Modelling ternary fluids in contact with elastic membranes

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We present a thermodynamically consistent model of a ternary fluid interacting with elastic membranes. Following a free-energy modelling approach for the fluid phases, we derive the governing equations for the dynamics of the ternary fluid flow and membranes. We also provide the numerical framework for simulating such fluid-structure interaction problems. It is based on the lattice Boltzmann method for the ternary fluid (Eulerian description) and a finite difference representation of the membrane (Lagrangian description). The ternary fluid and membrane solvers are coupled through the immersed boundary method. For validation purposes, we consider the relaxation dynamics of a two-dimensional elastic capsule placed at a fluid-fluid interface. The capsule shapes, resulting from the balance of surface tension and elastic forces, are compared with equilibrium numerical solutions obtained by Surface Evolver. Furthermore, the Galilean invariance of the proposed model is proven. The proposed approach is versatile, allowing for the simulation of a wide range of geometries. To demonstrate this, we address the problem of a capillary bridge formed between two deformable capsules.

I. INTRODUCTION

Multi-phase/component flows in interaction with deformable, thin structures are encountered in a broad range of applications. One example of such flows are capsules accommodating multiple aqueous solutions, such as polyethylene glycol and dextran, serving as model biological cells [1, 2]. Artificially fabricated elastic capsules [3] are also employed as container and delivery systems in many industrial applications, for instance in drug-delivery [4] and various processes in the cosmetic [5] and food [6] industries. Another case where a multiphase flow interacting with soft particles occurs is in the self-assembly of colloidal aggregates into patchy particles, a phenomenon of particular importance for the successful design of bottom-up materials [7, 8]. These patchy particles can be formed of compartments, each containing different polymers, bonded together by a solvent. The wetting of liquid droplets surrounded by air/vapour on soft solids is another example of multiphase flows in contact with deformable structures, with relevance in biology, e.g. cell locomotion [9], medicine and engineering, e.g. in the identification of cancer cells [10], and in the development of smart coatings [11]. Similar elastowetting problems also arise with fibrous materials, such as the coalescence of wet hair [12].

Even though these elastocapillary problems have been extensively studied both analytically and experimentally [13–26], the accurate computational modelling of such configurations is lagging behind. Elastic liquid-core capsules, obeying various constitutive laws, immersed in another fluid component constitute the most widely studied computational configuration [27–34]. A comprehensive review on such configuration is given by Barthès-Biesel [35]. All these computational works have not dealt with the contact line problem that arises when two or more fluids are in contact with the same side of a membrane. With respect to elastocapillary problems involving contact lines, there has been an attempt by Lubbers et al. [36] to numerically solve the equilibrium shape problem of liquid droplets on soft solids. The equilibrium shapes are obtained by minimizing the total elastocapillary energy of the vapour-liquid-substrate system; thus, no fluid dynamics properties of the droplets and surrounding air are available. Only recently methods capable of capturing contact line dynamics have started to be developed. For instance, Bueno et al. [37] developed a numerical framework for the simulation of binary fluids in contact with nonlinear hyperelastic solids to investigate the wetting of soft substrates and elastic micropillars. Wouters et al. [38] developed a numerical framework allowing for the simulation of soft particles at fluid-fluid interfaces.

The aim of the present work is to analytically derive a thermodynamically consistent model of multicomponent fluids in interaction with elastic membranes, and to provide a versatile numerical framework for the simulation of such fluid-structure interaction problems. We focus here on the case of a ternary fluid where one component is enclosed inside the membranes. The model can, however, be generalised to consider more fluid components contained in/surrounding the membranes. The availability of such computational method will allow us to systematically study a wide range of elastocapillary phenomena intractable to analytical solutions, in order to complement expanding experimental activities in this area.

We follow a top-down modelling approach for the ternary fluid using the lattice Boltzmann method [39–

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II. MATHEMATICAL MODELLING OF TERNARY FLUIDS IN INTERACTION WITH ELASTIC MEMBRANES

In this section, we first present briefly the free energy of the ternary fluid, which is based on the one proposed by Semprebon et al. [45] but modified accordingly in order to account for the interplay between the fluid and the membrane. We subsequently formulate the strain and bending energies of the elastic membrane. Finally, the equations of motion for the ternary fluid interacting with elastic membranes are introduced.

To clarify notations, bold lower case letters are employed for vector variables evaluated on the Eulerian lattices, while bold upper case ones refer to vector variables defined at the Lagrangian markers.

A. Free energy of the ternary fluid

In this article, we are concerned with cases in which there are three fluid components, and without any loss of generality, we will assume that the fluid component 3 is enclosed inside the membrane. The free energy $F_i$ of the ternary fluid system considered here is the sum of two contributions: a Landau free-energy functional $\tilde{E}_f$ allowing the coexistence of three fluid components, and a coupling-energy functional $\tilde{E}_c$ taking into account the interaction between the membrane and its confined fluid component

$$F_i = \tilde{E}_f + \tilde{E}_c.$$

In this work, we use the following forms for $\tilde{E}_f$ and $\tilde{E}_c$:

$$\tilde{E}_f = \int_V [f_{b,f} + f_{\nabla}] \, dV = \sum_{m=1}^{N=3} \int_V \left( c_s^2 \rho \ln \rho + \frac{\kappa_m}{2} C_m^2 (1 - C_m)^2 \right) dV,$$

$$\tilde{E}_c = \int_V f_{b,c} \, dV = \int_V \frac{\kappa_c}{2} (C_3 - \tilde{I})^2 \, dV,$$

where $C_m$ represents the density of the fluid component $m$ ($m = 1, 2, 3$), $\rho = \sum_m C_m$ is the total mass density, $c_s$ denotes the speed of sound, $\kappa_m$ are tunable parameters related to the fluid surface tensions, $\kappa_c$ denotes the coupling coefficient, and the integration takes place over the simulation volume $V$. The free energy density of the bulk fluid mixture $f_{b,f}$ is composed of the first two terms on the right-hand side of Eq. (2), while the interfacial free energy density $f_{\nabla}$ comprises the last term. The same form for $\tilde{E}_f$ is considered in the ternary fluid model proposed by Semprebon et al. [45], whereas the aforementioned model differs from the present one in that no coupling-energy functional $f_{b,c}$, Eq. (3), is taken into consideration. $\tilde{I}$ is an interfacial profile that we construct from the membrane, as described below in Eq. (7).

Following the form of the bulk free energy density $f_{b,f}$, each fluid density $C_m$ has two bulk minima at $C_m = \{0, 1\}$, where we drop the physical units in the following for brevity. For the ternary fluid system of interest, only the following three minimizers are relevant:

$C_1 = 1, \ C_2 = 0, \ C_3 = 0$;

$C_1 = 0, \ C_2 = 1, \ C_3 = 0$;

$C_1 = 0, \ C_2 = 0, \ C_3 = 1$. 

46]: namely, the free energy of the fluid system is initially formulated, including the desired thermodynamics features. Given the free energy, the macroscopic equations of motion of the multicomponent fluid can be derived. This technique is the contrary of the bottom-up modelling approach, where the macroscopic properties of the fluid arise from the microscopic interactions between the fluid particles, with these interactions usually taking the form of an interparticle potential [47, 48]. Krüger et al. [49] have discussed in detail the advantages and limitations of each of these modelling approaches. The lattice Boltzmann model proposed by Semprebon et al. [45] has been in particular chosen here for resolving the flow in a uniform, Cartesian grid. This is a diffuse interface model, meaning that the fluid-fluid interfaces are spread over several lattices, rather than being tracked explicitly. The capsules enclosing one fluid component are modelled here as infinitely thin membranes, composed of a homogeneous and isotropic material, able to undergo stretching/compression and bending. To resolve the interaction between the ternary fluid and the membranes, we adopt the immersed boundary method (IBM), initially proposed by Peskin [50]. This allows for solving the equations of motion of the ternary fluid in an Eulerian description, while the membranes’ ones are solved in a Lagrangian coordinate system, following the motion/deformation of the membrane’s boundary.

The article is organised as follows. In Sec. II, we present a thermodynamically consistent model of a ternary fluid in contact with elastic membranes. The methods employed here for the numerical solution of the ternary fluid and its interacting membranes equations of motion are subsequently presented in Sec. III A – Sec. III C. The Surface Evolver [51], an open-source software used for benchmarking purposes, is briefly discussed in Sec. III D. We then validate our model against Surface Evolver in Sec. IV A, considering a two-dimensional elastic capsule placed at a fluid-fluid interface as the benchmark configuration. We also prove the Galilean invariance of the equations governing the ternary fluid flow and the membranes’ dynamics. To demonstrate the versatility of our model, the problem of a capillary bridge taken as a problem of motion are subsequently presented in Sec. III A – Sec. III C. The Surface Evolver [51], an open-source software used for benchmarking purposes, is briefly discussed in Sec. III D. We then validate our model against Surface Evolver in Sec. IV A, considering a two-dimensional elastic capsule placed at a fluid-fluid interface as the benchmark configuration. We also prove the Galilean invariance of the equations governing the ternary fluid flow and the membranes’ dynamics. To demonstrate the versatility of our model, the problem of a capillary bridge formed between two elastic capsules is afterwards studied in Sec. IV B. For simplicity, here we focus on two dimensions. Finally, the key contributions of the present work and its future perspectives are summarized in Sec. V.
corresponding to the three bulk fluids in the ternary system. Our multicomponent fluid model is a diffuse interface model; when the fluid transitions from one bulk component to another, the interfacial profile assumes the form

$$C_m = \frac{1 + \tanh \left( \frac{d_{FI}}{2\alpha} \right)}{2},$$

(5)

where $d_{FI}$ measures the distance between the bulk fluid at position $\mathbf{x}$ and the fluid-fluid interface. Equation (5) ensures that $C_m \to 1$ for $d_{FI} \to \infty$, and $C_m \to 0$ for $d_{FI} \to -\infty$. The parameter $\alpha$ is proportional to the interface width, which here is chosen the same for all three fluid-fluid interfaces. The fluid-fluid surface tensions are expressed as

$$\gamma_{mn} = \frac{\alpha}{6} (\kappa_m + \kappa_n), \ m, n = 1, 2, 3 \text{ and } m \neq n.$$  

(6)

The coupling energy is formulated in such a way that for $\kappa_c > 0$ there is a minimum when $C_3 = I$. The interfacial profile $I$ across the elastic membrane is then defined as

$$I = \frac{1 + \tanh \left( d(\mathbf{x}, \mathbf{X}) / (2\alpha) \right)}{2},$$

(7)

where $d(\mathbf{x}, \mathbf{X})$ denotes the distance between the fluid at position $\mathbf{x}$ and the membrane located at $\mathbf{X}$, as shown in figure 1(a). This distance is assigned to be positive for the enclosed fluid component 3, and negative for the surrounding fluid components 1 and 2. The width of the fluid-membrane interfaces is kept the same as the fluid-fluid interfaces one by assigning the same value to the parameter $\alpha$ in Eqs. (5) and (7). Similarly to $C_m$, $I \to 1$ for $d(\mathbf{x}, \mathbf{X}) \to \infty$, and $I \to 0$ for $d(\mathbf{x}, \mathbf{X}) \to -\infty$, as depicted in figure 1(b). It should be noted that changes in the free energy functional will reflect on the definition of $I$. It is obvious that with an increasing coupling coefficient $\kappa_c$, the interfacial profile for the density $C_3$ of the enclosed fluid component will be superimposed onto the profile of the fluid 3-membrane interface. However, if $\kappa_c$ is too high, the coupling energy term will dominate over the rest of the free energy terms, which is undesirable and may even lead to numerical instabilities.

### B. Strain and bending energies of the elastic membrane

The formulations of $\mathcal{E}_s$ and $\mathcal{E}_b$ depend on the nature of the membrane material. Here, we consider the simple case of a linear elastic material, for which the strain energy takes the form of Hooke’s law, and the bending energy is given by the squared mean curvature. However, the strain and bending energy formulations can be modified to model more realistic materials. Thus, the strain and bending energies are expressed as:

$$\mathcal{E}_s = \int_S \frac{\kappa_s}{2} \left( \left| \frac{\partial \mathbf{X}}{\partial s} \right| - 1 \right)^2 \, ds,$$

(8)

and

$$\mathcal{E}_b = \int_S \frac{\kappa_b}{2} \left( \frac{\partial \theta}{\partial s'} \right)^2 \, ds.$$

(9)

The integration takes place over the surface $S$ of the elastic membrane, whose position in Eulerian coordinates at time $t$ is described by $\mathbf{X} = \mathbf{X}(s, t)$, with $s$ denoting its undeformed Lagrangian coordinates. The parameters $\kappa_s$ and $\kappa_b$ are, respectively, the stretching and bending moduli. $\partial \theta / \partial s'$ denotes the membrane curvature, which depends on how the tangential angle, $\theta = \theta(\mathbf{X})$, varies across the deformed membrane surface, described by $s' = s'(\mathbf{X})$.

### C. Equations of motion

Following the rationale of Kou and Sun [52], we derive the equations of motion for the fluids and the elastic membrane, making use of the first and second laws of
thermodynamics. The complete derivation is provided in the Appendix. For convenience, we will also introduce the total mass density $\rho$, and two auxiliary fields $\phi$ and $\psi$, given by

$$\rho = C_1 + C_2 + C_3, \quad \phi = C_1 - C_2, \quad \psi = C_3.$$  

(10)

Here we assume that all fluid components have the same density, and thus the total mass density is set to be $\rho = 1$.

Using $\rho$, $\phi$ and $\psi$, the equations of motion for the fluid components are given by the continuity, Navier-Stokes and two Cahn-Hilliard equations

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{u}) = 0,$$  

(11)

$$\frac{\partial (\rho \mathbf{u})}{\partial t} + \nabla \cdot (\rho \mathbf{u} \otimes \mathbf{u}) = -\nabla \rho_i + \nabla \cdot \left[ \eta \left( \nabla \mathbf{u} + \nabla \mathbf{u}^T \right) \right] - \rho \nabla \mu_{\rho} - \phi \nabla \mu_{\phi} - \psi \nabla \mu_{\psi} - \psi \nabla \left( \frac{\delta f_{b,c}}{\delta \psi} \right),$$  

(12)

$$\frac{\partial \phi}{\partial t} + \nabla \cdot (\phi \mathbf{u}) = M_{\phi} \nabla^2 \mu_{\phi},$$  

(13)

$$\frac{\partial \psi}{\partial t} + \nabla \cdot (\psi \mathbf{u}) = M_{\psi} \nabla^2 \mu'_{\psi} = M_{\psi} \nabla^2 \left( \mu_{\psi} + \frac{\delta f_{b,c}}{\delta \psi} \right),$$  

(14)

where $\mathbf{u}$, $\rho_i = \rho \rho_i^2$, $\eta$, $M_{\phi}$ and $M_{\psi}$ are, respectively, the mass-averaged velocity, the ideal gas pressure, the fluid dynamic viscosity, and the two mobility parameters in the Cahn-Hilliard equations. We assume that all fluid components $C_m$ ($m = 1, 2, 3$) have identical mobility parameters, resulting in $M_{\phi} = 3 M_{\psi}$ when the auxiliary variables are employed. $\mu_{\rho}$, $\mu_{\phi}$ and $\mu_{\psi}$ are the chemical potentials coming from the chosen free energy density of the ternary fluid. The detailed formulations of the chemical potentials $\mu_{\rho}$, $\mu_{\phi}$ and $\mu_{\psi}$ in terms of the variables $\rho$, $\phi$ and $\psi$ are given by

$$\mu_{\rho} = \frac{\kappa_1}{8} (\rho + \phi - \psi) (\rho + \phi - \psi - 1) (\rho + \phi - \psi - 2) + \frac{\kappa_2}{8} (\rho + \phi - \psi) (\rho - \phi - \psi - 1) (\rho - \phi - \psi - 2) + \frac{\alpha^2}{4} \left[ (\kappa_1 + \kappa_2) \nabla^2 \psi - \nabla^2 \rho + (\kappa_2 - \kappa_1) \nabla^2 \phi \right],$$  

(15)

$$\mu_{\phi} = \frac{\kappa_1}{8} (\rho + \phi - \psi) (\rho + \phi - \psi - 1) (\rho + \phi - \psi - 2) - \frac{\kappa_2}{8} (\rho - \phi - \psi) (\rho - \phi - \psi - 1) (\rho - \phi - \psi - 2) + \frac{\alpha^2}{4} \left[ (\kappa_2 - \kappa_1) \nabla^2 \rho - (\kappa_1 + \kappa_2) \nabla^2 \phi \right],$$  

(16)

$$\mu_{\psi} = -\frac{\kappa_1}{8} (\rho + \phi - \psi) (\rho + \phi - \psi - 1) (\rho + \phi - \psi - 2) - \frac{\kappa_2}{8} (\rho - \phi - \psi) (\rho - \phi - \psi - 1) (\rho - \phi - \psi - 2) + \frac{\alpha^2}{4} \left[ (\kappa_1 + \kappa_2) \nabla^2 \rho - (\kappa_2 - \kappa_1) \nabla^2 \phi \right] - (\kappa_1 + \kappa_2 + 4 \kappa_3) \nabla^2 \psi + \kappa_3 \psi (\psi - 1) (2 \psi - 1).$$  

(17)

It readily follows from Eq. (3) that $\delta f_{b,c}/\delta \psi = \kappa_c (\psi - 1)$.

To be consistent with the work of Semprebon et al. [45], the ideal gas pressure, $p_i$, is written as a separate term in the Navier-Stokes Eq. (A27) instead of being included into the definition of $\mu_{\rho}$, Eq. (15). Since the total mass density is set to $\rho = 1$ everywhere in this work, the chemical potential term $\mu_{\rho}$ is essentially a constant and it does not play any significant role in the Navier-Stokes equation. Compared to previous ternary free energy lattice Boltzmann models, such as by Semprebon et al. [45], the present model is differentiated by the last term on the right-hand side of Eq. (A27) and the additional term $\delta f_{b,c}/\delta \psi$ in the chemical potential $\mu'_{\psi}$, Eq. (14). Both terms are due to the coupling-energy functional between the fluid and the membrane. As we will discuss in Sec. III, another key difference to previously published models is the use of immersed boundary method to couple the fluid and membrane dynamics.

The membrane is discretised by a collection of Lagrangian markers. The variable $X_i$ denotes the position of the $i$th Lagrangian marker (in Eulerian coordinates), with $l = 1, \ldots, N_l$. The equation of motion for each Lagrangian marker is given by

$$m \frac{d^2 X_i}{d t^2} = F_{\text{mem},l} = F_{s,l} + F_{b,l} + F_{c,l},$$  

(18)

where $F_{\text{mem}}$ denotes the total force exerted on the elastic membrane, and

$$F_{s,l} = -\frac{\partial E_s}{\partial X_i}, \quad F_{b,l} = -\frac{\partial E_b}{\partial X_i}, \quad F_{c,l} = -\frac{\partial E_c}{\partial X_i},$$  

(19)

are the strain, bending and coupling forces, respectively. The detailed forms of $F_{s,l}$, $F_{b,l}$ and $F_{c,l}$ are provided in Sec. III B.

III. NUMERICAL METHODS

In this section, the numerical techniques employed for the solution of the equations of motion of the ternary fluid and its interacting elastic membrane are discussed. The governing equations of the ternary fluid are solved numerically by means of a lattice Boltzmann method, as detailed in Sec. III A. The discretized forms of the strain, bending and coupling energies are presented in Sec. III B along with the corresponding force formulations. The interaction between the ternary fluid and elastic membrane is solved by an immersed boundary method, described in Sec. III C, that is coupled to the lattice Boltzmann method following the algorithm presented in Krüger et al. [53]. Finally, we briefly report the equivalent energy implementation in Surface Evolver, a finite element approach employed to benchmark the equilibrium solutions of wetting elastic membranes, in Sec. III D.
A. Lattice Boltzmann method

To solve the equations of motion of the ternary fluid, Eqs. (A26) and (A27)–(14), we employ the lattice Boltzmann method with three sets of distribution functions \( f_i (x, t), \ g_i (x, t), \) and \( h_i (x, t), \) corresponding to the total fluid density \( \rho \) and the auxiliary fields \( \phi \) and \( \psi. \) The evolutions of the distribution functions are governed by the lattice Boltzmann equation, where the standard Bhatnagar-Gross-Krook (BGK) single relaxation time model \([54]\) is used for the collision operator, and the exact difference scheme \([55]\) is employed for the forcing term

\[
f_i (x + c_i \Delta t, t + \Delta t) = f_i (x, t) - \frac{\Delta t}{\tau} [f_i (x, t) - f_i^{eq} (\rho, u)] + \Delta t [f_i^{eq} (\rho, u + \delta u) - f_i^{eq} (\rho, u)],
\]

\[
g_i (x + c_i \Delta t, t + \Delta t) = g_i (x, t) - \frac{\Delta t}{\tau} [g_i (x, t) - g_i^{eq} (\phi, v)],
\]

\[
h_i (x + c_i \Delta t, t + \Delta t) = h_i (x, t) - \frac{\Delta t}{\tau} [h_i (x, t) - h_i^{eq} (\psi, v)].
\]

The variables \( f_i (x, t), \ g_i (x, t), \) and \( h_i (x, t) \) refer to the distribution functions \( f_i, g_i, \) and \( h_i \) at position \( x \) and time \( t \) with velocity \( c_i \) along the \( i \)th lattice direction. The relaxation times \( \tau, \tau_{\phi}, \) and \( \tau_{\psi} \) are linked to the dynamic viscosity \( \eta \) and the mobility parameters \( M_\phi \) and \( M_\psi \) by

\[
\eta = \rho c_s^2 \left( \frac{\tau - \Delta t}{2} \right),
\]

\[
M_\phi = \Gamma_\phi \left( \frac{\tau_\phi - \Delta t}{2} \right),
\]

\[
M_\psi = \Gamma_\psi \left( \frac{\tau_\psi - \Delta t}{2} \right),
\]

where \( c_s \) is the speed of sound, and \( \Gamma_\phi, \Gamma_\psi \) are tunable parameters. The speed of sound is given by \( c_s = \frac{1}{\sqrt{\rho}} \), where \( c = \frac{\Delta x}{\Delta t} \) is the lattice speed, and \( \Delta x, \Delta t \) are the lattice spacing and time step, respectively. The variables \( f_i^{eq}, g_i^{eq}, \) and \( h_i^{eq} \) denote the equilibrium distribution functions.

The equilibrium distribution functions are expressed as

\[
f_i^{eq} (\rho, u) = w_i \rho \left[ 1 + \frac{c_i \cdot u}{c_s^2} + \frac{uu : (c_i c_i - c_s^2 I)}{2c_s^4} \right],
\]

\[
g_i^{eq} (\phi, v) = \begin{cases} w_i \left[ \frac{\Gamma_\phi \phi}{c_s^2} + \frac{\phi c_i \cdot v}{c_s^2} + \frac{\phi \nabla \cdot (v c_i c_i - c_s^2 I)}{2c_s^4} \right], & i \neq 0 \\ \phi - \sum_{i,i \neq 0} g_i^{eq}, & i = 0, \end{cases}
\]

\[
h_i^{eq} (\psi, v) = \begin{cases} w_i \left[ \frac{\Gamma_\psi \psi}{c_s^2} + \frac{\psi c_i \cdot v}{c_s^2} + \frac{\psi \nabla \cdot (v c_i c_i - c_s^2 I)}{2c_s^4} \right], & i \neq 0 \\ \psi - \sum_{i,i \neq 0} h_i^{eq}, & i = 0, \end{cases}
\]

where \( w_i \) are weight coefficients depending on the chosen lattice arrangement for the velocity discretisation, and \( I \) is the identity tensor.

The macroscopic physical variables are defined as moments of the distribution functions

\[
\rho (x, t) = \sum_i f_i (x, t),
\]

\[
u (x, t) = \left( \sum_i c_i f_i (x, t) \right) / (\rho (x, t)),
\]

\[
\phi (x, t) = \sum_i g_i (x, t),
\]

\[
\psi (x, t) = \sum_i h_i (x, t).
\]

The densities \( C_m \) can then be reconstructed by using the inverse variables transformation of Eq. (10)

\[
C_1 = (\rho + \phi - \psi) / 2, \ C_2 = (\rho - \phi - \psi) / 2, \ C_3 = \psi.
\]

The variable \( \nu \) represents the bare fluid velocity, and it is related to the actual fluid velocity \( \bar{v} \) by

\[
\nu (x, t) = \bar{v} (x, t) + \delta \nu (x, t) / 2,
\]

where \( \delta \nu \) denotes the velocity correction given by

\[
\delta \nu (x, t) = f (x, t) / \rho (x, t) \Delta t.
\]

The forcing term \( f \) can be considered as the sum of three contributions: a force \( f_{FE} \) taking into account the gradient terms on the right-hand side of Eq. (A27), a force \( f_{IB} \) accounting for the interaction between the ternary fluid and elastic membrane, and a force \( f_{ext} \) allowing the existence of external forces

\[
f = f_{FE} + f_{IB} + f_{ext}.
\]

The force \( f_{FE} \) can be written as

\[
f_{FE} = -\rho \nabla \mu_\rho - \phi \nabla \mu_\phi - \psi \nabla \mu_\psi - \psi \nabla \left( \frac{\delta f_{h,c}}{\delta \psi} \right).
\]

The form of the force \( f_{IB} \) is discussed in Sec. III C. In the present work, no external forces are considered, that is \( f_{ext} = 0. \)

In summary, the present lattice Boltzmann model differs from the one proposed by Semperon et al. \([45]\) for ternary fluids in the following ways: (1) the added term \( \delta f_{h,c}/\delta \psi \) in the chemical potential \( \mu_\psi \), (2) the added term \( -\psi \nabla \left( \delta f_{h,c}/\delta \psi \right) \) in Eq. (28), and (3) the inclusion of the immersed boundary forces \( f_{IB} \) in Eq. (27).

In lattice units, the lattice spacing and time step are for simplicity set equal to 1, \( \Delta x = \Delta t = 1, \) resulting in \( c = 1 \) and \( c_s = 1/\sqrt{3}. \) Here, we employ the D2Q9 lattice arrangement, for which the lattice velocities (in columns) are defined as

\[
c_i = \begin{bmatrix} 1 & -1 & 0 & 0 & 1 & -1 & -1 & 1 \end{bmatrix},
\]

and the weight coefficients are given by

\[
w_0 = 4/9, \ w_{1-4} = 1/9, \ w_{5-8} = 1/36.
\]
The discretised strain and bending energies of the elastic membrane, given respectively by Eqs. (8) and (9) in its continuous-space forms, are formulated as

\[ E_s = \frac{\kappa_s}{2} \sum_{l=1}^{N_l} \left( \frac{X_{l+1} - X_l}{\Delta s} - 1 \right)^2 \Delta s, \quad (29) \]

\[ E_b = \frac{\kappa_b}{2} \sum_{l=1}^{N_l} \left( \frac{\Delta \theta}{\Delta s'} \right)^2 \Delta s. \quad (30) \]

The summation occurs over all the Lagrangian markers \( l = 1, \ldots, N_l \). The variable \( X_l \) denotes the position of the \( l \)th Lagrangian marker (in Eulerian coordinates), and \( \Delta s \) represents the initial distance between two consecutive Lagrangian markers. The membrane is initially discretized into Lagrangian markers such that \( \Delta s = \Delta x = 1 \). The tangential angle \( \Delta \theta \) and arc length \( \Delta s' \) of the circumscribed circle passing through the Lagrangian marker \( l \) and its neighboring points, depicted in figure 2, can be expressed as

\[ \Delta \theta (l) = 2 \arccos \left[ \frac{(X_{l+1} - X_l) \cdot (X_l - X_{l-1})}{|X_{l+1} - X_l||X_l - X_{l-1}|} \right], \quad (31) \]

\[ \Delta s' (l) = |X_l - X_{l-1}| + |X_{l+1} - X_l|. \quad (32) \]

Taking into account Eqs. (29)–(32), the corresponding discretised strain and bending forces can then be found as

\[ F_{s,l} = -\frac{\partial E_s}{\partial X_l} = -\sum_{m=1}^{N_l} \kappa_s \left( \frac{|X_{m+1} - X_m|}{|X_{m+1} - X_m|} - 1 \right) \frac{X_{m+1} - X_m}{|X_{m+1} - X_m|} (\delta_{m+1,l} - \delta_{m,l}) \quad (33) \]

and

\[ F_{b,l} = -\frac{\partial E_b}{\partial X_l} = -\sum_{m=1}^{N_l} \kappa_b \left( \frac{2 \Delta \theta (m)}{\Delta s' (m)} \frac{\partial \Delta \theta (m)}{\partial X_l} - \left( \frac{\Delta \theta (m)}{\Delta s' (m)} \right)^2 \frac{\partial \Delta s' (m)}{\partial X_l} \right), \quad (34) \]

where

\[ \frac{\partial \Delta \theta (m)}{\partial X_l} = -\frac{2}{\sqrt{1 - y^2 (m)}} \left[ \left( \frac{X_m - X_{m-1}}{|X_{m+1} - X_m||X_m - X_{m-1}|} - \frac{X_{m+1} - X_m}{|X_{m+1} - X_m|} \cdot \frac{X_m - X_{m-1}}{|X_{m+1} - X_m||X_m - X_{m-1}|} \right) (\delta_{m+1,l} - \delta_{m,l}) \right. \\

\[ + \left. \left( \frac{X_{m+1} - X_m}{|X_{m+1} - X_m||X_m - X_{m-1}|} - \frac{X_{m+1} - X_m}{|X_{m+1} - X_m|} \cdot \frac{X_m - X_{m-1}}{|X_{m+1} - X_m||X_m - X_{m-1}|} \right) (\delta_{m,l} - \delta_{m-1,l}) \right], \]

\[ \frac{\partial \Delta s' (m)}{\partial X_l} = \frac{X_m - X_{m-1}}{|X_m - X_{m-1}|} (\delta_{m,l} - \delta_{m-1,l}) + \frac{X_{m+1} - X_m}{|X_{m+1} - X_m|} (\delta_{m+1,l} - \delta_{m,l}). \]

The variables \( F_{s,l} \) and \( F_{b,l} \) denote the discretised strain and bending forces exerted on the \( l \)th Lagrangian marker.

The discretised coupling energy of the elastic membrane can be written as

\[ E_c = \frac{\kappa_c}{2} \sum_{x} (\psi - I)^2 (\Delta x)^d, \quad (35) \]
where \( \sum_x \) implies summation over all the Eulerian lattice nodes \( x \), and \( d \) is the domain dimensionality (\( d = 2 \) in the present case). The interfacial profile \( I \) is given by Eq. (7). The corresponding discretised coupling force can then be obtained as

\[
F_{c,t} = - \frac{\partial \mathcal{E}_c}{\partial X_l} = - \sum_x \frac{\delta f_{h,c}}{\delta t} \frac{\partial I}{\partial X_l}
\]

\[
= - \sum_x \frac{K_c}{4\alpha} (\psi - I) \text{sech}^2 \left( \frac{d(x, X_l)}{2\alpha} \right) \frac{\partial d(x, X_l)}{\partial X_l}.
\]

(36)

The variable \( F_{c,t} \) represents the discretised coupling force exerted on the \( l \)th Lagrangian marker.

In the computational implementation, we exert the forces \( F_s, I, F_h, l \) and \( F_{c,t} \) on each Lagrangian marker \( l \).

C. Immersed boundary method

To reproduce the effect of \( F_{\text{mem},l} \) on the Eulerian fluid flow, denoted here by \( f_{\text{IB}} \), a spreading operation is used

\[
f_{\text{IB}} (x, t) = S[F_{\text{mem},l}] (x) = \sum_{l=1}^{N_l} F_{\text{mem},l} \delta_h (x - X_l) \Delta s.
\]

(37)

The term \( \delta_h \) denotes the discretised Dirac delta function, and the following formulation proposed by Peskin [56] is chosen here to perform the convolution in the spreading and interpolation operations

\[
\delta_h (r) = \begin{cases} 
\frac{1}{8} \left( 3 - 2|r| + \sqrt{1 + 4|r| - 4r^2} \right), & |r| \leq 1 \\
\frac{1}{8} \left( 5 - 2|r| - \sqrt{-7 + 12|r| - 4r^2} \right), & 1 \leq |r| \leq 2 \\
0, & 2 \leq |r|.
\end{cases}
\]

If the variable \( r \) is a vector, \( r = (r_x, r_y) \), then the two-dimensional \( \delta_h \) is given by \( \delta_h (r) = \frac{1}{4\pi} \delta_h (r_x/\Delta x) \delta_h (r_y/\Delta y) \). Once the force \( f_{\text{IB}} \) is computed, the total density \( \rho \), auxiliary fields \( \phi \) and \( \psi \), and velocity \( v \) fields can be obtained at the next time step \( t + \Delta t \) by solving Eq. (20). To calculate the forces \( F_{\text{mem},l} \) at \( t + \Delta t \), the position of the Lagrangian markers \( X_l \), \( l = 1, \ldots, N_l \) at \( t + \Delta t \) needs to be known. For this reason, the known \( v (x, t + \Delta t) \) is interpolated at the Lagrangian markers as

\[
U (X_l, t + \Delta t) = \mathcal{I} [v] (X_l) = \sum_x v \delta_h (x - X_l) (\Delta x)^d.
\]

(38)

The updated position of the Lagrangian markers can then be found by Euler’s rule

\[
X_l (t + \Delta t) = X_l (t) + U (X_l, t + \Delta t) \Delta t.
\]

(39)

For simplicity, here we have used an explicit immersed boundary method. This is sufficient for the applications considered in this work. The temporal lag between the ternary fluid and elastic membrane can be eliminated by considering either sub-iterations of the coupled algorithm or implicit immersed boundary methods [57, 58].

D. Surface Evolver

In the absence of closed-form solutions for non-trivial elastocapillary problems, we have benchmarked the proposed coupled lattice Boltzmann-immersed boundary method against a finite element approach, the Surface Evolver. Surface Evolver has been extensively used to model the equilibrium shapes of liquid interfaces and capillary forces [59–61], and model deformations of elastic membranes [62]. In Surface Evolver, the interfaces are discretized by triangulated meshes, and configurations in mechanical equilibrium correspond to minima of the total energy, obtained through a conjugate gradient descent method.

For our benchmarks, we model 2D elastic capsules placed at a fluid-fluid interface in mechanical equilibrium. To minimize numerical deviations, the elastic capsules are initialized with exactly the same geometry as in the proposed model, described in Sec. IV A. The same free energy of the elastic membrane is implemented using the provided scripting language to formulate the strain and bending energies as in Eqs. (29) and (30). In particular, the functions \textit{edge}\_\textit{length} and \textit{scurve}\_\textit{string}\_\textit{marked} are employed for the calculation of the strain and bending energies. The main difference is in the definition of the surface tensions: here, the corresponding energy is simply accounted for by adding a term proportional to the total length of each fluid or membrane interface, multiplied by a constant parameter matching the surface tension arising from the diffuse interface in the lattice Boltzmann method. No coupling energy has been considered, as both the elastic energies and surface tension are provided by the discrete representation of the capsule and the fluid-fluid interface. The constraint of conservation of the total capsule area has also been considered.

IV. RESULTS

The proposed fluid-structure solver is validated in Sec. IV A. The steady configuration of an elastic capsule positioned at a fluid-fluid interface is chosen for this purpose. We perform a thorough comparison with the reference results of Surface Evolver for different cases of surface tension ratios, and various combinations of stretching and bending moduli. We also establish the Galilean invariance of the governing equations of the ternary fluid and elastic structure. Finally, a more complex configuration is considered in Sec. IV B to demonstrate the capabilities of the proposed model.
FIG. 3. Comparison of the elastic capsule’s position (•) and contours of ψ = 0.5 (—) and \( I = 0.5 \) (· · ·) for: (a) \( \alpha = 1.0 \), (b) \( \alpha = 1.5 \), and (c) \( \alpha = 2.0 \) in the symmetric case. The shown results correspond to a capsule of initial radius \( R = 20 \), considering a coupling coefficient of \( \kappa_c = 10^{-2} \). The middle of the diffuse interface distinguishing fluids 1 and 2 is illustrated by dashed lines (- - -).

A. Elastic capsule at fluid-fluid interface

To benchmark the proposed model, the configuration of an elastic capsule placed at a fluid-fluid interface, as shown in figure 1(a), is studied. An initially circular capsule of radius \( R \) is located at the center of a computational domain of dimensions \( 12R \times 6R \). The capsule relaxes to a mechanical equilibrium shape, which depends on the balance of the elastic strain and bending forces and the surface tensions \( \gamma_{12}, \gamma_{13} \) and \( \gamma_{23} \). To quantify the capsule deformation, we employ the Taylor deformation \( D = (L - B) / (L + B) \), where \( L \) and \( B \) are, respectively, the major and minor axes of the final elliptical capsule shape. Periodic boundary conditions are considered at all the domain boundaries. Simulations are performed for several combinations of stretching and bending moduli, \( \kappa_s = \{ 10^{-3}, 10^{-2}, 10^{-1} \} \) and \( \kappa_b = \{ 10^{-4}, 10^{-2} \} \). We consider two cases: (1) \( \gamma_{13} = \gamma_{23} \), resulting in a capsule shape that is symmetrical along the domain central lines, and (2) \( \gamma_{13} \neq \gamma_{23} \), resulting in the capsule to be more immersed in one of the surrounding fluid phases. We will refer thereafter to case 1 as the symmetric case, and case 2 as the asymmetric one. In the symmetric case, the surface tension ratio \( \gamma_{12}/\gamma_{13} = \gamma_{12}/\gamma_{23} \) is considered, varying from 0.888 to 1.882, while the ratio \( \gamma_{13}/\gamma_{23} \) ranging from 1.60 to 3.40 is examined in the asymmetric case. Both the relaxation times and the parameters \( \Gamma_\phi, \Gamma_\psi \) are all considered to be equal to 1, \( \tau = \tau_\phi = \tau_\psi = 1 \) and \( \Gamma_\phi = \Gamma_\psi = 1 \).

We first investigate the effect of the free-energy parameter \( \alpha \) on the capsule dynamics and flow field in both the symmetric and asymmetric cases. Simulations are conducted here for the most deformable capsule, that is \( \kappa_s = 10^{-3} \), at a bending modulus \( \kappa_b = 10^{-2} \) and the highest surface tension ratio studied in each case, namely \( \gamma_{12}/\gamma_{13} = 1.882 \) and \( \gamma_{13}/\gamma_{23} = 3.40 \). The elastic capsule is initialized with radius \( R = 20 \). The coupling coefficient is kept constant at \( \kappa_c = 10^{-2} \). The comparison of the Taylor deformation \( D \) and the corresponding relative error \( \delta D = | D_{\text{Ref}} - D | / D_{\text{Ref}} \), where \( D_{\text{Ref}} \) is the reference solution obtained by Surface Evolver, at the various parameters \( \alpha \) is shown in table I. At \( \alpha = 1 \), the Taylor deformation is highly underpredicted, with the relative error being greater than 5% in both the symmetric and asymmetric cases. The relative difference falls below 5% in both cases only at \( \alpha = 2 \). It is worth noting that the magnitude of the spurious currents decreases from approximately \( 5 \cdot 10^{-5} \) to \( 2 \cdot 10^{-5} \) when increasing the parameter \( \alpha \) from 1 to 2. As seen from Eqs. (5) and (7), the parameter \( \alpha \) has an effect on the interfacial profile for the density of the fluid components as well as the one across the elastic capsule. To assess this effect, the capsule shape is plotted along with the contour lines of \( C_3 = \psi = 0.5 \) and \( I = 0.5 \) at the various values of the parameter \( \alpha \) for the symmetric case in figure 3. For the optimal values of \( \alpha \) and \( \kappa_c \), the elastic capsule should be positioned at the middle of the interfacial profile \( I \), which in turn should be superimposed onto the middle of the interfacial profile for \( C_3 \). As observed from figure 3, the elastic capsule is indeed located at \( I = 0.5 \) for all values of \( \alpha \). However, deviations between the contour lines of \( \psi = 0.5 \) and \( I = 0.5 \) can be noticed close to the three-phase contact point for the lower values of \( \alpha \). These contour lines agree well with each other only for \( \alpha = 2 \). This establishes our choice of \( \alpha = 2 \) in subse-

<table>
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<th>( \alpha )</th>
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<th>Asymmetric</th>
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<td>( \delta D (%) )</td>
<td>( D )</td>
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quaint simulations. Similar observations can be made in the asymmetric case (data not shown). It is also worth noting that the suitable choice for \( \alpha \) will be affected by the form of the discretized Dirac delta function in the immersed boundary approach.

To examine the effect of the coupling coefficient on the superposition of the interfacial profiles of \( \psi \) and \( \mathcal{I} \), we perform simulations varying the value of \( \kappa_c \) from \( 5 \cdot 10^{-3} \) to \( 5 \cdot 10^{-2} \). As before, capsules of initial radius \( R = 20 \) are considered. Figure 4 shows the capsule’s position in conjunction with the contour lines of \( \psi = 0.5 \) and \( \mathcal{I} = 0.5 \) for the symmetric case. Similar to before, the elastic capsule is placed at \( \mathcal{I} = 0.5 \) for all values of \( \kappa_c \). Significant discrepancies between the contour lines of \( \psi = 0.5 \) and \( \mathcal{I} = 0.5 \) can be observed not only close to the three-phase contact point, but at the entirety of these contour lines for \( \kappa_c = 5 \cdot 10^{-3} \). For \( \kappa_c = 10^{-2} \), the desired contour lines agree well with each other, as previously mentioned. This coupling coefficient value is the threshold for the contour lines to overlap. For the highest coupling coefficient value shown here, that is \( \kappa_c = 5 \cdot 10^{-2} \), the contour lines of \( \psi = 0.5 \) and \( \mathcal{I} = 0.5 \) are perfectly superposed. However, this value of \( \kappa_c \) was found to be close to the threshold before the domination of the coupling energy over the rest of the free energy terms occurs. To avoid numerical instabilities, the use of \( \kappa_c = 10^{-2} \) in following simulations was deemed appropriate. Similar trends have been observed in the asymmetric case (data not shown).

To isolate the effect of increasing capsule radius on the Taylor deformation and capsule shape, simulations are carried out varying the value of \( R \), while keeping constant the ratio of the elastocapillary length \( l_{EC} = \sqrt{\kappa_b/\gamma_{12}} \) to the capsule’s radius. Table II demonstrates the Taylor deformation and capsule shape obtained for \( \alpha = 2 \) and coupling coefficient \( \kappa_c = 10^{-2} \) are considered here.

![Graphs showing Taylor deformation and capsule shape](image)

**FIG. 4.** Comparison of the elastic capsule’s position (●) and contours of \( \psi = 0.5 \) (—) and \( \mathcal{I} = 0.5 \) (· · ·) for: (a) \( \kappa_c = 5 \cdot 10^{-3} \), (b) \( \kappa_c = 10^{-2} \), and (c) \( \kappa_c = 5 \cdot 10^{-2} \) in the symmetric case. The presented results correspond to a capsule of initial radius \( R = 20 \), with the parameter \( \alpha \) being equal to 2.0. The dashed lines (· - ·) denote the middle of the diffuse interface separating fluids 1 and 2.

<table>
<thead>
<tr>
<th>( R )</th>
<th>( D )</th>
<th>( \delta D ) (%)</th>
<th>( L/R )</th>
<th>( B/R )</th>
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<th>( L/R )</th>
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<tr>
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<tr>
<td>Ref. 0.534</td>
<td>-</td>
<td>3.77</td>
<td>1.14</td>
<td>0.414</td>
<td>-</td>
<td>3.16</td>
<td>1.31</td>
</tr>
</tbody>
</table>

**TABLE II.** Taylor deformation \( D \), relative error \( \delta D \), and normalized major \( L \) and minor \( B \) axes at different capsule radii \( R \) for the symmetric and asymmetric cases. \( \alpha = 2 \) and coupling coefficient \( \kappa_c = 10^{-2} \) are considered here.
indicates that the Taylor deformation should not be considered as the sole convergence criterion (since different combinations of $L/R$ and $B/R$ may result in the same $D$), but in conjunction with the specific capsule length and width values and its shape. In figure 5(b), it can be seen that the capsule shape corresponding to the coarsest mesh exhibits evident differences with respect to those of the finer meshes and the reference shape. As the relative errors between $R = 20$ and $R = 40$ are only $\sim 2\%$ and $0.5\%$ in the symmetric and asymmetric cases, we have decided to perform subsequent simulations considering $R = 20$.

We subsequently explore the performance of our numerical scheme on various combinations of stretching and bending moduli and surface tension ratios. Figure 6 shows the Taylor deformations obtained by our simulations, whose results are represented by lines, compared to the ones measured by Surface Evolver, denoted by dot symbols. As expected, the variations in the Taylor deformation found for a particular combination of $\kappa_s$ and $\kappa_b$ become more apparent with a decrease in the stretching modulus. For stiff capsules, that is $\kappa_s \geq 10^{-1}$, the capsule deformation reaches a plateau for high surface tension ratios. On the contrary, the results for highly deformable capsules, that is $\kappa_s \leq 10^{-3}$, tend to the ones obtained for the pure liquid lens configuration, depicted by filled square symbols. Here, a liquid lens is formed when a droplet of fluid 3 is suspended at the interface between fluids 1 and 2. Its shape and size depend on the force balance between the surface tensions at the three-phase contact line. It is also obvious that the effect of the bending coefficient on the capsule deformation is negligible for a given stretching modulus. Our results agree well with those of Surface Evolver, with a typical relative error in $D$ of approximately 5.6\% in both the symmetric and asymmetric cases. These discrepancies in the Taylor deformation could be dampened by increasing the computational domain size and potentially the parameter $\alpha$, as demonstrated earlier, with an increase though in the computational cost.

To illustrate the effect of these discrepancies on the mechanical equilibrium shape of the elastic capsule, the latter is plotted for different combinations of $\kappa_s$ and $\kappa_b$ at the highest surface tension ratios studied here, that is $\gamma_{12}/\gamma_{13} = 1.882$ in the symmetric case and $\gamma_{13}/\gamma_{23} = 3.40$ in the asymmetric one, as presented in figure 7. For stiff and moderately deformable capsules, corresponding to $\kappa_s = 10^{-1}$ and $\kappa_s = 10^{-2}$, an excellent agreement is observed between our simulation results and those of Surface Evolver. No notable differences can be seen in the capsule shapes obtained by our numerical model and Surface Evolver close to the three-phase contact point. For highly deformable capsules, a slightly elongated shape is obtained by the reference software compared to the one found by our fluid-structure solver. Deviations close to the three-phase contact point could be attributed to the considerable membrane curvature, as the curvature radius reduces and becomes comparable to the interface width. The capsule width, however, matches well.

We finally prove the Galilean invariance of the pro-
posed model. To do so, simulations are performed in an inertial frame of reference, and their results are compared to the ones obtained by the previous stationary simulations. At \( t = 0 \), all the fluid components are given a constant horizontal velocity \( U_{z,0} \). Due to the fluid-structure interaction, the initially circular capsule travels in the same direction with an equal velocity. Three different velocity values are tested here, \( U_{z,0} = \{ 10^{-4}, 10^{-3}, 10^{-2} \} \). The Galilean invariance is checked in both the symmetric and asymmetric (data not shown) cases for \( \kappa_s = 10^{-3} \) and \( \kappa_b = 10^{-2} \) at the highest surface tension ratios examined respectively here. The shapes of the elastic capsule, after the capsule reaches mechanical equilibrium in the inertial reference frame, found for the different velocities are compared to each other and to the ones of the corresponding stationary simulations. These comparisons can be seen in figure 8. In both cases, the results for the capsule shapes in the moving and stationary frames are superposed, indicating that the proposed model is Galilean invariant.

### B. Capillary bridge between two elastic capsules

To show the capabilities of our model, the configuration of a capillary bridge formed between two deformable capsules, as shown in figure 9(a), is now investigated. Two initially circular capsules of radius \( R = 20 \) are placed at \( (x_{c1}, y_{c1}) = (9R/2, 3R) \) and \( (x_{c2}, y_{c2}) = (15R/2, 3R) \) of a computational domain of dimensions

---

**FIG. 6.** Comparison of the Taylor deformation \( D \) between the results of the fluid-structure solver (lines) and Surface Evolver (●) for: (a) the symmetric case, and (b) the asymmetric one. The results of the fluid solver for the pure liquid lens configuration are also presented (■).

---

**FIG. 7.** Mechanical equilibrium shapes of the elastic capsule with \( \kappa_b = 10^{-2} \) for the (a) symmetric, and (b) asymmetric cases. The results of our simulations (lines) are compared to those of Surface Evolver (●), which are sub-sampled by a factor of 4 for viewing clarity. The dashed lines (- - -) represent the middle of the diffuse interface separating fluids 1 and 2.
of the capsules is presented in figure 10 for the different aspect ratio of the elastic capsules can be defined as main boundaries. At the converged state, the dimensions of the computational domain. Both the capillary bridge and elastic capsules are surrounded by the fluid component. Three different surface tension ratios are tested here, a relative area sampled by a factor of 5. The middle of the diffuse interface separating fluids 1 and 2 is denoted by dash lines (- - -). The capillary bridge, composed of the fluid component 2, is initialized as a rectangular area of dimensions $2S \times H_b = R \times 31R/10$ located at the center of the computational domain. Both the capillary bridge and elastic capsules are surrounded by the fluid component 1. Due to the presence of the surface tensions $\gamma_{12}$, $\gamma_{13}$ and $\gamma_{23}$ as well as the elastic strain and bending forces, the capsules relax to a deformed mechanical equilibrium shape depending on the forces balance. This mechanical equilibrium shape depends also on the initial distance between the two capsules, $2S$, and the volume of the capillary bridge. To confine the parameter space of the current study, all the simulations are performed only for a dimensionless initial distance $S' = 2S/R = 1$, and a relative area $A_{rel} = A_b/A_c \approx 1.02$, where $A_b$ and $A_c$ are the areas of the capillary bridge and elastic capsules. Three different surface tension ratios are tested here, $\gamma_{12}/\gamma_{13} \approx 0.57, 0.67, 0.80$. The parameter $\alpha$ is set to be $\alpha = 2$. The simulations are performed for various stretching moduli, $\kappa_b = \{10^{-2}, 10^{-3}, 10^{-2}, 10^{-1}, 10^{0}\}$, and constant bending and coupling coefficients, $\kappa_c = 10^{-2}$ and $\kappa_c = 10^{-2}$. As before, $\tau = \tau_b = \tau_c = 1$ and $\Gamma_b = \Gamma_c = 1$. Periodic boundary conditions are applied at all the domain boundaries. At the converged state, the dimensionless aspect ratio of the elastic capsules can be defined as $L_c/H_c$. The effect of stretching modulus on the aspect ratio of the capsules is presented in figure 10 for the different surface tension ratios. For low $\gamma_{12}/\gamma_{13}$, the capsules seem to take similar mechanical equilibrium shapes independently of their degree of elasticity. For moderate and high $\gamma_{12}/\gamma_{13}$, the aspect ratio changes significantly between stiff and moderately deformable ($\kappa_b = 10^{-2}$) capsules, while it reaches a plateau for highly deformable ($\kappa_b \leq 10^{-3}$) capsules. It can also be noted that the surface tension ratio affects considerably the capsules’ aspect ratio for a given stretching modulus. This can be clearly seen in figure 9(b), where the mechanical equilibrium shapes of the highly deformable ($\kappa_c = 10^{-3}$) capsules are plotted for the different $\gamma_{12}/\gamma_{13}$. At $\gamma_{12}/\gamma_{13} \approx 0.57$, the capsules retain an almost circular shape, having only the part of their surface coming in contact with the capillary bridge slightly compressed. As the surface tension ratio increases, the capsules move towards each other, taking a semi-circular shape and causing the formation of a narrower and higher capillary bridge. Despite the fact that the stretching modulus has a notable effect on the aspect ratio of the elastic capsules at high $\gamma_{12}/\gamma_{13}$, the corresponding variations in their shapes are small for different $\kappa_c$, as shown in figure 9(c). The capsule becomes slightly shorter and wider with an increase in $\kappa_c$.

Finally, the transient shapes of the highly deformable capsules at a surface tension ratio $\gamma_{12}/\gamma_{13} \approx 0.80$ are shown in figure 11. For clarity, we present only the results of the left-hand side capsule; the transient shapes of the right-hand side capsule are symmetric to the ones presented across the vertical centreline. As mentioned previously, the elastic capsules have initially, at $t_0 = 0$, a circular shape. It is worth noting that the capsules take quickly, already at $t = 5 \times 10^2$, a semi-circular shape similar to the mechanical equilibrium one. As the time passes, the capsules become narrower and more elongated along the $x$- and $y$-axis, respectively. Minimal changes in the capsules shapes can be observed between $t = 1 \times 10^5$, which corresponds to half of the total simulation time, and $t = 2 \times 10^5$, where the capsules are considered to have reached the mechanical equilibrium shape.

V. CONCLUSIONS

In this work, we have presented a novel modelling technique for the coupling of a multicomponent fluid flow with deformable, infinitely thin structures. For the sake of simplicity, we have considered the case where elastic membranes enclosing a fluid component 3 are immersed in a two-component/phase flow consisted of fluids 1 and 2. For this case, we have introduced a modified formulation of the free energy of the ternary fluid, taking into consideration its interaction with the elastic membranes, to the original one proposed by Semprebon et al. [45]. Taking advantage of the laws of thermodynamics, we have also derived the governing equations of motion for the fluid and the membrane.

The macroscopic equations of motion of the fluid-
FIG. 9. (a) Schematic diagram of a capillary bridge, composed of fluid (2), formed between two elastic membranes (−), enclosing the fluid component (3), while suspended in fluid (1). Comparison of the mechanical equilibrium shapes of the elastic capsules for: (b) different surface tension ratios $\gamma_{12}/\gamma_{13}$ at $\kappa_s = 10^{-3}$, and (c) different stretching moduli $\kappa_s$ at $\gamma_{12}/\gamma_{13} \approx 0.80$. The dashed lines (- - -) depict the capillary bridge boundaries.

FIG. 10. Aspect ratio $L_c/H_c$ of the elastic capsules as a function of stretching modulus $\kappa_s$ at different surface tension ratios $\gamma_{12}/\gamma_{13}$.

FIG. 11. Shapes of the highly ($\kappa_s = 10^{-3}$) deformable capsule at $\gamma_{12}/\gamma_{13} \approx 0.80$ at different time steps $t$.

structure system are solved here by a partitioned numerical approach. This approach consists of: a mesoscopic lattice Boltzmann method for resolving the ternary fluid flow in an Eulerian description, a finite difference method to evolve the membranes equations of motion in a Lagrangian framework, and an immersed boundary method to couple the Eulerian and Lagrangian solvers. The fluid and structure solvers are coupled through a forcing source term in the lattice Boltzmann equation, acting as a feedback of the structure’s response on the flow.

We have subsequently validated our computational algorithm against Surface Evolver, an open-source software capable of modelling steady liquid surface problems employing an energy minimization approach. The configuration of an elastic capsule placed at a fluid-fluid interface was considered as the benchmark test. We have compared in detail the equilibrium shapes of the capsule, and its corresponding deformation parameters for different scenarios of the surface tensions and combinations of the capsule’s stretching and bending moduli. An overall good agreement has been observed between our results and the reference ones. We have also demonstrated the Galilean invariance of our model equations. Finally, our algorithm has been applied to a more complex configuration, that is the capillary bridge formed between two elastic capsules. Although an extensive investigation of the parameter space was beyond the scope of the present work, it should be noted that this configuration is a particularly rich phenomenon, where the criteria for the bridge rupture and the case of unequal capsules are worth being studied in the future. Importantly, the proposed model also enables the study of dynamic configurations.

We have assumed here that all fluid components have the same density and viscosity. By modifying the Landau free-energy functional $\mathcal{E}_f$ of Eq. (2) in an appropriate way [46] and following a rationale similar to the one presented in Sec. II, our model could be extended to the case where the components of the ternary fluid mixture have
different densities. Extension to multicomponent fluids of variable viscosity is straightforward. External forces, such as gravity, can be readily taken into consideration by assigning $f_{\text{ext}} \neq 0$ in Eq. (27). Extension to three dimensions is planned for the future, which will enable us to deal with more complex and realistic configurations encountered in experiments. The model could also be generalised so as to include more fluid components enclosed in or surrounding the elastic membranes, allowing us to tackle a wider range of applications, for example capsules containing multiple phases [16, 17]. In addition, the formulations of the strain and bending energies could be modified to consider materials obeying different constitutive laws, such as non-linear hyperelastic materials or biological membranes. Finally, the structure solver could be adapted in order to simulate open surfaces, encountered for instance in the wetting of a soft substrate [20, 21].

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Appendix A: Derivation of the equations of motion of ternary fluids in interaction with elastic membranes

1. Entropy equation

The first law of thermodynamics can be formulated as

$$\frac{d}{dt}(U + E) = \frac{dW}{dt} + \frac{dQ}{dt}, \quad (A1)$$

where $t$ is time, $U$ and $E$ are, respectively, the internal and kinetic energies, $W$ is the work done on the ternary fluid system by its surroundings, and $Q$ denotes the amount of heat supplied to the system for its temperature $T$ to be kept constant. The total entropy $S$ can be split into two parts: the entropy of the system $S_{\text{sys}}$, and the entropy of its surroundings $S_{\text{sur}}$. By definition, the total free energy of the system can be expressed as

$$F = U - TS_{\text{sys}}. \quad (A2)$$

It is also known that the entropy of the system’s surroundings is related to the heat $Q$ by

$$dS_{\text{sur}} = -\frac{dQ}{T}. \quad (A3)$$

Taking into account Eqs. (A1)–(A3), it can be shown that

$$\frac{dS}{dt} = \frac{dS_{\text{sys}}}{dt} + \frac{dS_{\text{sur}}}{dt} = -\frac{1}{T} \frac{d(F + E)}{dt} + \frac{1}{T} \frac{dW}{dt}. \quad (A4)$$

Here, the system of interest consists of the ternary fluid (denoted by subscript $f$) and the membrane. Let us first concentrate on the terms related to the ternary fluid.

Given the simulation volume $V$, the free and kinetic energies of the ternary fluid can be defined as

$$F_f = \int_V f_f dV, \quad E_f = \frac{1}{2} \int_V \rho |\mathbf{u}|^2 dV, \quad (A5)$$

where $f_f$ is the free energy density of the ternary fluid, $\rho$ is the total mass density of the fluid mixture, and $\mathbf{u}$ denotes the mass-averaged velocity. The fluid free energy density is the sum of two contributions: a free energy density of the bulk fluid $f_b$, and a local free energy density gradient contribution $f_\nabla$ allowing the existence of diffuse fluid-fluid interfaces

$$f_f = f_b + f_\nabla. \quad (A6)$$

The interfacial free energy density term is defined here as

$$f_\nabla = \frac{1}{2} \sum_{m,n=1}^N c_{mn} \nabla C_m \cdot \nabla C_n, \quad (A7)$$

where $N$ denotes the number of components of the fluid mixture ($N = 3$ here), $c_{mn}$ is the cross influence parameter, and $C_m$ represents the concentration fraction of fluid $m$ ($m = 1, 2, 3$). The total mass density is given by

$$\rho = \sum_{m=1}^N C_m M_{w,m}, \quad (A8)$$

where $M_{w,m}$ denotes the weight of component $m$.

Following the rationale of Kou and Sun [52], we can obtain expressions for $dF_f/dt$ and $dE_f/dt$, by applying the Reynolds transport and Gauss divergence theorems to Eq. (A5), identical to those in [52], and the mass conservation law of the fluid mixture

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{u}) + \sum_{m=1}^N M_{w,m} \nabla \cdot \mathbf{J}_m = 0. \quad (A9)$$

$\mathbf{J}_m = M_m \nabla \mu_m$ denotes the diffusion flux of component $m$, and $M_m$ and $\mu_m$ represent the corresponding mobility parameters. The chemical potential $\mu_m$ is discussed in Sec. II C.

The rate of change of the work done by the force $F_{\text{sur}}$ is given by

$$\frac{dW}{dt} = \int_{\partial V} (F_{\text{sur}} \cdot \mathbf{u}) ds, \quad (A10)$$
where the integration takes place over the volume’s surface boundary \( \partial V \). The force \( F_{\text{sur}} \) is related to the Cauchy stress tensor \( \sigma \) of the ternary fluid by \( F_{\text{sur}} = -\sigma \cdot n \), where \( n \) denotes the outward unit normal vector of \( V \). Equation (A10) can thus take the form

\[
\frac{dW}{dt} = -\int_{\partial V} \left( (\sigma \cdot n) \cdot u \right) \, ds = -\int_V \left( \sigma^T \cdot \nabla u + u \cdot (\nabla \cdot \sigma) \right) \, dV, \tag{A11}
\]

where \( \sigma^T \) denotes the transpose of \( \sigma \).

By making use of the expressions for \( dF_i/dt \), \( dE_i/dt \) and \( dW/dt \), the terms on the right-hand side of Eq. (A4) related to the ternary fluid can be collected as follows

\[
-\frac{d}{dt} (F_i + E_i) + \frac{dW}{dt} = \int_V \left[ -\frac{\partial f_i}{\partial t} - \nabla \cdot (f_i u) + \frac{1}{2} \sum_{m=1}^{N} M_{w,m} \nabla \cdot ((u \cdot u) J_m) - \sigma^T \cdot \nabla u - u \cdot \left( \rho \frac{du}{dt} + \sum_{m=1}^{N} M_{w,m} J_m \cdot \nabla u + \nabla \cdot \sigma \right) \right] \, dV, \tag{A12}
\]

where \( \frac{du}{dt} \) is the total derivative of \( u \), defined as \( \frac{\partial u}{\partial t} + \nabla u \).

2. Transport equation of the free energy density of the ternary fluid

The pressure \( p \) of the fluid mixture can be defined as

\[
p = \sum_{m=1}^{N} C_m \mu_m - f_t, \tag{A13}
\]

where \( \mu_m \) is the chemical potential of component \( m \), given by

\[
\mu_m = \frac{\delta f_t}{\delta C_m} = \mu^b_m - \sum_{n=1}^{N} \nabla \cdot (c_{mn} \nabla C_n), \tag{A14}
\]

where \( \mu^b_m = \frac{\delta f_b}{\delta C_m} \) and \( \mu^\nabla_m = \frac{\delta f_{b,c}}{\delta C_m} \) stand for the bulk and interfacial chemical potentials of component \( m \). Due to Eq. (A14), Eq. (A13) can be rewritten as

\[
p = \sum_{m=1}^{N} C_m \mu^b_m - f_b - \sum_{m,n=1}^{N} C_m \nabla \cdot (c_{mn} \nabla C_n) p_b - \frac{1}{2} \sum_{m,n=1}^{N} c_{mn} \nabla C_m \cdot \nabla C_n. \tag{A15}
\]

The bulk pressure \( p_b \) can be further divided into two parts: \( p_{b,f} \) accounting for the contribution of the bulk free energy density owing to the chosen free-energy functional \( f_{b,f} \) of the fluid mixture, and \( p_{b,c} \) taking into account the contribution of the bulk free energy density due to a fluid-membrane coupling-energy functional \( f_{b,c} \)

\[
p_b = \sum_{m=1}^{N} C_m \mu^b_m - f_{b,f} + C_3 \frac{\delta f_{b,c}}{\delta C_3} (C_3, I) - f_{b,c}, \tag{A16}
\]

where \( f_b = f_{b,f} + f_{b,c} \) and \( \mu^b_m = \frac{\delta f_{b,f}}{\delta C_m} \). The coupling-energy functional \( f_{b,c} \) depends only on the component enclosed in the membrane, which is assumed to be fluid 3 in this work, and an interfacial profile \( I \) across the elastic membrane for the ternary fluid. It should be noted that the term \( p_{b,c} \) on the right-hand side of Eq. (A16) differentiates the present model from the multi-component flow model proposed by Kou and Sun [52], as the latter does not account for \( f_{b,c} \). The gradient of the bulk pressure can be found as

\[
\nabla p_b = \nabla \left( \sum_{m=1}^{N} C_m \mu^b_m - f_b \right) = \sum_{m=1}^{N} \left( \mu^b_m \nabla C_m + C_m \nabla \mu_m \right) - \sum_{m=1}^{N} \mu^b_m \nabla C_m - \frac{\delta f_b}{\delta I} \nabla I = \sum_{m=1}^{N} C_m \nabla \mu^b_m - \frac{\delta f_{b,c}}{\delta I} \nabla I. \tag{A17}
\]

It can also be shown that

\[
\nabla p_{b,c} - C_3 \nabla \left( \frac{\delta f_{b,c}}{\delta C_3} \right) = \nabla \left( C_3 \frac{\delta f_{b,c}}{\delta C_3} - f_{b,c} \right) - C_3 \nabla \left( \frac{\delta f_{b,c}}{\delta C_3} \right) = -\frac{\delta f_{b,c}}{\delta I} \nabla I. \tag{A18}
\]

Taking into account the above, the time derivative of the bulk \( f_b \) free energy density can be deduced as

\[
\frac{\partial f_b}{\partial t} = \sum_{m=1}^{N} \mu^b_m \frac{\partial C_m}{\partial t} + \frac{\delta f_b}{\delta I} \frac{\partial I}{\partial t} = -p_b \nabla \cdot u - \nabla \left( p_{b,c} - C_3 \nabla \left( \frac{\delta f_{b,c}}{\delta C_3} \right) \right) - \nabla \cdot (f_b u) - \sum_{m=1}^{N} \mu^b_m \nabla \cdot J_m + \frac{\delta f_{b,c}}{\delta I} \frac{\partial I}{\partial t}, \tag{A19}
\]

where the second and last terms on the right-hand side of Eq. (A19) are introduced here due to the presence of the elastic membrane, not being included in the corresponding expression in [52]. The time derivative and divergence of the interfacial \( f_{b,c} \) free energy density are formulated similarly to those in [52].
The transport equation of the free energy density \( f_t \) of the ternary fluid can then be found as

\[
\frac{\partial f_t}{\partial t} + \nabla \cdot (f_t \mathbf{u}) = -p \nabla \cdot \mathbf{u} - \sum_{m=1}^{N} \sum_{n=1}^{N} \nabla \cdot (( \nabla \cdot (C_n \mathbf{u})) c_{mn} \nabla C_m) - \sum_{m=1}^{N} \nabla \cdot (\mu_m \mathbf{J}_m) + \sum_{m=1}^{N} \mathbf{J}_m \cdot \nabla \mu_m
\]

\[
- \sum_{m=1}^{N} \nabla \cdot (( \nabla \cdot J_n) c_{mn} \nabla C_m) - \mathbf{u} \cdot \left( \nabla p_{b,c} - C_3 \nabla \left( \frac{\delta f_{b,c}}{\delta C_3} \right) \right) - \frac{\delta f_{b,c}}{\delta I} \frac{\partial I}{\partial t}.
\]

(A20)

By substituting Eq. (A20) into Eq. (A12), the latter is reformulated as

\[
- \frac{d (F_t + E_t)}{dt} + \frac{dW}{dt} = \int_{V} \left[ (p \mathbf{I} + \sum_{m=1}^{N} c_{mn} (\nabla C_m \otimes \nabla C_n)) : \nabla \mathbf{u} + \sum_{m=1}^{N} \nabla \cdot (\mu_m \mathbf{J}_m) - \sum_{m=1}^{N} \mathbf{J}_m \cdot \nabla \mu_m
\]

\[
+ \sum_{m=1}^{N} \nabla \cdot (( \nabla \cdot (C_n \mathbf{u})) c_{mn} \nabla C_m) - \nabla \cdot \left( \sum_{m=1}^{N} c_{mn} (\nabla C_m \otimes \nabla C_n) \cdot \mathbf{u} \right)
\]

\[
+ \sum_{m=1}^{N} \nabla \cdot (( \nabla \cdot J_n) c_{mn} \nabla C_m) + \mathbf{u} \cdot \left( \nabla p_{b,c} - C_3 \nabla \left( \frac{\delta f_{b,c}}{\delta C_3} \right) \right) - \frac{\delta f_{b,c}}{\delta I} \frac{\partial I}{\partial t} - \sigma^T : \nabla \mathbf{u}
\]

\[
+ \frac{1}{2} \sum_{m=1}^{N} M_{w,m} \nabla \cdot ((\mathbf{u} \cdot \mathbf{u}) \mathbf{J}_m) - \mathbf{u} \cdot \left( \rho \frac{d \mathbf{u}}{dt} + \sum_{m=1}^{N} M_{w,m} \mathbf{J}_m \cdot \nabla \mathbf{u} + \nabla \cdot \mathbf{\sigma} \right) \right] dV,
\]

(A21)

where \( \mathbf{I} \) is the second-order identity tensor.

### 3. Free and kinetic energies of the elastic membrane

Let us focus now on the terms of Eq. (A4) related to the membrane. The free energy \( F_m \) of the membrane is the sum of two contributions: strain energy \( \mathcal{E}_s \) and bending energy \( \mathcal{E}_b \),

\[
F_m = \mathcal{E}_s + \mathcal{E}_b.
\]

(A22)

The kinetic energy \( E_m \) of the membrane is given by

\[
E_m = \frac{1}{2} \int_{S} m \left| \frac{d \mathbf{X}}{dt} \right|^2 ds,
\]

(A23)

where the integration takes place over the surface \( S \) of the elastic membrane, whose position in Eulerian coordinates at time \( t \) is described by \( \mathbf{X} = \mathbf{X}(\mathbf{s}, t) \), where \( \mathbf{s} \) denotes its Lagrangian coordinates, and \( m \) is the mass surface density of the membrane.

### 4. Equations of motion

By combining Eqs. (A21)–(A23), the entropy Eq. (A4) can be rewritten as

\[
T \frac{dS}{dt} = - \frac{d (F_t + E_t)}{dt} - \frac{d (F_m + E_m)}{dt} + \frac{dW}{dt}
\]

\[
= - \int_{V} \left[ \sigma^T - \rho \mathbf{I} + \sum_{m=1}^{N} c_{mn} (\nabla C_m \otimes \nabla C_n) : \nabla \mathbf{u} \right] dV
\]

\[
- \int_{V} \sum_{m=1}^{N} \mathbf{J}_m \cdot \nabla \mu_m dV - \sum_{m=1}^{N} M_{w,m} \mathbf{J}_m \cdot \nabla \mathbf{u}
\]

\[
+ \nabla \cdot \mathbf{\sigma} - \nabla p_{b,c} + C_3 \nabla \left( \frac{\delta f_{b,c}}{\delta C_3} \right) dV
\]

\[
- \int_{V} \frac{\delta f_{b,c}}{\delta I} \frac{\partial I}{\partial t} dV - \frac{d \mathcal{E}_s}{dt} - \frac{d \mathcal{E}_b}{dt} - \int_{S} m \frac{d \mathbf{X}}{dt} \frac{d^2 \mathbf{X}}{dt^2} ds.
\]

(A24)

Eq. (A24) differs from the expression of the rate of change of the total entropy \( S \) in [52] in the last two terms on the penultimate line and all the terms on the last line, which are present here due to the existence of elastic membranes. We consider the same natural boundary conditions and formulation of the stress tensor \( \mathbf{\sigma} \) of the ternary fluid as in [52].

According to the second law of thermodynamics, the total entropy cannot decrease over time. This, in combination with the non-negative nature of the third term on
the right-hand side of Eq. (A24), implies that
\[
\rho \frac{du}{dt} + \sum_{m=1}^{N} M_{w,m} \nabla u + \nabla \cdot \sigma - \nabla p_{v,c} + C_{3} \nabla \left( \frac{\delta f_{b,c}}{\delta C_{3}} \right) = 0.
\]  
(A25)

In the present work, we consider that the component weights \( M_{w,m} \) are equal, and \( \sum_{m=1}^{N} M_{w,m} J_{m} = 0 \). For consistency purposes, the ideal gas pressure term, \( p_{v} \), in the Navier-Stokes Eq. (A25) is separated in the following. As such, the continuity Eq. (A9) and the Navier-Stokes Eq. (A25) take, respectively, the form (Eqs. (11) and (12) in Sec. II C of the article)
\[
\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho u) = 0,
\]  
(A26)

where the auxiliary fields \( \phi = C_{1} - C_{2} \) and \( \psi = C_{3} \) (Eq. (10) in Sec. II C of the article) have also been introduced. The equation of motion of the elastic membrane can be derived by considering the last four terms on the right-hand side of Eq. (A24), and setting their sum equal to zero.