Optimization of flow-focusing devices for homogeneous extensional flow

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We present a methodology for the shape optimization of flow-focusing devices with the purpose of creating a wide region of homogeneous extensional flow, characterized by a uniform strain-rate along the centerline of the devices. The numerical routines employed include an optimizer, a finite-volume solver, and a mesh generator operating on geometries with the walls parameterized by Bézier curves. The optimizations are carried out for devices with different geometric characteristics (channel aspect ratio and length). The performance of the optimized devices is assessed for varying Reynolds numbers, velocity ratio between streams, and fluid rheology. Brownian dynamics simulations are also performed to evaluate the stretching and relaxation of λ -DNA molecules in the devices. Overall, the optimized flow-focusing devices generate a homogeneous extensional flow over a range of conditions typically found in microfluidics. At high Weissenberg numbers, the extension of λ -DNA molecules in the optimized flow-focusing devices is close to that obtained in an ideal planar extensional flow with an equivalent Hencky strain. The devices presented in this study can be useful in microfluidic applications taking advantage of homogeneous extensional flows and easy control of the Hencky strain and strain-rate. *Published by AIP Publishing.* <https://doi.org/10.1063/1.5037472>

I. INTRODUCTION

Extensionally dominated flows can be found in many natural processes. They are important in the compression of the synovial fluid inside articulations,¹ in blood circulation through stenosis or vasoconstrictions,² and also inside spiders' silk glands,³ among other examples. Similarly, several engineering applications are based on extensionally dominated flows. For example, such flows are used to stretch DNA molecules in single-molecule studies^{4,5} and for DNA mapping.⁶ The molecular extension imposed by such flows is on the basis of microfluidic extensional rheometry,^{7,8} which, beyond critical conditions, gives rise to the study of elastic instabilities.^{8,9} Biological cells can also be stretched in such type of flows for the assessment of mechanical properties,^{10,11} and phenotyping methods were already proposed to classify cells based on the cellular deformation observed.¹² Microfluidic interfacial tensiometry is another method making use of extensional flows, in this case to deform droplets with the aim of measuring the interfacial tension.^{13,14} Therefore, studying such ubiquitous flows and being able to generate them under controlled conditions is of considerable importance.

Extensionally dominated flows can be generated in different types of geometries. The devices frequently used for this purpose, in microfluidics, are converging/diverging channels,^{15,16} cross-slot devices,^{5,17} flow-focusing devices,^{18–20} T-junctions,²¹ and four-roll mill analog devices.²² Nevertheless, the extensional flow in such devices is frequently non-homogeneous (variable strain-rate), contains a non-negligible shear component, and is often limited to a small region of the device. The combination of these factors results frequently in poorly controlled and non-ideal flow conditions. The difficulty in generating pure extensional flows was recognized by Binding and

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Walters,²³ who also pointed out the need and importance of efforts directed to approach, as close as possible, these ideal flows.

Numerical optimization methods were proposed in an attempt to minimize these issues by adjusting the shape of microfluidic devices, in order to obtain a wide region of homogeneous extensional flow, typically near the centerline of the devices.^{24–26} These shape optimization methods were applied to two-dimensional (2D)²⁴ and three-dimensional (3D)²⁵ cross-slot devices and also to converging/diverging channels.²⁶ However, to the best of our knowledge, the numerical optimization of flow-focusing devices has still not been reported. These devices display some advantages compared to cross-slot and contraction geometries. For example, the Hencky strain can be dynamically controlled by simply adjusting the velocity ratio between the inlet streams, while the Hencky strain in a contraction device is constrained by its geometric dimensions. Flow-focusing devices also have the inherent capability to keep a given entity aligned at the center-plane (reproducible strain record), being also able to behave as mixers. Thus, finding the (non-trivial) optimal shape for homogeneous extensional flows in these devices is a relevant task which can potentially improve several applications that previously used non-optimized shapes.^{18–20,27} For example, Arratia *et al.*²⁷ attempted to measure the relaxation time of polymeric solutions by monitoring the filament thinning in a standard (non-optimized) flow-focusing device, making use of an analytical expression that assumes a constant and uniform strain-rate throughout the filament. These assumptions are rough approximations in a standard flow-focusing device but can be better ensured in a device specifically optimized for such purposes, as the ones sought in the present study.

In this work, we present the shape optimization of flow-focusing devices, with the purpose of creating a wide and delimited region of homogeneous extensional flow along the centerline of such devices. The optimizations are carried out for flow-focusing devices with different channel aspect ratios and different lengths in the extensional flow region, in order to widen the range of potential applications. The performance of the optimized devices is assessed in a variety of hydrodynamic conditions to determine the optimal working range of each geometry. Additionally, we use Brownian dynamics simulations to illustrate the application of the optimized devices in single-molecule studies.

The rest of this paper is organized as follows. In Sec. II, we describe the methods used for shape optimization, and Sec. III addresses the details of the Brownian dynamics simulations. The results obtained are presented and discussed in Sec. IV, and Sec. V summarizes the main outcomes of this study.

II. SHAPE OPTIMIZATION METHODS

A. Overview

The sequence of operations in the optimization loop adopted in this work is depicted in Fig. 1. For each iteration of this loop, the geometry and mesh are built for a given set of design parameters, the flow field is computed, and a cost function is evaluated based on the computed velocity field. The cost function value is then fed as the input in a black-box optimizer, which returns on the output a new set of design parameters (new trial geometry) aiming to minimize the cost function. These steps are repeated until the cost function drops below a minimum value previously established, once the variation in the design parameters drops below a prescribed threshold, or once the number of iterations exceeds a predefined limit. Sections II B–II E discuss in more detail each of the steps identified in Fig. 1.

B. Geometry parameterization

The flow-focusing geometry being optimized in this work is represented in Fig. 2. Due to symmetry relative to the Oxz plane, only half of the geometry is represented. Symmetry also holds in the Oxy plane, although the whole cross section is represented in the inset of Fig. 2. The four arms

FIG. 1. Schematic representation of the shape optimization loop.

of the geometry display a width $2H$ in the straight region, and the height (h) is uniform all over the geometry. The channel aspect ratio in the straight region of the arms is defined as $AR = h/(2H)$.

The two red curves in Fig. 2 are Bézier curves and correspond to the walls of the geometry that can be modified in order to minimize the cost function. The Cartesian coordinates $\mathbf{x} = (x, y, z)$ of the points lying in an n degree Bézier curve can be obtained from

$$\mathbf{x}(t) = \sum_{k=0}^n {}^nC_k (1-t)^{n-k} t^k \mathbf{P}_k, \quad 0 \leq t \leq 1, \quad (1)$$

where nC_k represent the binomial coefficients and $\mathbf{P}_k$ are the Cartesian coordinates of the $(n+1)$ control points. In general, Bézier curves only interpolate control points at the ends. The first Bézier curve is defined by points $\mathbf{P}_0$ – $\mathbf{P}_6$, but only the inner points ($\mathbf{P}_1$ – $\mathbf{P}_5$) are design variables. These five inner points are at fixed angles and can only be displaced radially, as illustrated in Fig. 2. The second Bézier curve is defined by points $\mathbf{P}_7$ – $\mathbf{P}_9$, and only the radial coordinate of point $\mathbf{P}_8$ is a design variable. Thus, for each geometry, there are six design variables, corresponding to the radial coordinate of points $\mathbf{P}_1$ – $\mathbf{P}_5$ and $\mathbf{P}_8$.

In this work, we optimized geometries with different channel aspect ratios, both for two-dimensional (2D) and for three-dimensional (3D) channels with $AR = \{0.5, 1, 2, 4\}$. The desired length for the extensional flow region was also varied by changing the angle of point $\mathbf{P}_0$, θ_0 . The pairs $\{AR; \tan(\theta_0)\}$ for all these conditions are presented in Table I, where we also attribute names to the geometries resulting from each set of conditions. Independent of these conditions, the following constraints hold for all geometries: $y_0 = H$, $\mathbf{P}_6 = (H, 14H, z)$, $\mathbf{P}_7 = (-H, 2H, z)$, $\theta_8 = 3\pi/4$, and $\mathbf{P}_9 = (-2H, H, z)$. Note that points $\mathbf{P}_1$ – $\mathbf{P}_5$ are uniformly distributed between points $\mathbf{P}_0$ and $\mathbf{P}_6$ in the azimuthal direction; i.e., the angle between consecutive points is $\Delta\theta = (\theta_6 - \theta_0)/6$ and depends on the value of $\tan(\theta_0)$ for each geometry.

The geometries and (unstructured) meshes used in the optimization routines are automatically generated in the Salome environment (<http://www.salome-platform.org/>), an open-source CAD

FIG. 2. Parameterized flow-focusing device used in the optimizations. The channel width of the arms in the straight, non-optimized region is $2H$ and the height is uniform (h) in the whole geometry. We define the geometry aspect ratio as $AR = h/(2H)$. Points P_0 – P_6 define one Bézier curve, whereas another Bézier curve is defined by points P_7 – P_9 . The extreme points in each of the Bézier curves (black squares) have fixed coordinates, while the remaining points (red circles) can be moved radially. Cartesian (x, y, z) and cylindrical (r, θ, z) coordinate systems are represented, both centered at the intersection between the lines connecting opposite channel entries (departing from the geometrical center of each entrance face). A symmetry plane is considered at $y=0$ (dashed line). The geometry displays three inlets (west, north, and south; the last is omitted due to the symmetry plane) and one outlet (east).

software. A fixed grid size independent of the trial geometry was used, typically $\Delta s = H/15$. This value of Δs used in the optimization cycle resulted in a good compromise between accuracy and computational time when solving for the flow field. However, we should note that all the results that we present in Sec. IV have been computed in more refined meshes, for which mesh-independency is ensured.

C. Flow solver

After the mesh is built, we compute the flow field using the open-source OpenFOAM[®] toolbox, which is based on the finite-volume method. We assume an isothermal, laminar flow and an incompressible fluid. In such conditions, the governing equations are mass conservation,

$$\nabla \cdot \mathbf{u} = 0, \quad (2)$$

and momentum balance,

$$\rho \left(\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} \right) = -\nabla p + \nabla \cdot (\boldsymbol{\tau}_S + \boldsymbol{\tau}), \quad (3)$$

TABLE I. Geometric characteristics and names of the flow-focusing devices optimized in this work.

Geometry	AR	$1/\tan(\theta_0)$
2D_14H	—	14
3D_AR4_14H	4	14
3D_AR2_14H	2	14
3D_AR1_14H	1	14
3D_AR1_10H	1	10
3D_AR1_5H	1	5
3D_AR1_2H	1	2
3D_AR0.5_14H	0.5	14

where $\mathbf{u} = (u, v, w)$ is the velocity vector, t is the time, p is the pressure, ρ is the fluid density, and $\tau_s = \eta_s(\nabla\mathbf{u} + \nabla\mathbf{u}^T)$ and τ represent, respectively, the solvent and polymer contributions to the stress tensor. The momentum equation is written in a general form, which is valid for both generalized Newtonian fluids [$\tau = \mathbf{0}$ and $\eta_s = \eta(\dot{\gamma})$] and viscoelastic fluids. However, the optimizations have been carried out exclusively for Newtonian fluids, i.e., $\tau = \mathbf{0}$ and $\eta_s = \eta = \text{const.}$ Other rheological models are considered later in Sec. IV B 3 to assess the performance of the optimized devices. The Reynolds number, $Re = \frac{\rho U_E R_h}{\eta}$, computed with the average velocity in the outlet (east) arm, U_E , is typically below unity (e.g., $Re = 0.1$) and only the steady-state solution is of interest for the optimization. The hydraulic radius, $R_h = \frac{Hh}{H + 0.5h} = \frac{2AR}{1 + AR}H$, is used in the definition of Re (for 2D cases, we consider $R_h = H$).

As depicted in Fig. 2, the fluid enters the computational domain through the north and south arms, with velocity magnitude U_N , and through the west arm with velocity U_W . The ratio between these two inlet velocities defines an important operational variable of flow-focusing devices, the velocity ratio, $VR = U_N/U_W$. The fluid exits the geometry through the east arm, with an average velocity $U_E = U_W + 2U_N$. At inlet boundaries, uniform velocity profiles are assigned, and a zero pressure gradient is assumed. At outlet boundaries, the pressure is fixed, $p = 0$, and the velocity is assumed fully developed. No-slip boundary conditions are imposed at the walls.

The velocity ratio is the parameter of flow-focusing devices that controls the Hencky strain, $\varepsilon_H = \ln(U_E/U_W) = \ln(2VR + 1)$. In order to avoid adding one additional degree of freedom to the optimizations, this parameter was fixed, $VR = 100$, corresponding to $\varepsilon_H \approx 5.3$. A relatively high VR was selected taking into account applications requiring considerable molecular extension, thus justifying the use of a high ε_H . As will be shown later, the geometries optimized at such high VR perform equally well for any $VR \gg 1$.

D. Cost function definition

The main purpose of the optimizations performed in this work is the creation of a region of homogeneous extensional flow in flow-focusing devices with different geometric dimensions (Table I). More specifically, we aim to have a plateau of constant strain-rate, $\frac{\partial u}{\partial x} = \dot{\varepsilon}$, along the centerline of the geometries, over the length predefined in Table I, which is equivalent to having a velocity profile of the form $\mathbf{u}(x, 0, 0) = (\dot{\varepsilon}x + b, 0, 0)$. Thus, the cost function should reflect how far the centerline velocity profile is, for each trial geometry, from this desired profile. This can be expressed in several ways from a mathematical perspective. For example, Zografos *et al.*²⁶ minimized the squared difference between the trial velocity profiles and an imposed one, forcing smooth transitions of velocity at the entrance/exit to/from the extensional flow region in a smooth contraction/expansion geometry. This allows a fine control of the velocity profile, since both the start/end of the profiles and the velocity values are specified in the whole region of interest. A different method is simply to require a linear velocity profile in the intended region, without specifying bounds or values, by minimizing the sum of squared residuals of a linear fit to the trial profiles. This method imposes no restrictions to the velocity profile outside the extensional flow region, and it does not explicitly fix the start/end of the profile such that the strain-rate is also not explicitly controlled. However, as will be shown later, the fixed cross section of the arms at the start/end of the extensionally dominated flow region imposes effective limits to the velocity, resulting, in practice, in a *quasi*-defined strain-rate. The mathematical expression of the cost function implemented is

$$c = \sum_{i=1}^m (u_{\text{trial},i} - u_{\text{fit},i})^2, \quad 0 \leq x \leq x_0, \quad (4)$$

where m is the number of equidistant sampling points along the centerline, whose location is held constant for each geometry in Table I, and x_0 is the x -coordinate of point $\mathbf{P}_0$. For example, we used

$m = 100$ for $\tan(\theta_0) = 1/14$. The term inside parentheses in Eq. (4) represents the residual of the linear fit at point i , where $u_{\text{trial},i}$ is the velocity computed numerically at point i for a given trial shape, and $u_{\text{fit},i}$ is the velocity predicted at that same point by the linear model fitted to the center-line velocity profile for that trial shape.

E. Black-box optimization algorithm

The search for a minimum of Eq. (4) was performed with NOMAD, an open-source black-box optimization software implementing the Mesh Adaptive Direct Search (MADS) algorithm.^{28,29} This optimizer does not require defining the derivatives of the cost function, being suitable for this work. The optional metaheuristic search provided by the optimizer to escape from local minima has been enabled and limited to 20% of the number of cost function evaluations, although not even this option can guarantee that the global minimum is found. The optimization cycle was stopped when the variation between consecutive trials of all the six design variables ($|\Delta r_i|$, $i = \{1, 2, 3, 4, 5, 8\}$) dropped below $0.01H$, or after reaching 1000 trial geometries. In general, 500 to 1000 trial geometries were needed to find the optimal design for each set of conditions in Table I.

The shape fed as an initial guess to the optimizer was either a straight (sharp corners) flow-focusing or an optimized shape previously found for a related set of conditions (this option improved the convergence in some cases).

III. BROWNIAN DYNAMICS SIMULATIONS

The optimized flow-focusing devices display a set of characteristics which make them affordable for single-molecule studies. Brownian dynamics simulations were carried out in order to evaluate the performance of the devices in the stretching of polymer molecules. The numerical algorithm used is described next.

A. Bead-spring model

The polymer molecules are represented by a bead-spring coarse-grained model, since this approach has proved to reproduce, at least qualitatively, most of the experimental observations for dilute solutions of flexible polymers.³⁰ In this model, each molecule is represented by N beads and $N_S = N - 1$ springs (assuming open chains), where each spring connects two consecutive beads. The position of all the N beads is contained in an $N \times 3$ matrix ($\mathbf{r}$) such that $\mathbf{r}_i$ corresponds to the position vector of bead i . Each bead is connected to other nsp beads through nsp springs. The set of connections for a given bead is denoted by $\mathbf{g}_i$, and group $\mathbf{G}$ contains the set of connections for all the N beads such that $\mathbf{g}_i \subseteq \mathbf{G}$. Each k spring connecting beads a and b is defined by a vector $\mathbf{R}_k = \mathbf{r}_b - \mathbf{r}_a$. Thus, $\mathbf{R}$ is an $(N - 1) \times 3$ matrix containing all spring vectors. For circular chains, not used in this work, the number of springs is equal to the number of beads.

B. Governing equations

The set of stochastic differential equations governing the motion of the beads in the chain is given by³¹

$$d\mathbf{r}_i = \left(\mathbf{u}_f + \sum_{j=1}^N \frac{\partial}{\partial \mathbf{r}_j} \mathbf{D}_{ij} + \sum_{j=1}^N \frac{\mathbf{D}_{ij} \mathbf{F}_j}{kT} \right) dt + \sqrt{2} \sum_{j=1}^i \boldsymbol{\sigma}_{ij} d\mathbf{W}_j, \quad (5)$$

where $\mathbf{u}_f$ is the velocity of the surrounding fluid, $\mathbf{D}_{ij}$ is the diffusion tensor, $\mathbf{F}_j = \mathbf{F}_j^S + \mathbf{F}_j^{\text{EV}}$ is the sum of spring ($\mathbf{F}_j^S$) and exclusion volume forces ($\mathbf{F}_j^{\text{EV}}$), $\boldsymbol{\sigma}_{ij}$ is a tensor resulting from the decomposition of the diffusion tensor that verifies the fluctuation-dissipation theorem, k is Boltzmann's constant, T is the absolute temperature, and $d\mathbf{W}_j$ is an independent Wiener process³² that can be

approached by $d\mathbf{W}_j = \sqrt{dt} \mathbf{n}_j$, with $\mathbf{n}_j$ being a randomly distributed Gaussian vector with zero mean and unit variance. Equation (5) results from the balance between drag, spring, exclusion volume, and Brownian forces, taking into account hydrodynamic interactions and neglecting the inertia of each bead. We will briefly explain the nature of each term (more details can be found in Larson,³⁰ Schroeder *et al.*,³¹ and Jendrejack *et al.*³³).

The drag force is responsible for the first term inside parentheses on the right-hand side of Eq. (5). If no other forces were considered, the beads would simply behave as normal fluid elements.

The springs in the chain constrain the movement of the beads. For a given bead i , connected by nsp springs to nsp adjacent beads, as specified in the corresponding set $\mathbf{g}_i$, the resulting spring force is³⁰

$$\mathbf{F}_i^S = \sum_{j \in \mathbf{g}_i} \frac{kT}{\lambda_p} \left(\frac{|\mathbf{X}_{ij}|}{l} - \frac{1}{4} + \frac{1}{4(1 - |\mathbf{X}_{ij}|/l)^2} \right) \frac{\mathbf{X}_{ij}}{|\mathbf{X}_{ij}|}, \quad (6)$$

where $\mathbf{X}_{ij} = \mathbf{r}_j - \mathbf{r}_i$ is the vector connecting bead i to bead j , λ_p is the persistence length, and l is the maximum length of a spring in a fully stretched state. In this case, $\mathbf{X}_{ij} = \mathbf{R}_k$ correspond to the spring vectors, because the summation is over $\mathbf{g}_i$. The force of each individual spring in Eq. (6) is approached by the Marko-Siggia model,³⁴ which limits the length of the springs to l . Therefore, the maximum length of a chain is limited by $L_{\max} = N_S l$, which is commonly known as the molecule contour length.

The exclusion volume forces impose a repulsive potential between beads, and they are represented in this work by the model of Jendrejack *et al.*,^{33,35}

$$\mathbf{F}_i^{\text{EV}} = -\frac{9}{2} v^{\text{EV}} \left(\frac{3}{4\sqrt{\pi}} \right)^3 \frac{kT}{\lambda_p^2 \sqrt{l}} \sum_{j=1}^N \exp \left(-\frac{9}{4} \frac{|\mathbf{X}_{ij}|^2}{\lambda_p l} \right) \mathbf{X}_{ij}, \quad (7)$$

where v^{EV} is the exclusion volume parameter.

The Brownian force accounts for the thermal agitation of the beads and is represented by the last term on the right-hand side of Eq. (5). According to the fluctuation-dissipation theorem, tensors $\boldsymbol{\sigma}$ and $\mathbf{D}$ are related by³⁵

$$\mathbf{D} = \boldsymbol{\sigma} \boldsymbol{\sigma}^T, \quad (8)$$

where both $\boldsymbol{\sigma}$ and $\mathbf{D}$ are $N \times N$ tensors, with each ij component corresponding to a 3×3 tensor. The decomposition of $\mathbf{D}$ to obtain $\boldsymbol{\sigma}$, according to Eq. (8), can be performed by several methods, differing in computational speed, accuracy, and generality.^{36–38} In this work, we use Cholesky decomposition for its simplicity, generality, and unconstrained application, at the expense of a higher computational cost.

The hydrodynamic interactions account for the disturbances in the local fluid velocity caused by each bead. These interactions are included in the model by considering anisotropic (conformation-dependent) diffusion, embodied by tensor $\mathbf{D}$. Several models are available to express the components of $\mathbf{D}$ as a function of the chain conformation (beads' positions), and in

this work, we select the Rotne–Prager–Yamakawa (RPY) model³⁹

$$\mathbf{D}_{ij} = \begin{cases} \frac{kT}{6\pi\eta a} \mathbf{I} = D\mathbf{I}, & i = j \\ \frac{3D}{4} \frac{a}{|\mathbf{X}_{ij}|} \left[\left(1 + \frac{2a^2}{3|\mathbf{X}_{ij}|^2} \right) \mathbf{I} + \left(1 - \frac{2a^2}{|\mathbf{X}_{ij}|^2} \right) \frac{\mathbf{X}_{ij}\mathbf{X}_{ij}}{|\mathbf{X}_{ij}|^2} \right], & i \neq j \wedge |\mathbf{X}_{ij}| \geq 2a, \\ D \left[\left(1 - \frac{9|\mathbf{X}_{ij}|}{32a} \right) \mathbf{I} + \frac{3}{32a} \frac{\mathbf{X}_{ij}\mathbf{X}_{ij}}{|\mathbf{X}_{ij}|} \right], & i \neq j \wedge |\mathbf{X}_{ij}| < 2a \end{cases} \quad (9)$$

where $\mathbf{I}$ is the 3×3 unit tensor and $D = \frac{kT}{6\pi\eta a}$ is the isotropic diffusion coefficient, which is commonly used in the free-draining approach and that corresponds to the Stokes-Einstein relation for the diffusion of spherical bodies. In fact, the bead-spring model accounting for hydrodynamic interactions degenerates in the free-draining approach if only isotropic diffusion is considered ($\mathbf{D}_{ij} = \mathbf{0}$, $i \neq j$). Note that η represents the fluid viscosity and a corresponds to the beads' radius ($\xi = 6\pi\eta a$ is the drag coefficient for each bead). For the Rotne–Prager–Yamakawa tensor, $\frac{\partial}{\partial \mathbf{r}_j} \mathbf{D}_{ij} = \mathbf{0}$ in Eq. (5). The intensity of the hydrodynamic interactions can be measured by parameter h^* ^{30,31}

$$h^* = \frac{a}{l} \sqrt{3 \frac{N_{k,s}}{\pi}}, \quad (10)$$

where $N_{k,s} = l/(2\lambda_p)$ is the number of Kuhn steps per spring.

C. Numerical implementation

The use of spring models with limited chain length requires special attention, since that constraint should not be violated during the numerical integration over time. In general, this requires the use of small time steps in explicit time integration schemes or the implementation of implicit/semi-implicit integration schemes, which allow for higher time steps at the cost of a higher computational time per time step. The last option is usually more robust and results in a lower total time of computation, being used in this work, based on the semi-implicit scheme described by Kim and Doyle,⁴⁰ here adapted to handle hydrodynamic interactions. By default, this scheme performs an explicit first-order Euler integration of Eq. (5), but when $\frac{|\mathbf{R}_k|}{l} > \alpha$ for any spring of the chain, the spring force is accounted for implicitly ($\alpha = 0.90$ is typically used in this work).⁴⁰ In this case, we first compute the intermediate beads' positions ($\mathbf{r}_i^*$) resulting from the explicit evaluation of all the right-hand side terms of Eq. (5), except the diagonal spring force

$$\mathbf{r}_i^* = \mathbf{r}_i^t + \Delta t \left[\mathbf{u}_i + \sum_{j=1, j \neq i}^N \frac{\mathbf{D}_{ij}\mathbf{F}_j}{kT} + \frac{\mathbf{D}_{ii}\mathbf{F}_i^{\text{EV}}}{kT} + \frac{\sqrt{2}}{\Delta t} \sum_{j=1}^i \boldsymbol{\sigma}_{ij} d\mathbf{W}_j \right]. \quad (11)$$

The diagonal spring force contribution is then added implicitly

$$\mathbf{r}_i^{t+\Delta t} = \mathbf{r}_i^* + \Delta t \left[\frac{\mathbf{D}_{ii}\mathbf{F}_i^{\text{S}}}{kT} \right]_{t+\Delta t}, \quad (12)$$

resulting in a non-linear system of equations, where the unknown is the matrix of beads' positions at the new time ($\mathbf{r}^{t+\Delta t}$). The non-linear system of equations is solved for each Cartesian coordinate at a time, using the iterative Newton-Raphson method. Without loss of generality, we will describe the procedure for the x -component of $\mathbf{r}^{t+\Delta t}$ (the steps are similar for the other components). We

should note that the decoupling between Cartesian components does not apparently impair the convergence of the Newton-Raphson method.

Firstly, we define a function $f_{i,x}$ using Eqs. (6), (9), and (12) such that

$$f_{i,x} = r_{i,x} - r_{i,x}^* - \Delta t \frac{D}{\lambda_P} \sum_{j \in \mathbf{g}_i} \left(\frac{|\mathbf{X}_{ij}|}{l} - \frac{1}{4} + \frac{1}{4(1 - |\mathbf{X}_{ij}|/l)^2} \right) \frac{r_{j,x} - r_{i,x}}{|\mathbf{X}_{ij}|}. \quad (13)$$

The Newton-Raphson method seeks to find a solution $r_{i,x}$ such that $f_{i,x} = 0$, leading to $r_{i,x} = r_{i,x}^{t+\Delta t}$. We recall that $|\mathbf{X}_{ij}| = \sqrt{(r_{j,x} - r_{i,x})^2 + (r_{j,y} - r_{i,y})^2 + (r_{j,z} - r_{i,z})^2}$. Accordingly, this root-finding problem is solved by following the direction pointed by the derivatives at each k^{th} iteration and the first step is to approach $f_{i,x} = 0$ [Eq. (13)] by

$$f_{i,x}^k + J_{i,x}^k (r_{i,x}^{k+1} - r_{i,x}^k) = 0 \Leftrightarrow J_{i,x}^k \Delta r_{i,x}^k = -f_{i,x}^k, \quad (14)$$

where function $f_{i,x}^k$ at each iteration is

$$f_{i,x}^k = r_{i,x}^k - r_{i,x}^* - \Delta t \frac{D}{\lambda_P} \sum_{j \in \mathbf{g}_i} \left(\frac{|\mathbf{X}_{ij}^k|}{l} - \frac{1}{4} + \frac{1}{4(1 - |\mathbf{X}_{ij}^k|/l)^2} \right) \frac{r_{j,x}^k - r_{i,x}^k}{|\mathbf{X}_{ij}^k|}, \quad (15)$$

and $\mathbf{J}_x^k$ is the Jacobian of $\mathbf{f}_x^k$ at each iteration,

$$\mathbf{J}_x^k = \begin{bmatrix} \frac{\partial f_{1,x}^k}{\partial r_{1,x}^k} & \frac{\partial f_{1,x}^k}{\partial r_{2,x}^k} & \cdots & \frac{\partial f_{1,x}^k}{\partial r_{N,x}^k} \\ \frac{\partial f_{2,x}^k}{\partial r_{1,x}^k} & \frac{\partial f_{2,x}^k}{\partial r_{2,x}^k} & \cdots & \frac{\partial f_{2,x}^k}{\partial r_{N,x}^k} \\ \vdots & \vdots & \ddots & \vdots \\ \frac{\partial f_{N,x}^k}{\partial r_{1,x}^k} & \frac{\partial f_{N,x}^k}{\partial r_{2,x}^k} & \cdots & \frac{\partial f_{N,x}^k}{\partial r_{N,x}^k} \end{bmatrix}. \quad (16)$$

Matrix $\mathbf{J}_x^k$ is square ($N \times N$) and in most of the cases is sparse, since each bead is only connected to a limited number of beads by springs. For example, considering an open, linear chain without branches, and with the beads numbered consecutively, $\mathbf{J}_x^k$ is a tridiagonal matrix because each bead is connected to only its two neighboring beads, except at the edges, where a single connection exists. For any generic chain topology, each non-zero element of $\mathbf{J}_x^k$ can be expressed by

$$J_{ij,x}^k = \frac{\partial f_{i,x}^k}{\partial r_{j,x}^k} = \begin{cases} \frac{c}{d_{ij}} \left[\left(\frac{1}{4(d_{ij}-1)^2} + d_{ij} - \frac{1}{4} \right) \left(\frac{\delta_{ij}^2}{d_{ij}^2} - 1 \right) - \frac{\delta_{ij}^2}{d_{ij}} \left(1 - \frac{1}{2(d_{ij}-1)^3} \right) \right], & i \neq j \wedge ij \in \mathbf{G} \\ 1 - \sum_{m=1, m \neq i}^N J_{im,x}^k, & i = j \end{cases}, \quad (17)$$

where $c = \Delta t \frac{D}{\lambda_P}$, $d_{ij} = |\mathbf{X}_{ij}^k|/l$, and $\delta_{ij} = (r_{j,x}^k - r_{i,x}^k)/l$. We can observe that $\mathbf{J}_x^k$ is only non-zero in the main diagonal and for the $\{ij, ji\}$ pairs defining a spring, i.e., included in $\mathbf{G}$. Moreover, it is

evident from the first branch of Eq. (17) that $\mathbf{J}_x^k$ is symmetric, as expected physically (the force of each spring affects equally both beads). Lastly, it can be shown that $\sum_{m=1, m \neq i}^N J_{im,x}^k < 0$, thus $\mathbf{J}_x^k$ is a diagonally dominant matrix.

After building $\mathbf{J}_x^k$ and $\mathbf{f}_x^k$, the system of equations (14) can be solved for $\Delta \mathbf{r}_x^k$ and the new vector of solutions is obtained by

$$\mathbf{r}_x^{k+1} = \mathbf{r}_x^k + \Delta \mathbf{r}_x^k. \quad (18)$$

The solution of the system in Eq. (14) is computed efficiently using Thomas' algorithm, due to the fact that $\mathbf{J}_x^k$ is tridiagonal and diagonally dominant for the chains considered in this work.

The steps described above correspond to one iteration of the Newton-Raphson method, for one Cartesian component. In practice, $\mathbf{J}^k$ and $\mathbf{f}^k$ are first computed for the three Cartesian components, and then $\Delta \mathbf{r}^k$ is obtained sequentially for each component. The newly computed $\mathbf{r}^{k+1}$ is then used in the next iteration of the Newton-Raphson method. In the transition between iterations, $\mathbf{r}^{k+1}$ is used to update $\mathbf{F}_{jj \neq i}^S$ in $\mathbf{r}_i^*$ [Eq. (11)]. The remaining terms of $\mathbf{r}_i^*$ are kept frozen during all the iterations. The process is stopped once $\max \left(\frac{|\mathbf{R}^k - \mathbf{R}^{k-1}|}{l} \right) < \varepsilon$, where $\mathbf{R}^k$ and $\mathbf{R}^{k-1}$ correspond to the spring vectors' matrices in consecutive iterations (k and $k-1$) and ε is a relative tolerance, typically set at 10^{-6} (the method converges typically in less than 10 iterations). The last estimate is transferred to the solution, $\mathbf{r}_i^{t+\Delta t} = \mathbf{r}_i^{k+1}$, and the algorithm progresses to the next time step. We should note that in this semi-implicit scheme, the freezing of almost all the terms of $\mathbf{r}_i^*$ [Eq. (11)] during the iterations of the Newton-Raphson method introduces some explicitness. The method can be rendered more implicit by using $\mathbf{r}_i^{t+\Delta t}$ to re-evaluate the second term on the right-hand side of Eq. (13) and applying again the Newton-Raphson method. This internal loop can be repeated multiple times within the same time step until the changes in $\mathbf{r}_i^{t+\Delta t}$ become negligible.

In Eq. (5), the term $\mathbf{u}_f$ corresponds to the fluid velocity evaluated at the current position of the bead. Before explaining the procedure adopted to compute this term, we should note that all the cells of the mesh undergo a tetrahedral decomposition at the beginning of each simulation, where one vertex of the tetrahedron is always the cell's center and the remaining three vertices correspond to vertices of one of the cell's faces. Such decomposition is performed for tracking the particles in generic meshes, where the beads are located by their barycentric coordinates inside a given tetrahedron of a given cell. After the steady-state velocity field of the fluid is computed at cell centers using the finite-volume method described in Sec. II C, these values are interpolated from cell centers to cell vertices using inverse distance weighting. Then, the term $\mathbf{u}_f$ is computed at the bead position using the corresponding barycentric coordinates to weight the velocity values at the vertices of the tetrahedron containing the bead. Therefore, sub-cell resolution can be achieved in the computation of $\mathbf{u}_f$, while also ensuring a smooth spatial variation of this term. It is worth mentioning that the method used to interpolate $\mathbf{u}_f$ from the cell-centered values of a generic mesh has a strong influence on the overall accuracy and stability of the Brownian dynamics algorithm, although further exploiting this subject is out of the scope of this work.

The cases addressed in this work do not involve confinement of the molecules, i.e., the smallest dimension of the geometries is always significantly larger than the molecules' coil size at equilibrium. Moreover, the molecules only occasionally contact with the walls. Therefore, no special treatment of the hydrodynamic interactions was needed near the walls and a simple blocking algorithm was used to reposition any bead that occasionally crossed the wall due to Brownian motion—the beads were returned back to the position where they crossed the wall, in the fluid side.

The solver for Brownian dynamics simulations was implemented in *rheoTool*,^{41,42} using the built-in algorithm of OpenFOAM (v.5.0) for particle tracking in generic meshes.⁴³ The code used is available as open-source therein.⁴¹

D. Model parameters

The molecule selected for the Brownian dynamics simulations was λ -DNA, since it is a well-characterized polymer, widely used in numerical and experimental single-molecule studies. Larson³⁰ discussed the range of physically admissible parameters to represent λ -DNA molecules using bead-spring models. In this work, we use the set of parameters reported by Jendreck *et al.*:³⁵ $N_S = 10$, $\lambda_P = 0.053 \mu\text{m}$, $a = 0.077 \mu\text{m}$ ($h^* = 0.16$), $v^{\text{EV}} = 0.0012 \mu\text{m}^3$, and $D = 0.065 \mu\text{m}^2/\text{s}$, which give a relaxation time of approximately 4.1 s in a 43.3 cP buffer, in good agreement with the value reported by Jendreck *et al.*³⁵ The relaxation time was measured from a fit to the ensemble average relaxation curve of the molecules, as discussed by Larson.³⁰ We assume a contour length, $L_{\text{max}} = 21 \mu\text{m}$, for stained λ -DNA.^{30,35} In these simulations, $H = 50 \mu\text{m}$ is considered for the channel half-width. The time step was adjusted as a function of the Weissenberg number (Wi), Δt (s) = A/Wi , with $A = 0.0029$ s for $Wi < 10$ and $A = 0.0058$ s for $Wi \geq 10$. This empirical relation allowed one to obtain time-step-independent results, without springs overstretch, while keeping an acceptable time of computation.

E. Model verification

The accuracy of the Brownian dynamics solver was assessed prior to the application in the optimized flow-focusing devices. The test case used was the DNA stretching in a planar extensional flow, as reported in the experimental and numerical study of Schroeder *et al.*³¹ This case was selected because it accounts for both exclusion volume and hydrodynamic interactions effects, it allows assessing the accuracy of both transient and steady-state results, and it reproduces the dominant flow-type of flow-focusing devices. The molecules considered are 7λ -DNA concatemers, presenting a contour length of approximately $150 \mu\text{m}$.³¹ The following parameters are used to simulate these molecules:³¹ $N_S = 28$, $\lambda_P = 0.066 \mu\text{m}$, $a = 0.101 \mu\text{m}$ ($h^* = 0.12$), $v^{\text{EV}} = 0.0034 \mu\text{m}^3$, and $D = 0.257 \mu\text{m}^2/\text{s}$, which give a relaxation time of approximately 21 s in an 8.4 cP buffer. Following Schroeder *et al.*,³¹ a second set of parameters was also tested: $a = 0.200 \mu\text{m}$ ($h^* = 0.23$), $v^{\text{EV}} = 0.00032 \mu\text{m}^3$, and $D = 0.130 \mu\text{m}^2/\text{s}$, which give a relaxation time of approximately 29.7 s in an 8.4 cP buffer. A planar extensional flow in the Oxy plane is defined by

$$\nabla \mathbf{u} = \begin{bmatrix} \dot{\epsilon} & 0 & 0 \\ 0 & -\dot{\epsilon} & 0 \\ 0 & 0 & 0 \end{bmatrix},$$

and the fluid velocity at a given bead position is computed analytically, $\mathbf{u}_f = (\dot{\epsilon}x_i, -\dot{\epsilon}y_i, 0)$. The Hencky strain accumulated over time in this flow field is $\epsilon_H = \dot{\epsilon}t$ and we define the Weissenberg number as $Wi = \lambda\dot{\epsilon}$. The molecules are initialized in random conformations and left to equilibrate for a time equal to 20λ in the absence of flow. The results were averaged over an ensemble of 1000 molecules.

The fractional molecular length is plotted in Figs. 3(a)–3(c) as a function of the Hencky strain (time), for $Wi = 0.75, 0.98$, and 4. The molecular length (chain size), L , is defined as the biggest inter-beads distance in the chain, $L = \max(|\mathbf{r}_i - \mathbf{r}_j|)$, $i = \{1 \dots N\}$, $j = \{1 \dots N\}$. The curves are plotted for two different time steps, $\Delta t = 10^{-3} \lambda$ and $\Delta t = 10^{-4} \lambda$, in order to assess the influence of the time step on the results. The steady fractional molecular length for different Wi 's is plotted in Fig. 3(d), for the two values of h^* considered. In general, a good agreement is observed between our numerical results and the numerical predictions of Schroeder *et al.*,³¹ which both reproduce the experimental data reasonably well. In addition, the curves depicted in Figs. 3(a)–3(c) for different time steps are visually indistinguishable, showing time-step independency.

FIG. 3. Fractional molecular length of 7λ -DNA molecules as a function of the Hencky strain for (a) $Wi = 0.75$, (b) $Wi = 0.98$, and (c) $Wi = 4$, in a planar extensional flow (results are for $h^* = 0.12$). The lines represent the numerical results obtained in this work, using different time steps, whereas the symbols are for the experimental and numerical data from Schroeder *et al.*³¹ In panel (d), the steady fractional molecular length is plotted as a function of Wi , for two different h^* . The results from Schroeder *et al.*³¹ are also plotted for comparison.

IV. RESULTS AND DISCUSSION

A. Shapes of the optimized flow-focusing devices

The shapes of the resulting optimized flow-focusing devices are depicted in Fig. 4, where each geometry corresponds to a pair $\{AR; \tan(\theta_0)\}$ specified in Table I (the names of the geometries can also be found therein). The radius of the movable points defining the Bézier curves are listed in Table II for each geometry such that any of the optimized devices obtained in this work can be easily reproduced.

Comparing the optimized flow-focusing devices (Fig. 4) with the original, non-optimized shape (Fig. 4, first panel), it can be seen that the minimization of the cost function is essentially achieved by displacing the downstream walls away from the Cartesian axes. The resulting shapes are non-trivial and most of them exhibit protuberances (except geometries 3D_AR1_10H and

FIG. 4. Shapes of the optimized flow-focusing devices obtained in this work. Correspondence between geometric characteristics $\{AR; \tan(\theta_0)\}$ and names can be found in Table I. Note that the shapes depicted correspond to the cross section of the devices at any plane $z = \text{const.}$

3D_AR0.5_14H), similar to the ones also observed in optimized cross-slot devices.^{24,25} Moreover, the Bézier curve of geometry 3D_AR0.5_14H, the one with lower aspect ratio, closely approaches a hyperbola, which is the expected optimal shape for a Hele-Shaw flow at high VR. The Bézier curve at the upstream corner has a less relevant contribution to the cost function than the Bézier curve at the downstream corner, and this partially explains the small variation of r_8 among the shapes obtained for different geometric parameters (Table II and Fig. 4).

B. Velocity and strain-rate profiles

The velocity profiles presented in this section are normalized by the centerline velocity in the fully developed flow region in the east arm of the devices ($U_{E,c}$). Note that $U_{E,c}$ represents the velocity magnitude, but at the centerline it is equivalent to the x -component of the velocity vector,

TABLE II. Radius of the movable points in the Bézier curves of the optimized flow-focusing devices (see Fig. 2). Each Bézier curve can be reconstructed using these values and the information provided in Sec. II B.

Geometry	r/H					
	P_1	P_2	P_3	P_4	P_5	P_8
2D_14H	5.4911	4.9337	4.9922	9.6512	-1.0331	0.9358
3D_AR4_14H	5.5475	5.9966	5.0522	8.8676	0.7835	1.5000
3D_AR2_14H	5.6408	5.6361	3.8883	9.7175	-0.5650	0.7514
3D_AR1_14H	5.4968	4.4486	5.6538	3.8116	4.1901	0.9003
3D_AR1_10H	4.9540	3.7076	4.4785	5.0700	0.1400	0.9000
3D_AR1_5H	4.2478	1.1252	4.8254	4.9775	-3.8240	0.9000
3D_AR1_2H	2.3175	2.2174	2.4488	4.0665	-4.9906	0.9005
3D_AR0.5_14H	5.3168	3.8090	6.6612	0.7523	7.7649	0.9001

since the remaining velocity components are zero due to symmetry. The strain-rate profiles at the centerline, $\dot{\epsilon} = \partial u / \partial x$, are normalized by $\dot{\epsilon}_P = U_{E,c} / [H / \tan(\theta_0)]$. In addition, all the profiles are taken at the mid-plane ($z = 0$).

In Fig. 5, the curves for $VR = 100$ correspond to the centerline velocity and strain-rate profiles obtained in the hydrodynamic conditions used in the optimizations ($VR = 100$; low Re ; Newtonian fluid), for the different geometries. Overall, the numerical optimization was effective in expanding the extensional flow region to the desired lengths, while keeping a nearly constant strain-rate in that region. The larger oscillations in the strain-rate profiles are observed for the geometries optimized for shorter lengths, $1/\tan(\theta_0) = 2, 5$, and 10 . Still, the maximum deviation relative to the average plateau strain-rate in those cases remains below 7%. We can, however, conclude that the optimization is less effective for shorter extensional regions, which we attribute to the short distance left to the velocity profile to rearrange and recover from any oscillation. In Sec. II D, it was argued that, although the cost function implemented does not fix explicitly the strain-rate value in the plateau region, an indirect constraint exists. In fact, we can see in Fig. 5 that the plateau of the normalized strain-rate profiles lies typically between 0.9 and 1, except in the geometries with a high value of $\tan(\theta_0)$.

Although the flow-focusing devices were optimized for fixed hydrodynamic conditions, it is relevant to know the response (how the centerline velocity and strain-rate profiles behave) of each device to different conditions. Therefore, each geometry was tested for different VR , Re , and fluid rheology, since these are relevant variables that can be changed in the experimental use of the devices.

1. Velocity ratio effect

The velocity and strain-rate profiles at the centerline of each device are plotted in Fig. 5 for $VR = \{5, 10, 50, 100\}$, $Re = 0$ [the convective term is removed from the momentum equation, Eq. (3)] and considering a Newtonian fluid. The velocity and strain-rate profiles for $VR = 100$ and 50 are indistinguishable from each other in all the devices, and the deviation to these profiles becomes only significant below $VR = 10$. Indeed, for $VR = 10$ and 5 , the velocity profiles at the entrance to the extensional flow region ($-3 \lesssim x/H \lesssim 0$) display a small overshoot followed by a stronger undershoot in almost all the devices, which induce some oscillations in the strain-rate profiles. The overshoot and undershoot, respectively, are essentially due to the shrinking of the cross section in the upstream corners and the increase of area in the transition from the west arm to the outlet arm. These effects are not significant at high VR , because the central stream velocity is small compared to the lateral streams, having also a lower penetration in the confluence region. Thus, in general, the optimized devices provide a linear velocity profile for $VR \gtrsim 10$.

2. Inertia effect

The velocity and strain-rate profiles at the centerline of each device are plotted in Fig. 6 for $Re = \{0, 0.1, 1, 10\}$, $VR = 100$ and considering a Newtonian fluid. The profiles are independent of

FIG. 5. Velocity ratio (VR) effect on the centerline velocity and strain-rate profiles in the optimized flow-focusing devices. All profiles were obtained for a Newtonian fluid and $Re = 0$.

Re up to $Re = 1$, corresponding to Stokes flow conditions. For $Re = 10$, inertial effects become significant and the strain-rate plateau deteriorates. The profiles in Fig. 6 also suggest that the geometries optimized for lower aspect ratios are less sensitive to inertia, which is attributed to the confinement effect of the top and bottom walls. Overall, the optimized devices show a Re -independent behavior for $Re \lesssim 1$; above that threshold, the extensionally dominated flow can no longer be considered homogeneous.

3. Fluid rheology effect

For the fluid rheology tests, both power-law and Oldroyd-B fluids were evaluated. A power-law fluid is inelastic but displays shear-thinning/thickening, whereas Oldroyd-B fluids are elastic but have a constant shear viscosity. Thus, shear-dependent viscosity and elasticity effects on the velocity and strain-rate profiles can be isolated using these two constitutive equations.

The shear viscosity of the power-law fluid is given by⁴⁴

$$\eta(\dot{\gamma}) = K\dot{\gamma}^{n-1}, \quad (19)$$

where K is the flow consistency index, n is the power-law index, and $\dot{\gamma} = \sqrt{2\mathbf{D}:\mathbf{D}}$ is the local

FIG. 6. Inertia effect on the centerline velocity and strain-rate profiles in the optimized flow-focusing devices. All profiles were obtained for a Newtonian fluid and $VR = 100$.

strain-rate, with $\mathbf{D} = \frac{1}{2}(\nabla \mathbf{u} + \nabla \mathbf{u}^T)$ representing the rate-of-strain tensor. For creeping flow conditions ($Re \rightarrow 0$), the flow field depends only on n (for fixed VR and geometry).

The differential constitutive equation for the polymeric extra-stress tensor ($\boldsymbol{\tau}$) of an Oldroyd-B fluid can be written as⁴⁵

$$\boldsymbol{\tau} + \lambda \left(\frac{\partial \boldsymbol{\tau}}{\partial t} + \mathbf{u} \cdot \nabla \boldsymbol{\tau} - \boldsymbol{\tau} \cdot \nabla \mathbf{u} - \nabla \mathbf{u}^T \cdot \boldsymbol{\tau} \right) = \eta_p (\nabla \mathbf{u} + \nabla \mathbf{u}^T), \quad (20)$$

where λ is the fluid relaxation time and η_p is the polymeric component of the viscosity, such that $\beta = \frac{\eta_s}{\eta_p + \eta_s}$ defines the solvent viscosity ratio, with η_s representing the solvent (Newtonian) component of the viscosity. For creeping flow conditions and fixed VR , the flow of the Oldroyd-B fluid is controlled by the Weissenberg number, $Wi = \lambda \frac{U_{E,c}}{H/\tan(\theta_0)}$, and β . Note that $\dot{\epsilon}_p = \frac{U_{E,c}}{H/\tan(\theta_0)}$ is approximately the constant strain-rate in the plateau. Further details about the numerical algorithm used for the simulation of viscoelastic fluid flows can be found in Pimenta and Alves.⁴²

FIG. 7. Velocity and strain-rate profiles at the centerline of geometry 3D_AR1_14H for power-law fluids with different power-law indices (n). All profiles were obtained for $VR = 100$ and $Re = 0$.

The results for power-law fluid flows in geometry 3D_AR1_14H are plotted in Fig. 7 for $n = \{0.6, 0.8, 1, 1.2, 1.4\}$, $Re = 0$, and $VR = 100$. We should note that the flow was computed in some of the remaining geometries, and the behaviors were qualitatively similar to the ones illustrated in Fig. 7 (for the sake of conciseness, these results are not plotted). The most noticeable effect of changing the value of n is the increase/decrease of the strain-rate overshoot at the start of the extensional flow region. Further downstream, the strain-rate profile is also affected by the change of n , but the result is essentially a vertical translation of the profile. Hence, we can conclude that the optimized devices can still keep a reasonably homogeneous extensional flow region for weakly shear-thinning/thickening power-law fluids, in the range $0.8 \lesssim n \lesssim 1.2$.

The tests with the Oldroyd-B fluid were carried out for $\beta = 0.5$, representative of semi-dilute Boger fluids, $Wi = \{0, 0.1, 0.2, 0.4, 0.8\}$, $Re = 0$, and $VR = 100$. The centerline velocity and strain-rate profiles are plotted in Fig. 8 for geometry 3D_AR1_14H. For $Wi < 0.4$, the profiles do not deviate significantly from the Newtonian case. However, for $Wi \geq 0.4$, the overshoots in the strain-rate profiles at the start/end of the extensional flow region are enhanced due to pronounced elastic effects. For higher Wi and/or lower β , elasticity increases and these undesired effects are amplified. Moreover, the flow symmetry relative to the Oxz plane is lost above a critical Wi dependent on β (lower β leads to a lower critical Wi —results not shown). This is due to the onset of an elastic instability, which has been reported and investigated in Oliveira *et al.*⁴⁶ Such conditions are not suitable for rheological measurements^{27,47} but can be useful to explore elastic instabilities in extensionally dominated flows.⁴⁸

4. Overview

We have seen that the performance of the optimized flow-focusing devices can be affected up to some extent by several factors, among which the VR , Re (inertia), and fluid rheology were analyzed above. From the perspective of practical applications, the devices' performance should ideally be independent of the operating conditions. In practice, some dependency is inevitable, especially far from the “optimization point,” as it has also been shown for cross-slot²⁵ and contraction/expansion devices.²⁶ In the situations where the performance is not acceptable, the best that one can do is to optimize the geometry specifically for such conditions. This on-demand optimization starts to be a numerically feasible option in view of the increasing availability of computational power and the advance of fabrication techniques, including 3D printing with increasingly better resolution.

FIG. 8. Velocity and strain-rate profiles at the centerline of geometry 3D_AR1_14H for an Oldroyd-B fluid ($\beta=0.5$), at varying Wi . All profiles were obtained for $VR=100$ and $Re=0$.

C. Extension of λ -DNA molecules in an optimized flow-focusing device

Brownian dynamics simulations were only carried out in the 3D_AR1_14H optimized geometry. In order to extract additional information on forced molecular relaxation, this geometry was connected at the outlet to its reflected image on the Oyz plane, as depicted in Fig. 9(a). The boundary conditions imposed while computing the fluid flow (for $Re=0$) ensured an equal average velocity (in absolute value) in each opposite inlet/outlet pair [Fig. 9(a)]. Thus, the velocity profile at the centerline is symmetric relative to the Oyz plane, whereas the strain-rate profile is anti-symmetric [Fig. 9(b)]—stretching flow for $-14.5 < x/H < -0.5$ and compressive flow for $0.5 < x/H < 14.5$. The two symmetric parts of the geometry are connected by a short straight channel spanning $-0.5 < x/H < 0.5$ [Fig. 9(a)]. Unless otherwise stated, 250 molecules were uniformly distributed over the dashed line represented in Fig. 9(a) ($x=-18H$, $-0.9 < y/H < 0.9$, $z=0$) and left to equilibrate in the local flow. This procedure, where we fix artificially the molecules' center of mass at their initial positions, aims to reproduce approximately the molecular extension that would be achieved in a long (west) entry channel, where the flow is shear-dominated. After the molecules reach a steady extension, they are allowed to freely flow. All results presented were obtained from an ensemble average over 4 groups of 250 molecules (a total of 1000 molecules). Note that the molecules are initially distributed over the west channel width (y -direction), but the z -coordinate is similar for all the molecules and corresponds to the mid-plane ($z=0$) of the device. The molecules behavior at other z -coordinates is certainly different, but such analysis is out of the scope of this work, since the homogeneous extensional flow is only kept close to the mid-plane.

The average fractional molecular length is plotted in Fig. 10 as a function of the x -coordinate of the trajectories (all the trajectories converge to the centerline upon entry in the extensional flow region). We use the plateau Weissenberg number to characterize the flow strength, $Wi = \lambda \bar{\epsilon}_{\text{plat}}$, where λ is the relaxation time of the molecules and $\bar{\epsilon}_{\text{plat}}$ is the average strain-rate in the plateau at $-13 < x/H < -2$, in the centerline [the symmetric value is observed in the plateau at $2 < x/H < 13$; see Fig. 9(b)]. The results are shown for $VR=10$ ($\epsilon_H \approx 3$), Fig. 10(a), and $VR=100$ ($\epsilon_H \approx 5.3$), Fig. 10(b). As expected, for $Wi < 0.5$ the fractional length remains nearly constant, since the molecules recoil faster than the flow-induced changes of conformation. Above this critical Wi , the molecules are effectively stretched in the region $x < 0$, they arrive at $x=0$ with their backbone predominantly aligned with the flow (Ox axis), and they relax downstream. The maximum fractional length reached is higher for the higher VR , and the difference between both VR would be even higher if the molecules were not pre-stretched to equilibrium in the west arm flow.

FIG. 9. (a) Double flow-focusing device geometry used in Brownian dynamics simulations. This device is composed by geometry 3D_AR1_14H ($x < 0$) and its reflected image ($x > 0$) in plane Oyz . The two parts of the geometry are connected by a straight channel (red lines in the scheme), between $x = -0.5H$ and $x = 0.5H$. The green arrows point the flow direction and the velocity magnitude in each entrance is also identified, where three inlets and three outlets can be found. The black dashed line, at $x = -18H$, $z = 0$, represents the region where the molecules are initially distributed and released. (b) Velocity and strain-rate profiles at the centerline of the double flow-focusing device represented in (a), for $Re = 0$ and considering a Newtonian fluid. The velocity profile is symmetric relative to the Oyz plane, whereas the strain-rate profile is anti-symmetric. The shaded regions represent the two plateaus of strain-rate, with average values $\bar{\dot{\epsilon}}_{plat}$ and $-\bar{\dot{\epsilon}}_{plat}$.

Interestingly, for $Wi \geq 20$, the fractional length decreases linearly with x in the relaxation region ($x > 0$). The slope of the profiles in these cases follows an affine deformation, which occurs when the molecules deform as the fluid elements. It follows from the definition of strain that $L = L_0 \exp(\epsilon)$ and, for the extensional flow observed in region $x > 0.5H$, it can be shown that $\frac{dL}{dx} = -\frac{\bar{\dot{\epsilon}}_{plat} L_0}{U_0}$ $\approx -\frac{L_0 \tan(\theta_0)}{H(1 + 0.5/VR)}$ is expected for an affine deformation, where U_0 and L_0 are the fluid velocity and the molecular extension at the departure point, respectively; in this case, $x = 0.5H$. The dimensionless slope, $m = -\frac{L_0 \bar{\dot{\epsilon}}_{plat} H}{L_{max} U_0}$, is plotted in Fig. 10 (black dashed line) for the profiles at $Wi = 100$,

FIG. 10. Fractional molecular length at different Wi , for (a) $VR = 10$ and (b) $VR = 100$. The x -axis represents the x -coordinate of the molecules trajectories. The dashed lines represent the slope expected for an affine deformation at $Wi = 100$.

and we can observe a good fit for those curves. Note that the profiles of Fig. 10 are only plotted up to $x = 13H$ because the molecules follow different paths (arms) downstream, leading to a diversity of strain records and molecular extension—some molecules keep relaxing, while others, the majority, start to stretch again.

The homogeneity of the extensional flow is the distinguishing feature claimed for the optimized devices proposed in this work. In order to further assess this property, we compared between the molecular extension obtained in the flow-focusing devices and the one observed in an analytical planar extensional flow with limited Hencky strain. For this comparison, in particular, the molecules in the flow-focusing device were all released from the centerline at $\mathbf{x} = (-18H, 0, 0)$, where the strain-rate upstream to the extensional flow region is negligible, leading to a negligible molecular extension therein. The analytical planar extensional flow was imposed using the following expression for the velocity field:

$$\mathbf{u} = \begin{cases} \bar{\dot{\epsilon}}_{\text{plat}}(x, -y, 0), & 0 < t < t_s \\ -\bar{\dot{\epsilon}}_{\text{plat}}(x, -y, 0), & t_s \leq t \leq 2t_s \end{cases}, \quad (21)$$

where $t_s = \ln(2VR + 1)/\bar{\dot{\epsilon}}_{\text{plat}}$ is the shifting time. The molecules are first equilibrated in no-flow conditions ($t = 0$). In the first branch of Eq. (21), the molecules are stretched by the flow, whereas they are compressed upon switching to the second branch. In practice, the two branches correspond to the same type of flow, and the transition between both only represents a shift of axes, which are rotated by 90° . In fact, after some relaxation in the second branch, the molecules stretch again while they change their orientation by 90° , although this last event is not of interest for this study. The Hencky strain in the first branch of Eq. (21) is made equivalent to the one obtained in the flow-focusing device through an appropriate choice of t_s . The accumulated Hencky strain in this analytical flow is given by

$$\epsilon_H = \begin{cases} \bar{\dot{\epsilon}}_{\text{plat}} t, & 0 < t < t_s \\ \bar{\dot{\epsilon}}_{\text{plat}}(2t_s - t), & t_s \leq t \leq 2t_s \end{cases}. \quad (22)$$

The Hencky strain in the flow-focusing device, for molecules travelling at the centerline, is computed by numerical integration. The results obtained for $VR = 10$ and $VR = 100$, and $Wi = 1, 5$, and 100 are depicted in Fig. 11. In general, the profiles resulting from each approach are in good agreement, but some deviation is observed at low Wi , especially for the case $VR = 100$. This deviation is mainly due to the non-abrupt transitions in the strain-rate profiles of the optimized devices (cf. Fig. 5), which lead to an average strain-rate smaller than the plateau value ($\bar{\dot{\epsilon}}_{\text{plat}}$). In practice, this always results in trajectories at an average Wi smaller than the one computed at the plateau, which is the one effectively imposed in the simulations with the analytical planar extensional flow [Eq. (21)]. This effect is more evident at low Wi , because it is in this region of the $L = f(Wi)$ plot that the rate of change is higher (Fig. 12). Thus, the optimized devices having a longer plateau of constant strain-rate should be especially advantageous in this regard (the relative difference between the average and the plateau strain-rate is smaller). In Fig. 11, we also plot (dashed lines) the results obtained in a non-optimized flow-focusing device ($AR = 1$, $H = 50 \mu\text{m}$, and straight connector of length H) in similar conditions. Since the strain-rate profile in this geometry is bell-shaped (cf. first panel of Fig. 5 for a 2D geometry), the average strain-rate value estimated by integration has been used in the computation of Wi instead of the maximum value, which is nearly 30% higher than the former. Overall, the fractional molecular length profiles obtained in the optimized flow-focusing device are always closer (in average) to the analytical ones than the profiles for the non-optimized flow-focusing device. The difference of performance is more evident for $VR = 100$ and low Wi and would be even higher if we had computed Wi based on the maximum strain-rate value for the non-optimized flow-focusing device case. Thus, the standard (non-optimized) flow-focusing device lacks a strain-rate plateau that could characterize it, and considering its average strain-rate is not sufficient, in general, to ensure an ideal planar extensional flow at that strain-rate, owing to the non-homogeneity of the flow. The plots in Fig. 11 also show the hysteretic

FIG. 11. Fractional molecular length as a function of the accumulated Hencky strain in the double 3D_AR1_14H flow-focusing device (symbols), in a non-optimized double flow-focusing device (dashed lines) and in an analytical planar extensional flow (solid lines). The arrows point the variation of ε_H in each section of the curves, composed by a stretching stage (increasing ε_H) and a relaxation stage (decreasing ε_H). The separation between both stages occurs for $\varepsilon_H = \ln(21) \approx 3$ in (a) and $\varepsilon_H = \ln(201) \approx 5.3$ in (b).

behavior of λ -DNA molecules, since the curves for increasing ε_H do not follow the ones for decreasing ε_H .

In the previous comparison, the stretch in the first branch of the analytical flow [Eq. (21)] has been limited to match the one in the flow-focusing device. In such conditions, the maximum fractional length reached in the flow-focusing device is nearly equal to that obtained in the analytical flow (Fig. 11). However, it is also relevant to compare these values against the steady extension attained for an *infinite* strain in the analytical flow [first branch of Eq. (21) with large t_S], which would represent an *infinite* VR in the flow-focusing device. Such analysis is presented in Fig. 12 as a function of Wi . For each VR of the flow-focusing device, two curves are plotted. The curves denoted as “no shear” were obtained from simulations where the molecules start their trajectories with the configuration resulting from equilibrium in no-flow conditions, while the other curves are

FIG. 12. Maximum fractional molecular length as a function of the Weissenberg number. The curves denoted as “no shear” are for molecules pre-equilibrated in no-flow conditions. The curves are for the double 3D_AR1_14H flow-focusing device, except curve “Infinite strain”, which is obtained from an analytical planar extensional flow with unlimited strain (in practice, $\varepsilon_H = 30$ was sufficient to reach steady extension for all Wi tested). Lines are only a guide to the eye.

FIG. 13. Distribution of fractional molecular length measured at $x = -0.5H$, in the double 3D_AR1_14H flow-focusing device, for (a) $VR = 10$ and (b) $VR = 100$.

from equilibrium in the west arm channel flow, as described before. Although the curves are denoted as “no shear,” the molecules outside the centerline still experiment some degree of shear while they travel the short distance ($\sim 2H$) between their initial positions and the entrance to the converging region, especially the molecules close to the walls, where the shear-rate is higher. As expected, the molecular extension for any of the finite VR s remains below the extension achieved under *infinite* strain, although the difference seems to decrease for increasing Wi . In addition, we can observe that the pre-shearing in the west arm has a small influence for $VR = 100$ but plays an important role for $VR = 10$ due to the higher average strain-rate in the west arm.

Besides reaching a high molecular extension, some applications also require a narrow distribution of molecular lengths (e.g., Chan *et al.*⁶). The dispersion in the molecular length is mainly due to molecular individualism—molecules can present kinks, coils, knots, or other structures, which do not allow them to fully unravel, even under strong flows. It is precisely due to molecular individualism, originated from the random Brownian force term in the simulations, that the numerical

results need to be averaged over a large number of molecules (molecules with the same strain history do not necessarily present the same extension). Therefore, we computed such distribution of molecular lengths at the exit of the stretching region, $x = -0.5H$, and the results are shown in Fig. 13. We can observe that the dispersion in the molecular length is significant for $Wi \leq 2$, and especially around $Wi \approx 1$, which represents the region of flow strengths at which the molecules start to unfold. In general, for $Wi \geq 5$, the distribution of molecular lengths is narrow, and the peak frequency shifts toward higher fractional lengths for increasing Wi .

V. CONCLUSIONS

This work explored the shape optimization of flow-focusing devices for homogeneous extensional flow. The main goal of the optimization routines was to generate a delimited region of constant strain-rate along the centerline of flow-focusing devices with different channel aspect ratios and lengths. The optimizations were carried out based on an automatized cycle that involved geometry and mesh generation, fluid flow solution with a finite-volume solver, and estimation of new geometric parameters using an optimizer. The optimizations were performed for a fixed velocity ratio between streams, low Reynolds number, and assuming Newtonian fluid flow. Additionally, the performance of the resulting optimal shapes was assessed for varying conditions, showing a satisfactory behavior, in general, for $VR \gtrsim 10$, $Re \lesssim 1$, and weakly shear-thinning/shear-thickening or weakly elastic fluids. The devices with a longer extensional flow region display smaller oscillations in the plateau region of the strain-rate profiles at the centerline.

Anticipating the potential advantages from using the optimized devices in single-molecule studies, we also analyzed numerically the extension of λ -DNA molecules in one of the optimized flow-focusing devices. The analysis was based on Brownian dynamics simulations, using a bead-spring model to represent the biomolecules. Overall, it was shown that the extensional flow in the selected optimized device performs closely to an analytical planar extensional flow with limited strain and outperforms a non-optimized (standard) device. Still, the numerical results obtained call for future experimental validation.

The optimized devices obtained can be advantageous in generic microfluidic applications profiting from the conjugation of homogeneous extensional flows, high throughputs, focusing of streams, and easy/precise control of the Hencky strain and strain-rate. In particular, we envisage possible applications in extensional rheometry,²⁷ in the controlled deformation of biological cells^{12,49,50} or other deformable entities,^{13,14} and in the study of the behavior of complex fluids under controlled flow conditions.⁴⁸

A secondary result of this work was the development and testing of a generic solver for Brownian dynamics simulations, which is made available as open-source in *rheoTool*.⁴¹

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