Numerical simulation of contraction and expansion flows of Langmuir monolayers

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1. Introduction

In recent years there is a growing trend in the field of non-Newtonian fluid mechanics towards validating experimental results in non-trivial flows. To that effect, a series of papers have appeared (see, for example, Refs. [1–4]). Olson and Fuller [5] performed experiments for two model fluids, which are typical examples of two-dimensional Langmuir monolayers. These are arachidyl alcohol (Newtonian) and PODMA [poly(octadecyl methacrylate)] (viscoelastic) in a 4:1 planar contraction and a 4:1 planar expansion (see Fig. 1). They also provided limited rheological data (loss modulus, $G''$), which were fitted linearly. They calculated experimentally (in an elementary level) the basic material functions, such as the zero-shear viscosity. One of the objectives was to entice the computational rheological community to undertake simulations for direct comparisons with the experiments.

On the other hand, much progress has been made in the viscoelastic simulation of polymeric liquids with integral constitutive equations of the K-BKZ type. A particular variant of this equation, the PSM model [6], has been extensively used in viscoelastic simulations, especially in axisymmetric contraction flows [7–9]. A particular defect of this model for planar flows of branched polymers has been corrected by Olley [10] and used in simulations in comparison with experiments by Mitsoulis et al. [11].

It is the purpose of the present paper to undertake simulations for the above-mentioned Langmuir monolayers with the integral K-BKZ model as modified by Olley [10], and to provide detailed numerical results corresponding to the experimental conditions. These results can be used for further comparison with the work from other researchers and with other constitutive equations.

2. Mathematical modeling

2.1. Governing equations

The isothermal contraction or expansion flows shown in Fig. 2 are being modeled by governing equations that are deduced from the equations of continuity and motion. For an incompressible fluid, the equation of continuity gives

$$\nabla \cdot \bar{u} = 0,$$

and the equation of motion with inertia terms for a steady motion gives

$$\rho \ddot{\bar{u}} + \nabla p = \nabla \cdot \bar{\tau},$$

where $\rho$ is the fluid density, $\bar{u}$ is the particle velocity, $p$ is the pressure, and $\bar{\tau}$ is the stress tensor.
where $\vec{u}$ is the velocity vector, $p$ is the pressure, $\overline{T}$ is the extra stress tensor, and $\rho$ is the fluid density.

### 2.2. Constitutive equation

The constitutive equation that relates the stresses $\overline{T}$ to the deformation history is a variant of the K-BZK equation proposed by Papanastasiou et al. [6], which is written as

$$
\overline{T} = \frac{1}{1-\theta} \int_{-\infty}^{t} \sum_{k=1}^{N} \frac{\alpha_k}{\lambda_k} \exp \left(-\frac{t-t'}{\lambda_k} \right) h(\overline{l}_{c-1}) \times \left( \frac{1}{C_1} \left( \frac{1}{C_1(t')-1} \right) + \theta \left( \frac{1}{C_1(t')-1} \right) \right) dt',
$$

where $h(\overline{l}_{c-1})$ is a strain-memory function proposed by Olley [10], so that the K-BZK model can predict an equal amount of strain-thickening behavior in uniaxial and in planar extension. In the above, $\lambda_k$ and $\alpha_k$ are the relaxation times and relaxation modulus coefficients, respectively, $\alpha_k$, $\beta_k$ and $\theta$ are constants, and $\overline{l}_{c-1}$ is the first invariant of the Finger strain tensor, $\overline{C}_1^{-1}$, being the inverse of the Cauchy–Green tensor, $\overline{C}_1$. The constant $\theta$ is given by

$$
\frac{N_2}{N_1} = \frac{\theta}{1-\theta},
$$

where $N_1$ and $N_2$ are the first and second normal stress differences, respectively. The value of $\theta$ is non-zero for polymeric liquids exhibiting a second normal stress difference. Its usual range is between 0 and −0.3 [9]. However, due to lack of measurements, for the present work we have set $\theta = 0$. In 2D planar flows this assumption does not alter the results [11].

Note that multiple $\lambda_k$ and $a_k$ may be used, corresponding to different strain-memory functions for each relaxation mode [11]. Thus, the separability of the time-memory and the strain-memory functions that has been questioned in the past [12], does not have to be assumed.

The effect of $N_2$ in various flow simulations has been examined by several workers [13–15], while the effect of multiple strain-memory functions has been also examined by Luo and Tanner [14] and by Mitsoulis [15].

In the case of entry flow in a contraction, it has been customary to define a dimensionless Weissenberg number by

$$
Ws = \frac{\lambda(\dot{\gamma}_0)}{H_d},
$$

where $\lambda$ is a relaxation time given by

$$
\lambda = \frac{N_2}{2\tau V_0},
$$

where $\tau$ is the wall shear stress for fully developed flow in the small channel (die), and $\dot{\gamma}_0$ is the apparent shear rate, related to the average velocity $V_{avg}$ by [5]:

$$
\dot{\gamma}_0 = \frac{V_{avg}}{H_d},
$$

where $H_d$ is the downstream channel (die) half width (see Fig. 2), and it can be used as a convenient representation of flow rate in the absence of a single relaxation time. In the case of a relaxation spectrum (as it is used here), a zero-shear rate Weissenberg number can also be calculated by using the “Maxwell” relaxation time, $\lambda_0$, evaluated at the limit of zero-shear rate, as done in the work by Olson and Fuller [5], i.e.

$$
Ws_0 = \lambda_0 \dot{\gamma}_0.
$$

### 3. Rheological characterization

The test fluids used by Olson and Fuller [5] are arachidyl alcohol and PODMA, which is a 23.4 wt.% solution in toluene. Measurements and calculations revealed that arachidyl alcohol is a Newtonian fluid, which has a surface viscosity $\mu = 0.028$ mN s/m at 22 °C, while PODMA is a viscoelastic fluid, which has a surface zero-shear rate viscosity $\eta_0 = 0.034$ mN s/m and a “Maxwell” relaxation time $\lambda_0 = 2.9$ s at 25 °C. Our first concern in our efforts to model the viscoelastic fluid (PODMA) was to obtain as much information as we could from the experimental data. The exponent $n$ of the power-law model was calculated by Olson and Fuller [5] ($n = 0.53$) but the consistency index $m$ was unknown.

In order to obtain the value of $m$ we consider flow between parallel plates (with the known gap $2H_d = 5$ mm of the narrow channel in Fig. 2). We also know the average velocity, $V_{avg}$, of the fully developed flow in the narrow channel from $\dot{\gamma}_0$ and $H_d$ (see Fig. 2), according to

$$
V_{avg} = \dot{\gamma}_0 H_d.
$$

The maximum velocity $V_{max}$ can now be calculated easily from

$$
V_{max} = \frac{2n + 1}{n + 1} V_{avg}.
$$
Table 1
Relaxation spectrum and associated parameters for a PODMA solution at 25 °C found from non-linear regression for the K-BKZ/Olley model (a = 1.513, \( \theta = 0 \)).

<table>
<thead>
<tr>
<th>( k )</th>
<th>( \lambda_k ) (s)</th>
<th>( a_k ) (mN/m)</th>
<th>( \bar{\eta}_k )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>( 5.06 \times 10^{-7} )</td>
<td>( 1.36 \times 10^{-4} )</td>
<td>( 0.48 \times 10^{-1} )</td>
</tr>
<tr>
<td>2</td>
<td>( 1.40 \times 10^{-5} )</td>
<td>( 1.12 \times 10^{-2} )</td>
<td>( 0.22 \times 10^{-1} )</td>
</tr>
<tr>
<td>3</td>
<td>( 2.51 \times 10^{-3} )</td>
<td>( 5.80 \times 10^{-2} )</td>
<td>( 0.31 \times 10^{-2} )</td>
</tr>
<tr>
<td>4</td>
<td>( 2.46 \times 10^{-4} )</td>
<td>( 3.39 \times 10^{-4} )</td>
<td>( 0.18 \times 10^{-4} )</td>
</tr>
<tr>
<td>5</td>
<td>( 4.72 \times 10^{-5} )</td>
<td>( 2.09 \times 10^{-5} )</td>
<td>( 0.11 \times 10^{-5} )</td>
</tr>
<tr>
<td>6</td>
<td>( 1.12 \times 10^{-3} )</td>
<td>( 8.03 \times 10^{-2} )</td>
<td>( 0.07 \times 10^{-4} )</td>
</tr>
<tr>
<td>7</td>
<td>( 1.13 \times 10^{-4} )</td>
<td>( 8.31 \times 10^{-5} )</td>
<td>( 0.75 \times 10^{-4} )</td>
</tr>
<tr>
<td>8</td>
<td>( 2.30 \times 10^{-5} )</td>
<td>( 2.04 \times 10^{-5} )</td>
<td>( 0.35 \times 10^{-5} )</td>
</tr>
</tbody>
</table>

The \( V_{\text{max}} \) is related to the parameter \( m \) according to

\[
V_{\text{max}} = \frac{n}{n+1} \left[ \frac{H_{d}^{H+1}}{m} \left( \frac{\Delta P}{T} \right) \right]^{1/n},
\]

where \( \Delta P \) is the surface pressure difference that drives the flow for PODMA (\( \Delta P = 5 \) mN/m) and \( L = 6.5 \) mm. From the above equations, we find \( m = 0.080 \) mN s/m.

The next step was to model PODMA with the K-BKZ integral Eq. (3). We needed hypothetical experimental values \( \eta \) vs. \( \dot{\gamma} \), and for that purpose we calculated the viscosity \( \eta \) from hypothetical \( \dot{\gamma} \) in the region where the power-law model is valid.

Next we performed a non-linear optimization fitting of those hypothetical \( \dot{\gamma} \) and for that purpose we calculated the viscosity \( \eta \) from hypothetical experimental values \( \dot{\gamma} \) vs. \( \eta \). From the relaxation spectrum it follows that the zero-shear viscosity, \( \eta_0 = 0.044 \) mN s/m, the infinite-shear rate viscosity, \( \eta_\infty = 0.007 \) mN s/m, and the Cross time constant, \( \lambda_c = 0.058 \) s.

By modeling those hypothetical experimental values \( \eta \) vs. \( \dot{\gamma} \) for the PODMA with the K-BKZ integral Eq. (3), we determined the parameters of this model. These are reproduced in Table 1 and have been determined with a spectrum of 8 relaxation times.

From the relaxation spectrum it follows that the zero-shear viscosity \( \eta_0 = \sum a_k \lambda_k \) of the polymer contribution is 0.047 mN s/m, and the average relaxation time \( \lambda = \sum a_k \lambda_k^2 / \sum a_k \lambda_k \) is 0.48 s. Note that Olson and Fuller [5] have used another relaxation time \( \lambda_0 = 2.89 \) s, by dividing the surface zero-shear viscosity by the surface modulus, in an effort to have some relaxation time for their calculation of the \( \psi \) number. Obviously, the two \( \lambda \)'s are not related.

With the parameters of Table 1, it is possible to fit the experimental data for different shear rates and obtain the shear-thinning behavior for the shear viscosity, as well as the quadratic behavior at very low shear rates for the first normal stress difference, as shown in Fig. 3b. Regarding the extensional-thickening behavior for the extensional viscosities and due to lack of experimental data, this was found by making use of the flow patterns evident in the experiments. Namely, the amount of strain-thickening was adjusted (by a judicious choice of the beta-parameters) so that the experimental vortex formation was reproduced (see below). It should be noted that due to the planar type of flow only the planar viscosity, \( \eta_p \), is relevant here. However, we also present the other two extensional viscosities, the elongational viscosity, \( \eta_e \), and the biaxial viscosity, \( \eta_b \), as it is customary, in order to show the response of the model in all three types of extension.

The predictions of the model for the dynamic functions \( G' \) (storage modulus) and \( G'' \) (loss modulus) are presented in Fig. 3a. We must point out here that lack of sufficient data for the storage modulus \( G' \) gives poor predictions for this function.

Another important point concerns the density of the monolayers, which is necessary for calculating the inertia terms in the equation of momentum (Eq. (2)). Olson and Fuller [5] used arachidyl alcohol at \( \Delta P = 30 \) mN/m, which corresponds to an average area per surfactant molecule of 25 Å² (cf. their Fig. 4). Since arachidyl alcohol has a molecular weight MW = 298 g/mol, this corresponds to a surface density of \( \rho = 1.98 \times 10^{-6} \) kg/m². The same thing can be done for the PODMA, with a MW = 950 × 500 = 322,500 g/mol.

Calculating the Reynolds number, \( Re \),

\[
Re = \frac{\rho \psi \bar{H}_d}{\mu},
\]

of the surface is not very fruitful since nearly all flows have negligible surface inertia [5]. For example, for the arachidyl alcohol at this surface density, using a surface viscosity \( \mu = 2.8 \times 10^{-5} \) N s/m, the range of surface Reynolds numbers, corresponding to experimental shear rates, is

\[
Re_\psi = \frac{\bar{\rho} \psi \bar{H}_d^2}{\mu} = (1.98 \times 10^{-6} \text{ kg/m}^2) \times (43.1 – 107.81 \text{ s}) \\
\times (2.5 \times 10^{-3} \text{ m}^2) / (2.8 \times 10^{-5} \text{ N s/m}) \\
= 1.9 \times 10^{-5} \text{ to } 4.8 \times 10^{-5}.
\]
Inertia can only arise from the subphase, and a Reynolds number based on the subphase properties makes the most sense. To correctly account for the inertia from the subphase, the fully coupled monolayer-subphase flow problem must be solved, which is a much harder 3D problem. It is known that the subphase can affect rheological measurements, and accounting for it has been quite recently investigated [16]. Although this approach is best, we choose here, as a first approximation, to find by an iterative process the value of surface density which gives the correct streamline patterns. Thus, a value of $\rho_{\text{eff}} = 0.0029 \text{ kg/m}^2$ was found. This gives rise to Reynolds numbers ranging from $0 < Re < 11.2$ in the range of simulations for the Newtonian monolayer. This density can then be seen as an effective surface density, $\rho_{\text{eff}}$, which takes into account the interaction between monolayer and subphase.

4. Method of solution

The special numerical scheme developed by Luo and Mitsoulis [17] for the implementation of integral-type constitutive equations with the finite element method (FEM) has been used. This scheme is effectively an EVSS-G/SI scheme in the jargon of viscoelastic simulations. The simulations have also been repeated with the Adaptive Viscoelastic Stress Splitting with Streamline Integration (AVSS/SI) introduced by Sun et al. [18], but the results were identical. Apparently, the latter method is particularly well suited for flows with steep stress boundary layers, such as the ones encountered in flows around a sphere.

The numerical algorithm for convergence is Picard iteration, sometimes used with an under-relaxation factor of 0.5 for the stresses in order to avoid divergence for the higher flow rates. Convergent solutions have been obtained for the whole range of experimental flow rates for the Newtonian monolayer, but for the PODMA viscoelastic case, convergence was more difficult to obtain at higher flow rates.

The solution procedure advances slowly from low flow rates (Newtonian behavior) to higher ones by using a flow rate increment scheme. On average 35 CPU s per iteration were needed with a mesh having 1740 elements on an Intel Core2-Duo at 2.66 GHz PC with 2GB of RAM, for a total of 1000 iterations up to $\dot{\gamma}_0 = 16 \text{ s}^{-1}$. The criterion for convergence was $10^{-3}$ for the maximum changes in the velocities and $10^{-2}$ for the pressure. The behavior noticed by Barakos and Mitsoulis [19] was again apparent for the maximum changes of the primary variables, i.e. all relative changes go to machine accuracy ($\sim 10^{-15}$ to $10^{-16}$) except for the variables at the entrance singularity, which fluctuate around a mean value ($\sim 10^{-2}$ to $10^{-3}$), which in turn depends on the grid density around the singularity. It should be noted that in all our works Galerkin averaging has been used for the rates-of-strain, and on top of this, arithmetic averaging has been used for these variables around the singularity to help reduce the very steep velocity gradients [19].

With regard to Fig. 2, let $L_u$, $L_d$ be the lengths upstream and downstream of the contraction, respectively, and let $H = H_d$ be the half the small channel gap and $H_{\text{res}} = 4H$ the reservoir gap. Due to different requirements for flow development in contractions and expansions, different lengths are required. For our initial calculations, we had chosen for the contraction the ratios $L_u/H$ and $L_d/H$ to be 16 and 30, as done previously [11]. For the expansion we chose $L_u/H$ and $L_d/H$ to be 4 and 16 [20]. Caution must be exercised in choosing the downstream length, so that there is an adequate length to accommodate fully developed profiles at the exit even for the highest flow rates. However, in the course of the present work,
the open boundary condition (OBC) at the outflow of Papanastasiou et al. [21] proved a valuable addition, as it allowed curtailing the domain without loss of quality of the solution. For the boundary conditions we assume there is no slip at the die walls (zero velocities imposed) and set a fully developed velocity profile at the entry while at the exit the OBC is applied (see also Fig. 2). All lengths are scaled with \( H \), all velocities with \( V_{\text{avg}} \) (the mean downstream velocity) and all pressures and stresses with \( \eta_0 V_{\text{avg}}/H \).

The finite element meshes used in the simulations are shown in Fig. 4 while details regarding domain lengths, number of elements, number of nodes and number of unknown degrees of freedom (DOF) are given in Table 2. Fig. 4a shows the preliminary mesh M1 for the contraction, while Fig. 4b shows the preliminary mesh M2 for the expansion. These two meshes were initially used for a series of fast runs to study the phenomena and find out the influence of parameters, especially for the viscoelastic simulations. Then, the denser meshes M3 and M4 were used, shown in the upper half of Fig. 4c and d for the contraction and the expansion, respectively. The lower halves of these figures show meshes 4 times denser, which are constructed by further subdivision of each parent element into \( 2 \times 2 \). These much denser meshes are used for sample runs to check that results are indeed mesh-independent.

### 5. Results and discussion

#### 5.1. Newtonian monolayer

The simulations have been undertaken for the whole range of the experimental flow rate values [5] including inertia terms with the effective surface density, \( \rho_{\text{eff}} \). The quantities of interest are:

(i) the dimensionless vortex length \( X \) defined by

\[
X = \frac{L_v}{H_{\text{res}}},
\]

where \( L_v \) is the vortex length (see Fig. 2) and \( H_{\text{res}} = 4H \) is the reservoir half gap;

(ii) the dimensionless vortex intensity \( -\psi_r_{\text{max}} \) defined by

\[
-\psi_r_{\text{max}} = \frac{\psi_{r,\text{max}} - \psi_w}{\psi_{cl} - \psi_w},
\]

where \( \psi_{r,\text{max}} \) is the stream function value in the eye of the vortex, \( \psi_w \) is the stream function value at the wall and \( \psi_{cl} \) is the stream function value at the centreline; and

(iii) the dimensionless entrance correction \( n_{\text{en}} \) defined by

\[
n_{\text{en}} = \frac{\Delta P - (\Delta P_{\text{res}} + \Delta P_0)}{2 \tau},
\]

where \( \Delta P \) is the overall pressure drop in the system, \( \Delta P_{\text{res}} \) and \( \Delta P_0 \) are the pressure drops based on the fully developed flow in the reservoir and the small channel, respectively.

#### 5.1.1. 4:1 planar contraction

In the 4:1 contraction flow, the simulations for the Newtonian fluid (arachidyl alcohol) are shown in comparison with the experiment for apparent shear rate \( \dot{\gamma}_0 = V_{\text{avg}}/H_0 \) in 10.8 s\(^{-1}\) in Fig. 5. Fig. 5a shows the streamlines as found out from the simulations, while Fig. 5b exhibits the streamlines of the experimental flow as observed by Olson and Fuller [5].

Olson and Fuller [5] have not shown streamline patterns for higher shear rates but report that no observable recirculation was present. Our simulation results for higher apparent shear rates corroborate their observation, as shown in Fig. 6a–c. The vortices at the salient corners disappear as the apparent shear rates become (a) 43.1 s\(^{-1}\), (b) 86.2 s\(^{-1}\), and (c) 107.8 s\(^{-1}\). From (b) to (c), the changes are negligible.

#### 5.1.2. 4:1 planar expansion

In the 4:1 expansion flow, the simulations for the Newtonian fluid (arachidyl alcohol) are shown in comparison with the experiments at apparent shear rates of 43.1 s\(^{-1}\), 86.2 s\(^{-1}\), and 107.8 s\(^{-1}\), in Fig. 7, respectively. Fig. 7a1–a3 show the streamlines as found out from the simulations, while Fig. 7b1–b3 exhibit the streamlines of the experimental flow as observed by Olson and Fuller [5].

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**Table 2**

Finite element meshes and their characteristics used in the 4:1 planar flow simulations.

<table>
<thead>
<tr>
<th>Mesh</th>
<th>Flow type</th>
<th>Lengths</th>
<th>No. of elements</th>
<th>No. of nodes</th>
<th>No. of DOF</th>
</tr>
</thead>
<tbody>
<tr>
<td>M1</td>
<td>Contraction</td>
<td>(-16,+30)</td>
<td>640</td>
<td>2065</td>
<td>4843</td>
</tr>
<tr>
<td>M2</td>
<td>Expansion</td>
<td>(-4,+16)</td>
<td>512</td>
<td>1649</td>
<td>3867</td>
</tr>
<tr>
<td>M3</td>
<td>Contraction</td>
<td>(-16,+16)</td>
<td>1740</td>
<td>7155</td>
<td>15606</td>
</tr>
<tr>
<td>M4</td>
<td>Expansion</td>
<td>(-8,+16)</td>
<td>1740</td>
<td>7155</td>
<td>15606</td>
</tr>
<tr>
<td>4×M3</td>
<td>Contraction</td>
<td>(-16,+16)</td>
<td>6960</td>
<td>28229</td>
<td>61059</td>
</tr>
<tr>
<td>4×M4</td>
<td>Expansion</td>
<td>(-8,+16)</td>
<td>6960</td>
<td>28229</td>
<td>61059</td>
</tr>
</tbody>
</table>

---

**Fig. 5.** (a) Predicted streamlines for the Newtonian fluid (arachidyl alcohol) for apparent shear rate of 10.8 s\(^{-1}\) and (b) experimental streamlines observed by Olson and Fuller [5] for the same apparent shear rate in flow through a 4:1 planar contraction.
The effects of inertia ($Re \neq 0$) are apparent in producing strong vortex activity in the expansion. It is seen that the vortex activity is increased with apparent shear rate (or Reynolds number), and the agreement between theory and experiment is rather good for the whole range of experimental rates.

5.1.3. Overall results

The overall results for the Newtonian fluid are given collectively in Table 3, while they are shown in Figs. 8–10, for the dimensionless vortex length, vortex intensity and entrance correction as a function of the apparent shear rate. For the creeping flow case ($Re = 0$), they compare well with the values known for the abrupt 4:1 planar contraction to be in the ranges of 0.14–0.18, 0.001–0.0013, and 0.34–0.38, respectively, given in the literature (see, for example, Ref. [22]). For creeping flow, the results are the same for both flow cases. The effect of inertia is to reduce the vortex size for the 4:1 planar contraction but to increase it substantially in a 4:1 planar expansion, as the apparent shear rate increases (see Fig. 8). Quantitatively, and for the 4:1 expansion, the vortex size is in good agreement with the experimentally observed. For the 4:1 contraction, its minute size did not allow a good quantitative measurement experimentally.

The vortex intensity is only found by simulations and is very small for Newtonian fluids, as shown in Fig. 9. Typically, it is in the order of $10^{-2}$ to $10^{-3}$ in the contraction, but it gets higher for the expansion, but at much higher apparent shear rates ($\dot{\gamma} > 100 \text{ s}^{-1}$, $Re > 1$).

Finally, the dimensionless excess pressure losses in the systems, known as entrance correction, also show opposite trends in the two geometries. While for the contraction there is a linear increase with apparent shear rate, for the expansion there is a linear decrease, going even below 0 at some value (here at $\dot{\gamma} > 40 \text{ s}^{-1}$). This is the direct result of the vortex kinematics, which creates a sub-pressure, and as a result less pressure is needed to push the fluid through (see Fig. 10). The contraction results are also in good agreement with results given some time ago by asymptotic analysis [23] and used by Olson and Fuller [5].

5.2. Viscoelastic monolayer

5.2.1. 4:1 planar contraction

In the 4:1 contraction flow, the simulations for the viscoelastic monolayer (PODMA) are shown in comparison with the experiments at various apparent shear rates of 2.7 $\text{s}^{-1}$, 5.4 $\text{s}^{-1}$, and 21.8 $\text{s}^{-1}$, in Fig. 11. Again inertia terms have been included with the effective surface density $\rho_{eff}$. Fig. 11a1–a3 show the streamlines as found out from the simulations, while Fig. 11b1–b3 exhibit the streamlines of the experimental flow as observed by Olson and Fuller [5].

We observe that the vortex size increases with apparent shear rate, up to the highest rate for which stable experimental patterns were found (21.8 $\text{s}^{-1}$). Beyond that value the 2D steady-state simulations showed an eventual decrease in the vortex size and intensity, while the experiments showed an oscillating flow with an almost constant vortex size.

It is evident from the simulations that the strong vortices exhibited by the elasticity of PODMA are rather well captured by the K-BKZ/Olley model. It should be emphasized that this was possible after many trials to fix the elongational response of the model to obtain the substantial strain-hardening in planar extension (two orders of magnitude higher than the Trouton ratio of 4, see Fig. 3b). Thus, the K-BKZ/Olley model performs well also in planar flows, and corrects the deficient behavior of the PSM model, which predicts strain-thinning in planar extension. It is well known that strong extension thickening is responsible for the production of large vortices in contraction flows for branched polymers (LDPE melts), both in axisymmetric [8] and planar [11] contractions. The Olley modification has exactly this ability, i.e. to predict strong strain-hardening behavior in the planar (and biaxial) extensional viscosity.

### Table 3

Results for the planar 4:1 contraction and 4:1 expansion for different flow rates for the Newtonian monolayer (arachidyl alcohol).

<table>
<thead>
<tr>
<th>$\dot{\gamma}$ ($\text{s}^{-1}$)</th>
<th>$Re^a$</th>
<th>X contraction</th>
<th>X expansion</th>
<th>$n_{in}$ contraction</th>
<th>$n_{in}$ expansion</th>
<th>$n_{in}$ [23] contraction</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.7</td>
<td>0</td>
<td>0.175</td>
<td>0.174</td>
<td>0.369</td>
<td>0.375</td>
<td>0.490</td>
</tr>
<tr>
<td>2.7</td>
<td>0.028</td>
<td>0.162</td>
<td>0.187</td>
<td>0.406</td>
<td>0.348</td>
<td>0.521</td>
</tr>
<tr>
<td>5.4</td>
<td>0.0559</td>
<td>0.152</td>
<td>0.197</td>
<td>0.440</td>
<td>0.322</td>
<td>0.552</td>
</tr>
<tr>
<td>10.8</td>
<td>1.12</td>
<td>0.129</td>
<td>0.221</td>
<td>0.507</td>
<td>0.270</td>
<td>0.613</td>
</tr>
<tr>
<td>21.6</td>
<td>2.24</td>
<td>0.097</td>
<td>0.274</td>
<td>0.654</td>
<td>0.170</td>
<td>0.736</td>
</tr>
<tr>
<td>43.2</td>
<td>4.47</td>
<td>0.063</td>
<td>0.395</td>
<td>0.947</td>
<td>0.011</td>
<td>0.982</td>
</tr>
<tr>
<td>86.2</td>
<td>8.93</td>
<td>0.042</td>
<td>0.665</td>
<td>1.547</td>
<td>0.308</td>
<td>1.472</td>
</tr>
<tr>
<td>107.2</td>
<td>11.2</td>
<td>0.036</td>
<td>0.814</td>
<td>1.837</td>
<td>0.426</td>
<td>1.718</td>
</tr>
</tbody>
</table>

* Based on an effective surface density $\rho_{eff} = 0.0029 \text{ kg/m}^2$. 
Fig. 7. (a) Predicted streamlines for the Newtonian fluid (arachidyl alcohol) for different apparent shear rates and (b) experimental streamlines observed by Olson and Fuller [5] for the same apparent shear rates in flow through a 4:1 planar expansion.
Fig. 8. Dimensionless vortex length for the Newtonian fluid (arachidyl alcohol) as a function of the apparent shear rate in a 4:1 planar contraction and expansion. Symbols are experimental observations by Olson and Fuller [5].

5.2.2. 4:1 planar expansion

In the 4:1 expansion flow, the simulations for the viscoelastic monolayer (PODMA) are shown in comparison with the experiments at the much higher apparent shear rates of 43.1 s\(^{-1}\), 86.2 s\(^{-1}\), and 107.8 s\(^{-1}\), in Fig. 12. Fig. 12a1–a3 show the streamlines as found out from the simulations, while Fig. 12b1–b3 exhibit the streamlines of the experimental flow as observed by Olson and Fuller [5].

As opposed to the contraction flow, the expansion flow is much easier to solve and reaches much higher rates with little difficulty. Here, the viscoelastic material relaxes in the reservoir its high stresses coming out of the small channel. We observe that the small Newtonian-like vortex increases with apparent shear rate, but this increase is modest. Actually, the viscoelastic nature of the mono-

layer tends to reduce the large inertial vortices observed with the Newtonian fluid (see Fig. 7). The comparison with the experimental observations is again favorable and seems to capture the main features of the expansion flow.

5.2.3. Overall results

The overall results for the PODMA viscoelastic monolayer are shown in Figs. 13–15, for the dimensionless vortex length, vortex intensity and entrance correction as a function of the apparent shear rate.

The effect of viscoelasticity is to increase the vortex size for the 4:1 planar contraction much faster than in a 4:1 planar expansion, as the apparent shear rate increases (see Fig. 13). Qualitatively, the vortex size increase is in agreement with the experimentally observed. However, the results of the simulations underestimate the vortices, giving a conservative evaluation of their size. It should be noted, however, that in the experimental data there are error bars, that may reach ±0.1 for the average point, and these difficulties to estimate the vortex size have been pointed out [5]. In the simulations, the point where the shear stress changes sign is used for the calculation of \(L_v\) [11]. Also, at low flow rates, the simulation results go asymptotically to the Newtonian creeping values, as expected.

The vortex intensity is only found by simulations and it rises very rapidly for the contraction flow, reaching 3% of the flow rate at the highest stable conditions of 21.8 s\(^{-1}\), as shown in Fig. 14. In the expansion flow, it is one order of magnitude less, and again it increases rapidly but at much higher apparent shear rates (\(\dot{\gamma}_0 > 100\) s\(^{-1}\)).

Finally, the dimensionless entrance correction shows opposite trends in the two geometries, as was the case with the Newtonian fluid (see Fig. 15). While for the contraction there is a rapid increase with apparent shear rate, for the expansion there is a gradual decrease, but always positive in the range of experimental data. Again, this behavior is the direct result of the vortex kinematics. Namely, in the contraction an extra pressure drop is needed to push the strain-thickening fluid through, while in the expansion a sub-pressure is created, and as a result less pressure is needed to push the fluid through.
Fig. 1. (a) Predicted streamlines for the viscoelastic fluid (PODMA) for different apparent shear rates and (b) experimental streamlines observed by Olson and Fuller [5] for the same apparent shear rates in flow through a 4:1 planar contraction.
Fig. 12. (a) Predicted streamlines for the viscoelastic fluid (PODMA) for different apparent shear rates and (b) experimental streamlines observed by Olson and Fuller [5] for the same apparent shear rates in flow through a 4:1 planar expansion.
Overall, it is encouraging that the model with the predictions of Fig. 3 can predict the strong vortices for the PODMA monolayer. Any discrepancies may be attributable to lack of any reliable extra information on the material, supposing of course that the effective surface density assumption holds. For example, apart from lacking any extensional data, there is also lack of good data for the storage modulus $G'$ and the associated first normal stress difference $N_1$ in shear flow. Indeed, Olson and Fuller [5] describe the difficulty of obtaining good data for these material functions, and Olson [24] has measured negative values for $G'$ for many frequencies. Obviously, more work would be required to elucidate this phenomenon.

6. Concluding remarks

Finite element simulations have been undertaken for the flow of Langmuir monolayers in the benchmark problems of entry flow through a 4:1 planar contraction and a 4:1 planar expansion. The monolayers used are a Newtonian fluid and a polymeric viscoelastic fluid (PODMA). The test fluids have been modeled by the viscous Newtonian model and the viscoelastic K-BKZ/Olley model with a spectrum of 8 relaxation times, which fits well shear viscosity data. Lack of data for the rest of the material functions have obliged us to make some assumptions, such as the behavior in extension and the normal stresses in shear.

The Newtonian monolayer is rather well simulated in both contraction and expansion, assuming a value for an effective surface density (which may be seen as taking into account interactions between monolayer and subphase) to get an appropriate Reynolds number and hence inertia effects. The vortex disappears in the contraction with increasing apparent shear rate (hence Reynolds number). The opposite is true in the expansion, where strong vortex activity is found both experimentally and numerically. The rest of the results concern the vortex size and intensity, and the excess pressure losses in the two benchmark geometries.

The viscoelastic monolayer, modeled by the integral K-BKZ equation with a suitable modification by Olley to account for strain-hardening in planar extension, follows closely the experimental flow observations. The model captures well the overall flow behavior, although it underestimates somewhat the vortex size in both geometries. It is argued that accurate data for the normal stresses in shear and extensional data are needed, something which is still lacking and may be difficult to measure. Then, the simulations offer an attractive alternative, since the effect of different parameters can be studied, as was done here for the planar extensional viscosity to help capture the vortex behavior in this kind of flow field, where both shear and extensional properties play an important combined role.

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References


