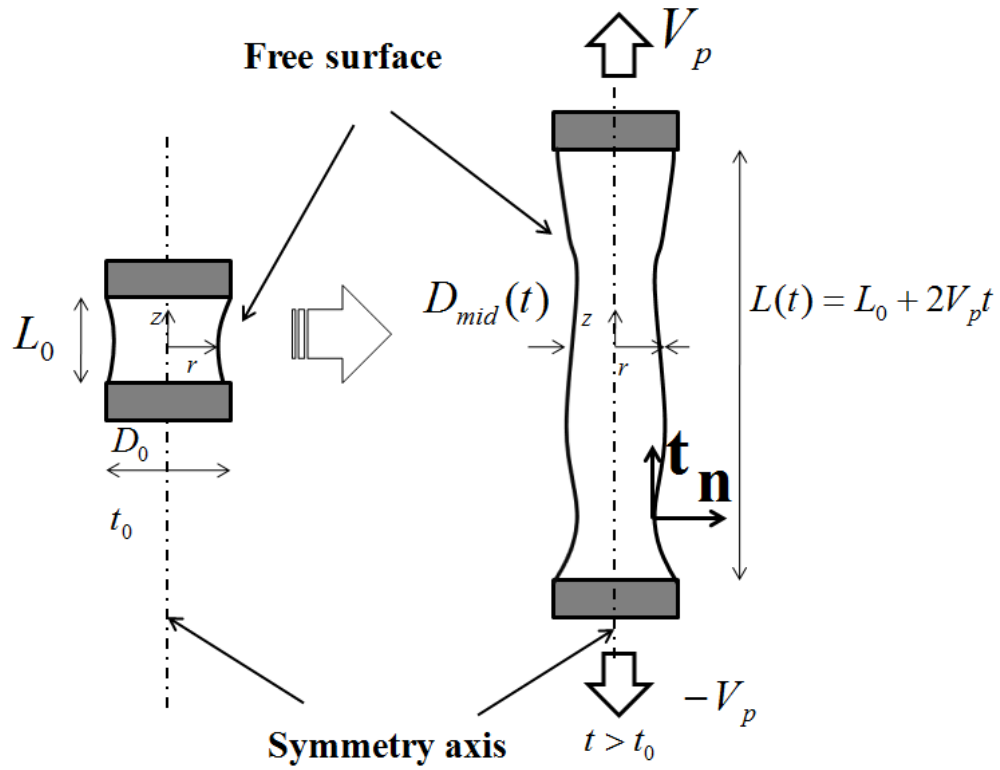
**Figure 2**: Diagram of filament stretch and thinning geometry  and the computational domain, shown midway through the stretching phase as the pistons move outwards and the fluid column necks in the middle.  Initially the fluid column is cylindrical. Extracted from [Tembely *et al.*, 2012]

37
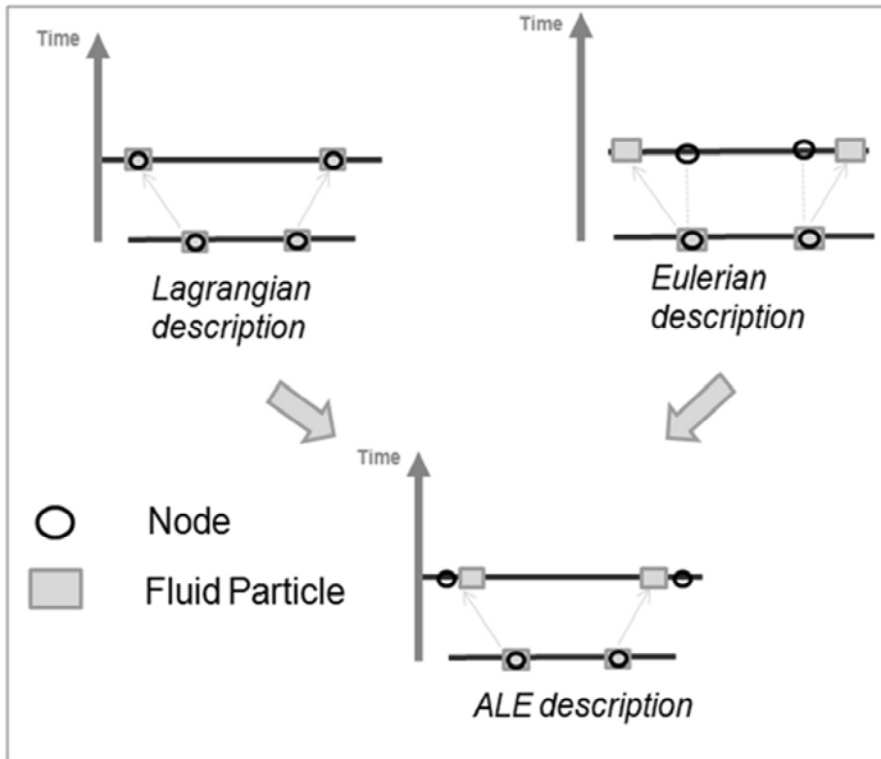
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724    **Figure 3:**  mesh evolution of the ALE method for the 1D simulation

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726
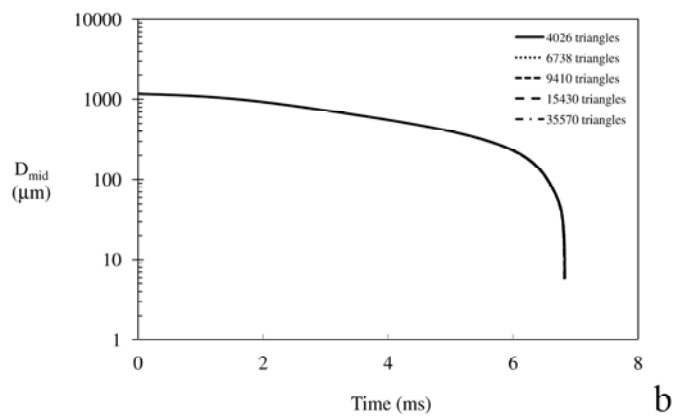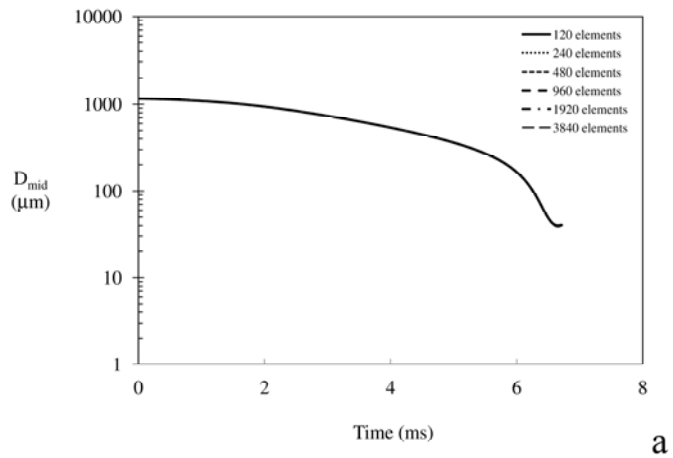
727
728
729 **Figure 4**: Evolution of the simulated mid-filament for different number of mesh
730 elements for (a) 1D simulation approach (b) 2D simulation approach. In the 2D
731 simulation, the legend gives the number of triangles at t = 7.2ms.
732

733



734
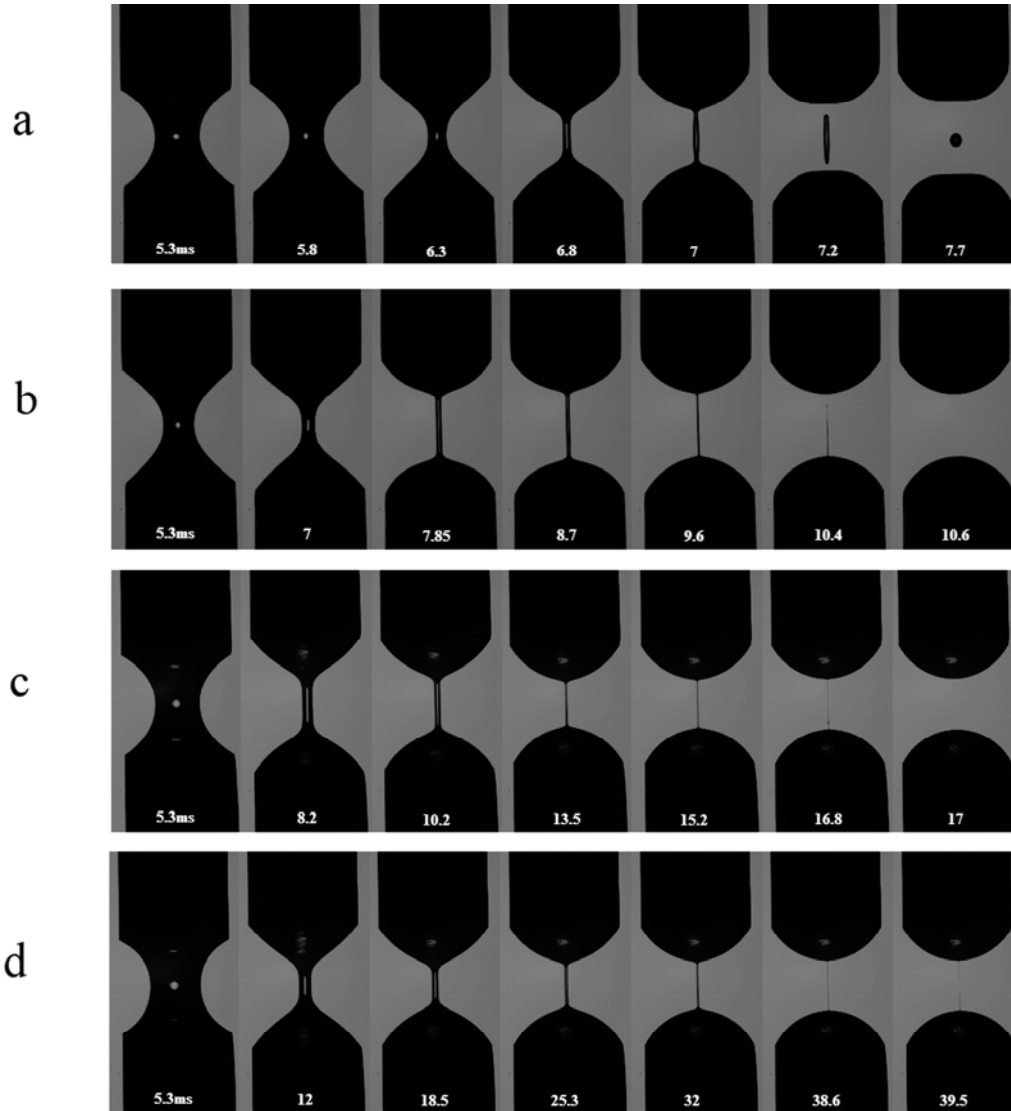
**Figure 5**: Photograph of the filament stretch, thinning and break up captured with the Trimaster for (a) DEP, (b) DEP + 1wt% PS110, (c) DEP + 2.5wt% PS110, (d) DEP + 5wt% PS110. The first picture of each series (t = 5.3ms) corresponds to the piston cessation of motion

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**Figure 7:** Newtonian base case. Plot of the mid filament diameter evolution as a function of time. Vertical line (---) corresponds to piston cessation of motion.

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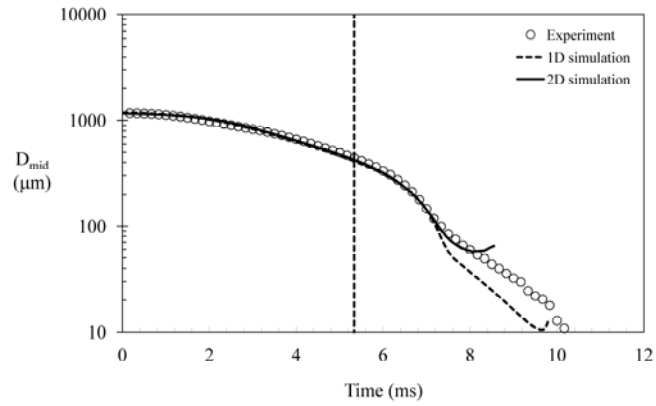


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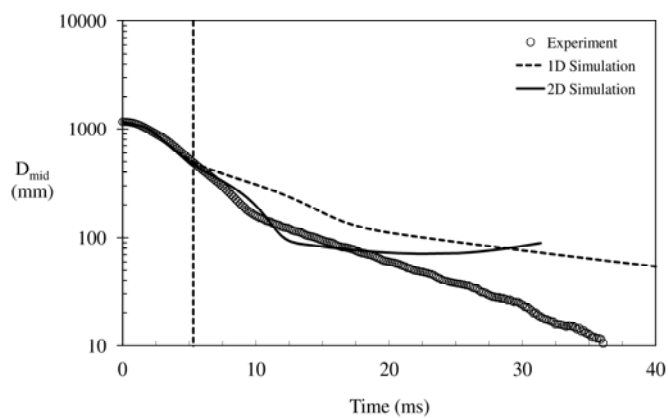754

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**Figure 10:** Single mode, 5wt% PS110 in DEP solution. Plot of the mid filament diameter evolution as a function of time. Constitutive equation: Fene-CR, relaxation time $\lambda$ = 3.2 ms, shear modulus g = 17Pa and polymer extensibility L = 30.   Vertical Line (---) corresponds to piston cessation of motion.

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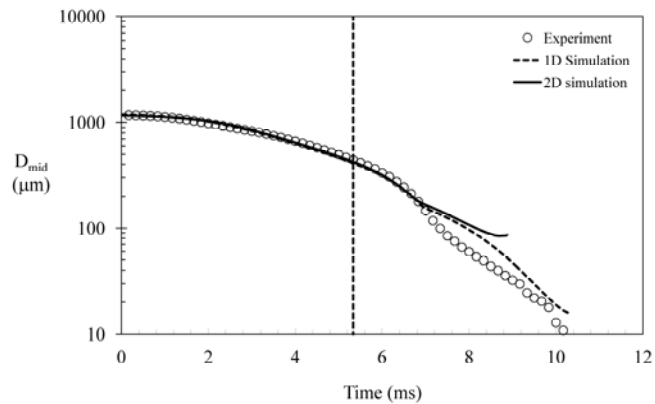


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**Figure 12:** Multi mode, 2.5wt% PS110 in DEP solution. Plot of the mid filament

785 diameter evolution as a function of time. Constitutive equation: Fene-CR, relaxation

786 times $\lambda_i$ and shear modulus $g_i$ for the different modes i are given in Table II and

787 polymer extensibility L = 30. Vertical line (---) corresponds to piston cessation of

788 motion.

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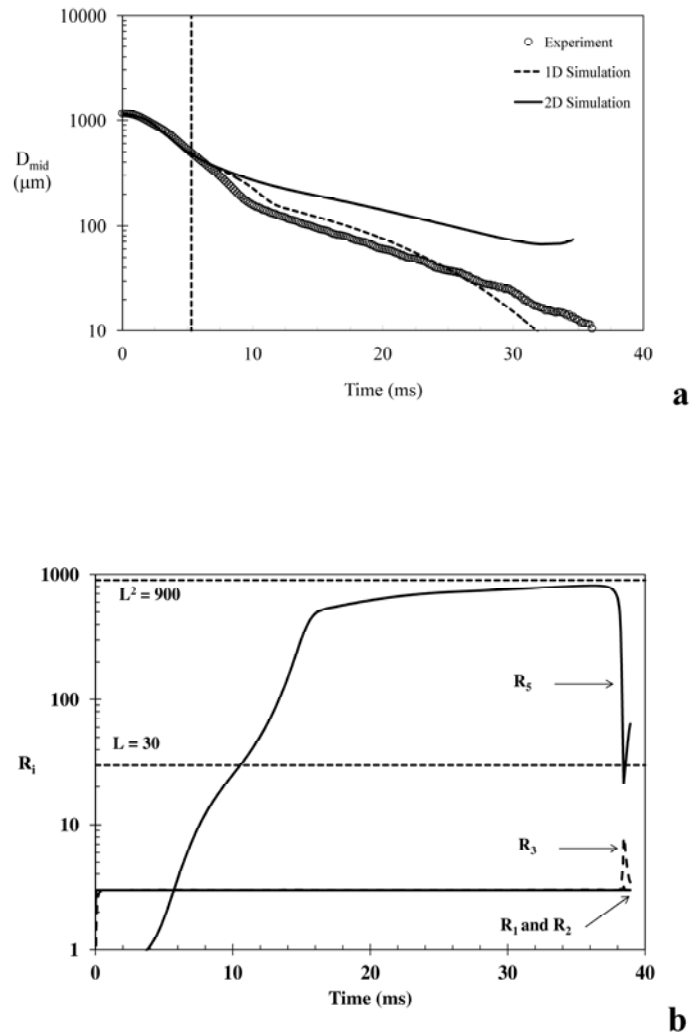
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793   **Figure 13:** (a) Multi mode, 5% solution. Plot of the mid filament diameter evolution

794   as a function of time. Constitutive equation: Fene-CR, relaxation times $\lambda_i$ and shear

795   modulus $g_i$ for the different modes i are given in Table II and polymer extensibility L

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**Figure 14:** Multi modes, 5wt% PS110 in DEP solution. Plot of the mid filament diameter evolution as a function of time. Constitutive equation: Oldroyd-B, relaxation times $\lambda_i$ and shear modulus $g_i$ for the different modes i are given in Table II. Vertical line (---) corresponds to piston cessation of motion.

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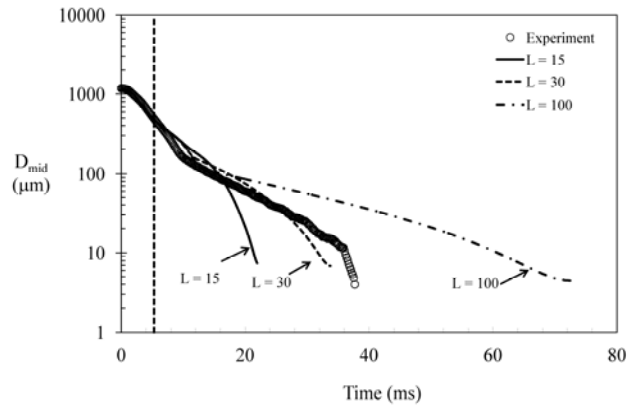
**Figure 15:** Effect of extensibility parameter L. Symbols represent the experimental

data of the evolution of the mid-filament as a function time and lines represent 1D

multi-mode numerical simulations for different polymer chain extensibilities L.

Constitutive equation: Fene-CR , relaxation times $\lambda_i$ and shear mdulus $g_i$ for the

different modes i are given in Table II.. Vertical line (---) corresponds to piston

cessation of motion.

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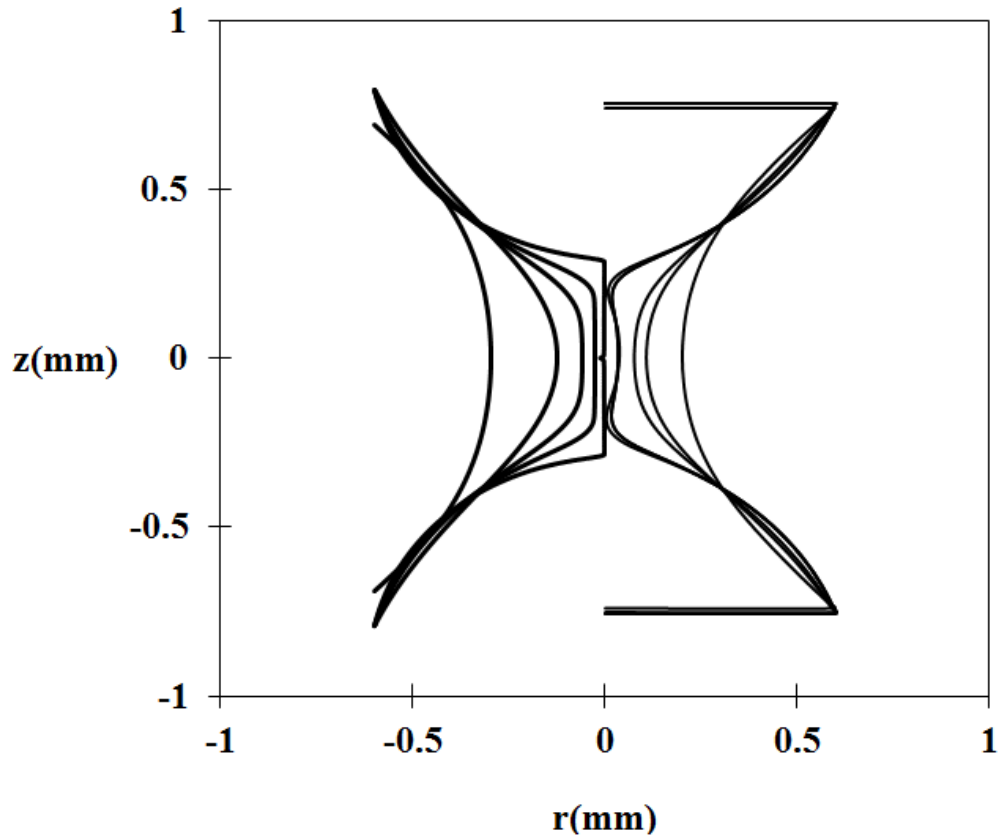
**Figure 16:** Comparison between the 1D numerical FENE-CR multimode transient

profiles (left), and the corresponding 2D simulations (right) for the DEP+5%PS. The

prescribed times are 5.3ms, 12ms, 18.5ms, 25.5 ms, 38ms.
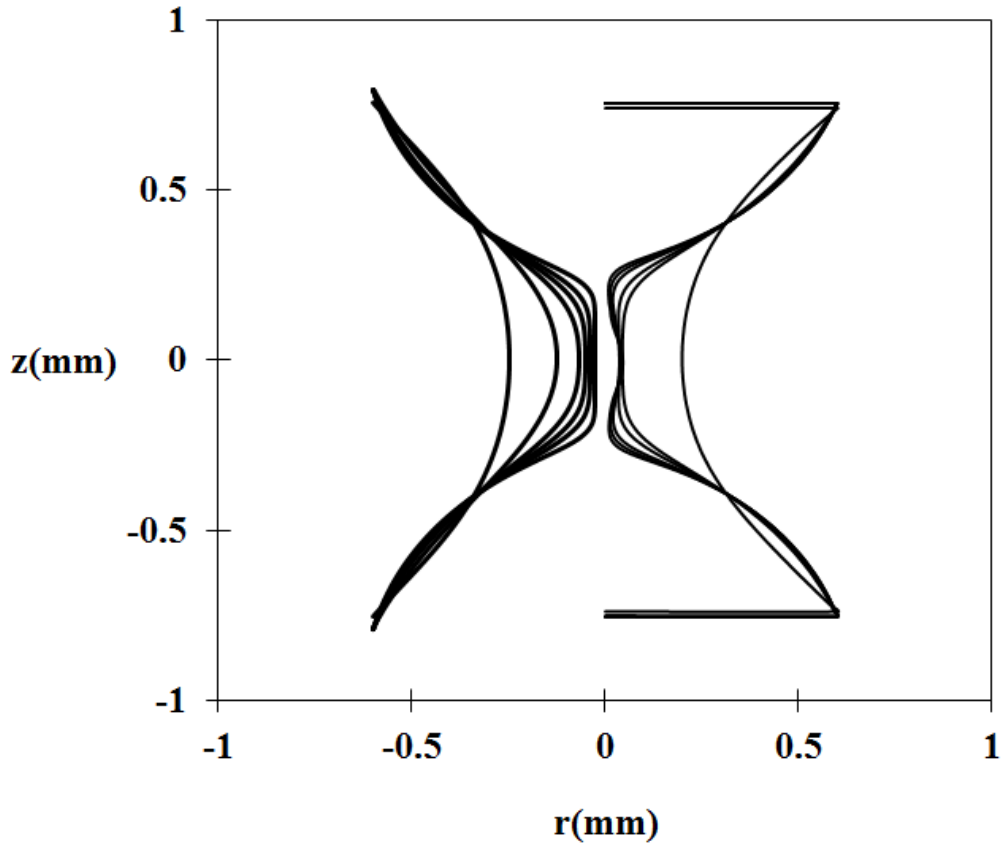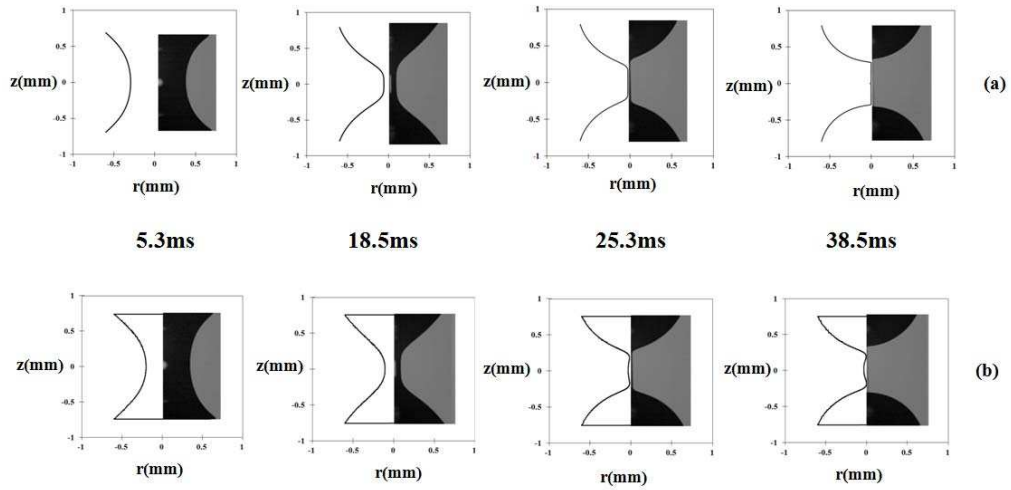
**Figure 17:** A comparison between the 1D numerical Oldroyd-B multimode transient

profiles (left), and the corresponding 2D simulations (right). The prescribed times are

5.3ms, 12ms, 18ms, 25ms, 32ms and 44ms for 1D simulation and 5.3ms, 12ms, 18ms,
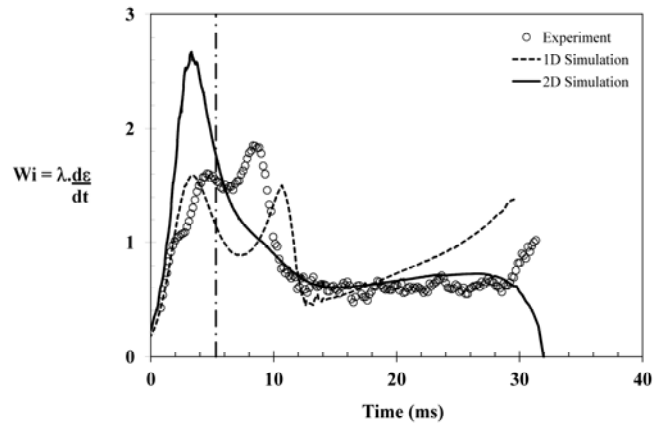
25ms, 28ms, 32.5ms for 2D simulation

**Figure 18:** Comparison between the experimental transient profiles for the DEP+5wt%PS110 and the simulations of (a) the 1D and (b) the 2D cases using the FENE-CR multimode constitutive equations.

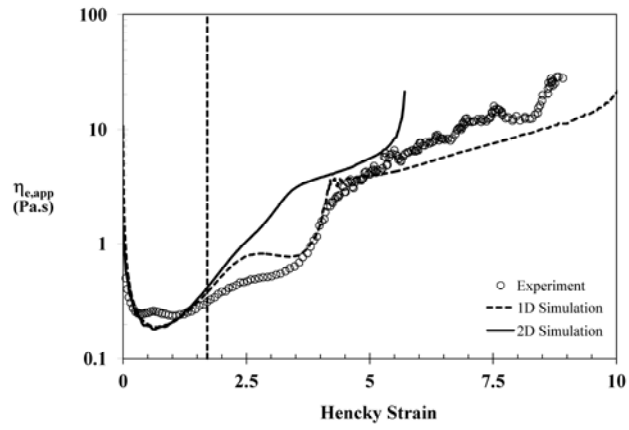834

**Figure 19**: Evolution of the Weissenberg number as a function of the Hencky strain.
Transient Weissenberg numbers were calculated using $\lambda = 3.2$ms for experimental
data, $\lambda = 2.89$ms and $\lambda = 5.1$ms for 1D simulation and 2D simulation data using multi
modes FENE-CR as constitutive equation.

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**Figure 20**: Evolution of the transient apparent extensional viscosity $\eta_{e,app}$ as a function of the Hencky strain $\varepsilon$ for computed from the mid filament evolution shown in Fig. 12.

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