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Interaction between surface shape and intra-surface viscous flow on lipid membranes

Padmini Rangamani · Ashutosh Agrawal · Kranthi K. Mandadapu · George Oster · David J. Steigmann

Abstract The theory of intra-surface viscous flow on lipid bilayers is developed by combining the equations for flow on a curved surface with those that describe the elastic resistance of the bilayer to flexure. The model is derived directly from balance laws and augments an alternative formulation based on a variational principle. Conditions holding along an edge of the membrane are emphasized, and the coupling between flow and membrane shape is simulated numerically.

Keywords Lipid bilayers · Viscous membranes · Surface flow · Bending elasticity

1 Introduction

A formulation of the nonlinear mechanics of lipid membranes, incorporating intra-membrane viscous flow and accounting for viscous interaction with bulk liquids, has recently been developed in Arroyo and DeSimone (2009). There, it is demonstrated that intra-membrane viscosity has a significant effect on the dynamics of the system vis a vis bulk viscosity at biologically relevant length scales, contrary

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to earlier claims made in the literature (Seifert 1997). This finding furnishes impetus for the general theory of surface flow on lipid membranes.

The work reported in Arroyo and DeSimone (2009) relies on a variational principle combined with exterior differential calculus. While such a framework is entirely satisfactory from a theoretical point of view, we believe that understanding of the subject would be promoted by the availability of an alternative treatment-based a priori on balance laws and associated constitutive equations. Indeed, the authors of Arroyo and DeSimone (2009) allude to the possibility of developing the theory from the balance laws and boundary conditions of the elastic model (Steigmann 1999a), suitably extended to account for the effects of intra-membrane viscosity. They refer to such a development as a 'conceptually interesting exercise' which, however, lay beyond the scope of their investigation. Our purpose in the present work is precisely to provide such a development. Thus, while the variational approach has intrinsic value, here we adopt Scriven's (1960) approach to the theory of surface flow, which was developed entirely on the basis of balance laws and constitutive equations. In this way, the methods of the older theory are retained, allowing us to distinguish more sharply the non-standard effects associated with the bending stiffness of lipid membranes.

Moreover, because differential forms are not widely used in continuum mechanics, we work in the setting of tensor analysis on surfaces. Our view is that this framework, which is more in keeping with the methods underlying conventional bulk-fluid mechanics, is adequate for formulating the general theory and addressing boundary-value problems.

Section 2 contains a complete summary of the conventional elastic theory and boundary conditions arising from a treatment based directly on balance laws rather than variational principles (Steigmann 1999a). These of

course are equivalent to the model derived from variational considerations (Steigmann et al. 2003; Agrawal and Steigmann 2009a,b). In the present work, the notions of area incompressibility and uniformity of the material properties are developed in detail, and the adjustments to the theory required to accommodate non-uniform material response are indicated. In Sect. 3, we focus on uniform membranes and introduce intra-membrane viscosity via a well-established interfacial flow model (Scriven 1960; Aris 1989). There we also develop the adjustments to the equilibrium and boundary conditions required by the inclusion of intra-membrane viscosity. Section 4 is concerned with the development of the equations of the model in the Monge parametrization, as a prerequisite to the numerical analysis of some example problems. For simplicity, we do not include viscous bulk interactions and thus impose a uniformly distributed pressure on the lipid membrane; that is, we effectively assume the bulk liquid to be inviscid and thus that it transmits a uniform pressure to the membrane in the absence of inertial effects. In light of the work reported in Arroyo and DeSimone (2009), this simplification may be regarded as realistic at sufficiently small length scales. In any case, viscous interaction with the bulk may be taken into account in a straightforward manner and so is not considered here (see for example Secomb and Skalak 1982; Arroyo and DeSimone 2009).

2 Inviscid membranes

In this section, we present a thorough overview of the conventional purely elastic theory of lipid membranes based on a free energy per unit mass. Our purpose is to establish a framework that is sufficiently general to subsume the current literature and to allow for straightforward extension to accommodate surface flow and additional effects such as diffusion (Agrawal and Steigmann 2011). The present work is limited to the consideration of elastic response combined with surface flow. Later, in Sect. 2.7, we connect this formulation to the more commonly used framework based on a free energy per unit area of the surface occupied by the membrane in its current configuration.

2.1 Elastic surfaces

Lipid membranes are special elastic surfaces with energy densities that respond to changes in surface metric and curvature. The equations of motion in the absence of inertia are simply the equations of mechanical equilibrium. For an elastic surface, subjected to a net lateral pressure p in the direction of the local surface unit normal $\mathbf{n}$, these may be summarized in the compact form (Steigmann 1999a):

$$\mathbf{T}^\alpha_{;\alpha} + p\mathbf{n} = \mathbf{0}, \tag{1}$$

where $\mathbf{T}^\alpha$ are the so-called *stress vectors* and Greek indices range over $\{1, 2\}$. These are proportional to the forces, per unit length, transmitted across the curves on which the surface coordinates θ^α are constant. Here the semi-colon refers to covariant differentiation with respect to the metric $a_{\alpha\beta} = \mathbf{a}_\alpha \cdot \mathbf{a}_\beta$, where $\mathbf{a}_\alpha = \mathbf{r}_{,\alpha}$ and commas identify partial derivatives with respect to θ^α. The $\mathbf{a}_\alpha$ comprise the natural tangent basis on the surface induced by the parametrization $\mathbf{r}(\theta^\mu, t)$ of the position field. The connection to the unit normal field is $\mathbf{n} = \mathbf{a}_1 \times \mathbf{a}_2 / |\mathbf{a}_1 \times \mathbf{a}_2|$. Here and henceforth, Greek indices range over $\{1, 2\}$ and, if repeated, are summed over that range. We assume familiarity with tensor analysis and curvilinear coordinate systems. Useful introductions for mechanicians are given in Sokolnikoff (1964), Kreyzsig (1959).

We denote the membrane surface by ω. The coordinate system θ^μ on ω plays a role analogous to that of a fixed-coordinate system used to parametrize a control volume in the Eulerian or spatial description of classical fluid mechanics.

The differential operation in (1) is the surface divergence, given explicitly by

$$\mathbf{T}^\alpha_{;\alpha} = (\sqrt{a})^{-1}(\sqrt{a}\mathbf{T}^\alpha)_{,\alpha}, \tag{2}$$

where $a = \det(a_{\alpha\beta})$. The metric is a positive-definite matrix, with $a > 0$, and so the divergence is well defined. The metric (the coefficients of the first fundamental form) is one of the two basic variables in surface theory; the other is the curvature $b_{\alpha\beta}$ (the coefficients of the 2nd fundamental form), defined by $b_{\alpha\beta} = \mathbf{n} \cdot \mathbf{r}_{,\alpha\beta}$.

This framework encompasses all elastic surfaces for which the energy density responds to metric and curvature. For example, if the energy density per unit mass of the surface is $F(a_{\alpha\beta}, b_{\alpha\beta})$, then (Steigmann 1999a)

$$\mathbf{T}^\alpha = \mathbf{N}^\alpha + S^\alpha \mathbf{n}, \tag{3}$$

where

$$\mathbf{N}^\alpha = N^{\beta\alpha}\mathbf{a}_\beta \quad \text{with} \quad N^{\beta\alpha} = \sigma^{\beta\alpha} + b^\beta_\mu M^{\mu\alpha}, \quad S^\alpha = -M^{\alpha\beta}_{;\beta}. \tag{4}$$

These are given in terms of the energy density by Steigmann (1999a)

$$\sigma^{\beta\alpha} = \rho\left(\frac{\partial F}{\partial a_{\alpha\beta}} + \frac{\partial F}{\partial a_{\beta\alpha}}\right)$$
$$M^{\beta\alpha} = \frac{1}{2}\rho\left(\frac{\partial F}{\partial b_{\alpha\beta}} + \frac{\partial F}{\partial b_{\beta\alpha}}\right), \tag{5}$$

where ρ is the surface mass density. Substituting (3) and (4) into (1), invoking the Gauss and Weingarten equations (Sokolnikoff 1964) $\mathbf{a}_{\beta;\alpha} = b_{\beta\alpha}\mathbf{n}$ and $\mathbf{n}_{,\alpha} = -b^\beta_\alpha\mathbf{a}_\beta$, where $b^\alpha_\beta = a^{\alpha\lambda}b_{\lambda\beta}$, and projecting the result onto the tangent and normal spaces to ω furnishes the three equations

$$N^{\beta\alpha}_{;\alpha} - S^\alpha b^\beta_\alpha = 0, \qquad S^\alpha_{;\alpha} + N^{\beta\alpha}b_{\beta\alpha} + p = 0. \tag{6}$$

This setting subsumes classical capillarity theory in which the energy depends in a special manner on metric alone ($M^{\beta\alpha} = 0$). It also incorporates the well-known Helfrich theory (Helfrich 1973; Ou-Yang et al. 1999) of lipid bilayers and monolayers in which the lipids are of fixed length and everywhere aligned with the surface normal.

2.2 Edge conditions

Edge conditions may be deduced from a mechanical power balance (Steigmann 1999b) which is simply the global form of the equation of motion in which inertia is suppressed. In the purely elastic theory, this is

$$\frac{d}{dt}E = P, \tag{7}$$

where

$$P = \int_{\omega} p\mathbf{n} \cdot \mathbf{u}\,da + P_b \tag{8}$$

is the power supplied to the membrane, $\mathbf{u}(\theta^{\alpha}, t)$ is the membrane velocity field,

$$E = \int_{\omega} \rho F\,da \tag{9}$$

is the net film energy and P_b is the power supplied by the forces and moments acting at the boundary $\partial\omega$. This is Steigmann (1999a,b)

$$P_b = \int_{\partial\omega} (\mathbf{f} \cdot \mathbf{u} + M\mathbf{n} \cdot \mathbf{u}_{,\nu})ds + \sum \mathbf{f}_i \cdot \mathbf{u}_i, \tag{10}$$

where $\mathbf{u}_{,\nu}$ is the normal derivative of $\mathbf{u}$ on $\partial\omega$ with exterior unit normal $\mathbf{\nu} = \nu_{\alpha}\mathbf{a}^{\alpha}$ and unit tangent $\mathbf{\tau} = \mathbf{n} \times \mathbf{\nu}$,

$$\mathbf{f} = \mathbf{T}^{\alpha}\nu_{\alpha} - (M^{\alpha\beta}\tau_{\alpha}\nu_{\beta}\mathbf{n})', \qquad M = M^{\alpha\beta}\nu_{\alpha}\nu_{\beta} \tag{11}$$

are the distributed force and bending couple on the edge, $(\cdot)' = d(\cdot)/ds$ is the arclength derivative on $\partial\omega$ in the direction of $\mathbf{\tau}$, and

$$\mathbf{f}_i = -M^{\alpha\beta}[\tau_{\alpha}\nu_{\beta}]_i\mathbf{n} \tag{12}$$

is the force exerted at the ith corner of $\partial\omega$ if the boundary is piecewise smooth with a finite number of points where $\mathbf{\tau}$ (and hence $\mathbf{\nu}$) is discontinuous; here the square bracket refers to the forward jump as the ith corner is traversed, and the sum in (10) accounts for all corners. The corner forces vanish if the boundary is smooth in the sense that its tangent $\mathbf{\tau}$ is continuous.

The expression (10) for the power is convenient because $\mathbf{u}$ and $\mathbf{u}_{,\nu}$ may be specified independently on $\partial\omega$. In particular (Agrawal and Steigmann 2009a) $\mathbf{n} \cdot \mathbf{u}_{,\nu} = -\mathbf{\tau} \cdot \mathbf{\omega}$ where $\mathbf{\omega}$ is the rate of change of the surface orientation; i.e., the rotational

velocity of the unit normal $\mathbf{n}$. This yields $M\mathbf{n} \cdot \mathbf{u}_{,\nu} = -M\mathbf{\tau} \cdot \mathbf{\omega}$ and hence the interpretation of M as a bending couple.

2.3 Lipid membranes

The elastic theory of lipid membranes is based on free-energy densities of the form $F(\rho, H, K)$, where

$$H = \frac{1}{2}a^{\alpha\beta}b_{\alpha\beta}, \qquad K = \frac{1}{2}\varepsilon^{\alpha\beta}\varepsilon^{\lambda\mu}b_{\alpha\lambda}b_{\beta\mu}, \tag{13}$$

respectively, are the mean and Gaussian curvatures of the surface. Here $(a^{\alpha\beta}) = (a_{\alpha\beta})^{-1}$ is the dual metric and $\varepsilon^{\alpha\beta}$ is the permutation tensor defined by $\varepsilon^{12} = -\varepsilon^{21} = 1/\sqrt{a}$, $\varepsilon^{11} = \varepsilon^{22} = 0$. The set $\{\rho, H, K\}$ of independent variables in the free-energy function is dictated by material symmetry requirements pertaining to the fluid-like response characteristics of lipid membranes. The underlying concept is developed fully in Steigmann (1999a, 2003).

In this case, the functions listed in (4) and (5) reduce to (Steigmann 1999a)

$$\sigma^{\alpha\beta} = -[\rho^2 F_{\rho} + 2H(\rho F_H) + 2K(\rho F_K)]a^{\alpha\beta} + (\rho F_H)\tilde{b}^{\alpha\beta},$$

$$M^{\alpha\beta} = \frac{1}{2}(\rho F_H)a^{\alpha\beta} + (\rho F_K)\tilde{b}^{\alpha\beta},$$

$$N^{\alpha\beta} = -[\rho^2 F_{\rho} + H(\rho F_H) + K(\rho F_K)]a^{\alpha\beta} + \frac{1}{2}(\rho F_H)\tilde{b}^{\alpha\beta},$$

$$-S^{\alpha} = \frac{1}{2}(\rho F_H)_{,\beta}a^{\alpha\beta} + (\rho F_K)_{,\beta}\tilde{b}^{\alpha\beta}, \tag{14}$$

where

$$\tilde{b}^{\alpha\beta} = 2Ha^{\alpha\beta} - b^{\alpha\beta} \tag{15}$$

is the cofactor of the curvature, in which $b^{\alpha\beta} = a^{\alpha\lambda}a^{\beta\mu}b_{\lambda\mu}$, and use has been made of the Mainardi–Codazzi equations of surface theory (Sokolnikoff 1964) in the form (Steigmann 1999a)

$$\tilde{b}^{\alpha\beta}_{;\beta} = 0. \tag{16}$$

Later, we use the connection

$$b^{\beta}_{\mu}\tilde{b}^{\mu\alpha} = Ka^{\alpha\beta}. \tag{17}$$

From (14) we have,

$$N^{\beta\alpha}_{;\alpha} - S^{\alpha}b^{\beta}_{\alpha} = -[(\rho^2 F_{\rho})_{,\alpha} + \rho(F_K K_{,\alpha} + F_H H_{,\alpha})]a^{\beta\alpha}, \tag{18}$$

and the positive definiteness of the metric then furnishes the tangential equilibrium equations (cf. (6)$_1$)

$$(\rho^2 F_{\rho})_{,\alpha} + \rho(F_K K_{,\alpha} + F_H H_{,\alpha}) = 0, \tag{19}$$

whereas the normal part of the equilibrium equation reduces to

$$p = \Delta\left(\frac{1}{2}\rho F_H\right) + (\rho F_K)_{;\alpha\beta}\tilde{b}^{\alpha\beta} + 2H\rho(\rho F_{\rho} + K F_K)$$
$$+ \rho(2H^2 - K)F_H, \tag{20}$$

in which $\Delta(\cdot) = (\cdot)_{;\alpha\beta} a^{\alpha\beta}$ is the surface Laplacian, also known as the Beltrami operator. The subscripts ρ, H, K refer to partial derivatives of the energy with respect to the indicated variables. Equations (19) and (20) together constitute the generalization of the well-known *shape equation* (Ou-Yang et al. 1999) to films with possibly non-uniform properties (Jenkins 1977; Steigmann 1999a).

In the absence of bending elasticity, this system reduces to the equations of classical capillarity theory:

$$p = 2H(\rho^2 F_\rho) \quad \text{in which} \quad (\rho^2 F_\rho)_{,\alpha} = 0. \tag{21}$$

2.4 Convected coordinates

To ease the formulation of balance laws and to facilitate their correct interpretation, we parametrize the material manifold by a convected-coordinate system ξ^α. This may be identified with the system θ^α at a fixed instant t_0, say. The associated surface Ω, with parametric representation $\mathbf{x}(\xi^\alpha) = \mathbf{r}(\xi^\alpha, t_0)$, is fixed and may serve as a reference surface in a Lagrangian or referential description of the motion. That is, we regard these coordinates as being convected in the sense that they identify, via a map $\mathbf{r} = \hat{\mathbf{r}}(\xi^\alpha, t)$, the current position at time t of a material point that was located at $\mathbf{x}(\xi^\alpha) \in \Omega$ at time t_0. The notion may be generalized by regarding Ω as a surface that is in one-to-one correspondence with that occupied at time t_0, so that Ω need not actually be occupied in the course of the motion. The connection with the θ^α-parametrization of ω is provided by Scriven (1960), Aris (1989)

$$\hat{\mathbf{r}}(\xi^\alpha, t) = \mathbf{r}(\theta^\beta(\xi^\alpha, t), t). \tag{22}$$

Thus, we specify the fixed surface coordinates θ^α as functions of ξ^α and t subject to $\theta^\alpha(\xi^\beta, t_0) = \xi^\alpha$. We assume the relations giving θ^α in terms of ξ^α to be invertible, to reflect the notion that at fixed t, the coordinates θ^α can be associated with a unique material point (identified by fixed values of ξ^α). Any function, $f(\theta^\alpha, t)$, say, may then be expressed in terms of convected coordinates as $\hat{f}(\xi^\alpha, t)$, where

$$\hat{f}(\xi^\alpha, t) = f(\theta^\beta(\xi^\alpha, t), t). \tag{23}$$

The material derivative of f is its partial time derivative in the convected-coordinate representation, i.e., $\dot{f} = \partial \hat{f}(\xi^\alpha, t)/\partial t$, whereas its local time derivative in the fixed-coordinate parametrization is $f_t = \partial f(\theta^\alpha, t)/\partial t$. By the chain rule, the two are related by $\dot{f} = f_t + (\theta^\alpha)^{\cdot} f_{,\alpha}$.

The velocity of a material point on Ω that has been convected by the motion to ω is $\mathbf{u} = \dot{\mathbf{r}} = \partial \hat{\mathbf{r}}/\partial t$. We may write this in terms of components on the natural basis induced by the fixed-coordinate (θ^α) parametrization. Thus,

$$\mathbf{u} = v^\alpha \mathbf{a}_\alpha + w\mathbf{n}. \tag{24}$$

This is not the same as the time derivative $\mathbf{r}_t$. However, the two are related by

$$\mathbf{u} = (\theta^\alpha)^{\cdot} \mathbf{a}_\alpha + \mathbf{r}_t. \tag{25}$$

Following (Aris 1989), we adopt the fixed-coordinate parametrization defined by

$$\frac{d}{dt}\theta^\alpha = v^\alpha(\theta^\beta, t), \quad \theta^\alpha_{|t_0} = \xi^\alpha, \tag{26}$$

where the derivative is evaluated at a fixed value of the doublet $\{\xi^\alpha\}$ and is therefore equal to $(\theta^\alpha)^{\cdot}$. Accordingly, the normal velocity in (24) is given by

$$w\mathbf{n} = \mathbf{r}_t, \tag{27}$$

and the convected and fixed-coordinate time derivatives satisfy

$$\dot{f} = f_t + v^\alpha f_{,\alpha}. \tag{28}$$

Later, we require an expression for the material derivative $\dot{a}_{\alpha\beta}$ in terms of the θ^α-parametrization. To this end, we adopt convected coordinates ξ^α whose values coincide with the instantaneous values of θ^α. The two coordinate systems will of course differ at different instants due to the fact that material is convecting with respect to the θ^α-system. Said differently, the material point instantaneously located at the place with surface coordinates θ^α will have different locations at different instants and hence be associated with different values of θ^α, whereas the values of ξ^α remain invariant. Accordingly, while it is always permissible to identify ξ^α with θ^α at any particular instant t_0, say, it is not possible to do so over an interval of time. However, for our purposes, this limitation is not restrictive. Using

$$\dot{a}_{\lambda\mu} = \dot{\mathbf{a}}_\lambda \cdot \mathbf{a}_\mu + \mathbf{a}_\lambda \cdot \dot{\mathbf{a}}_\mu \quad \text{and}$$

$$\begin{aligned}
\dot{\mathbf{a}}_\lambda &= (\partial \mathbf{r}/\partial \theta^\lambda)^{\cdot} = [\partial \mathbf{r}/\partial \xi^\mu (\partial \xi^\mu/\partial \theta^\lambda)]^{\cdot} \\
&= \partial \mathbf{u}/\partial \xi^\mu (\partial \xi^\mu/\partial \theta^\lambda) + \partial \mathbf{r}/\partial \xi^\mu (\partial^2 \xi^\mu/\partial \theta^\lambda \partial \theta^\alpha) v^\alpha,
\end{aligned} \tag{29}$$

together with $\partial \xi^\mu/\partial \theta^\lambda = \delta^\mu_\lambda$ (the Kronecker delta) and hence $\partial^2 \xi^\mu/\partial \theta^\lambda \partial \theta^\alpha = 0$ at time t_0, we derive $\dot{\mathbf{a}}_\alpha = \partial \mathbf{u}/\partial \xi^\alpha$ and

$$\dot{a}_{\lambda\mu} = \mathbf{u}_{,\lambda} \cdot \mathbf{a}_\mu + \mathbf{a}_\lambda \cdot \mathbf{u}_{,\mu}, \tag{30}$$

where $\mathbf{u}_{,\lambda} = \partial \mathbf{u}/\partial \theta^\lambda$ at the considered instant.

Combining (24) with the Gauss and Weingarten equations yields

$$\mathbf{u}_{,\lambda} = (v_{\alpha;\lambda} - wb_{\alpha\lambda})\mathbf{a}^\alpha + (v^\alpha b_{\alpha\lambda} + w_{,\lambda})\mathbf{n}, \tag{31}$$

where $\mathbf{a}^\alpha = a^{\alpha\beta}\mathbf{a}_\beta$ and $v_{\alpha;\lambda}$ is the covariant derivative defined by

$$v_{\alpha;\lambda} = v_{\alpha,\lambda} - v_\beta \Gamma^\beta_{\alpha\lambda}, \tag{32}$$

in which $\Gamma^\beta_{\alpha\lambda}$ are the Christoffel symbols on ω computed using the θ^α-system (Sokolnikoff 1964). This delivers the desired expression (Steigmann et al. 2003):

$$\dot{a}_{\lambda\mu} = v_{\mu;\lambda} + v_{\lambda;\mu} - 2wb_{\lambda\mu}. \tag{33}$$

The corresponding result in Aris' book (Aris 1989; eqs. (10.21.3), (10.21.4)) is given, in our notation, by

$$\dot{a}_{\lambda\mu} = v_{\mu;\lambda} + v_{\lambda;\mu} + (a_{\lambda\mu})_t, \tag{34}$$

where $(\cdot)_t$ is computed at fixed θ^α. The latter is $(a_{\lambda\mu})_t = (\mathbf{a}_\lambda)_t \cdot \mathbf{a}_\mu + \mathbf{a}_\lambda \cdot (\mathbf{a}_\mu)_t$, where $(\mathbf{a}_\lambda)_t = (\mathbf{r}_{,\lambda})_t = (\mathbf{r}_t)_{,\lambda}$, and (cf. (27))

$$(\mathbf{r}_t)_{,\lambda} = (w\mathbf{n})_{,\lambda} = w_{,\lambda}\mathbf{n} - wb_{\lambda\alpha}\mathbf{a}^\alpha, \tag{35}$$

yielding $(a_{\lambda\mu})_t = -2wb_{\lambda\mu}$, in agreement with (33). However, (33) is not given explicitly in Aris (1989).

2.5 Mass balance

The foregoing relationships facilitate the derivation of balance laws. For example, if f is the areal density of a particular quantity on ω, then the rate of change of the total quantity in a part π of ω is

$$\frac{d}{dt} \int_\pi f \, da = \frac{d}{dt} \int_\Pi f J \, dA = \int_\pi (\dot{f} + f \dot{J}/J) \, da, \tag{36}$$

where Π is the part of the fixed surface Ω that is convected to π and J is the local areal dilation of the surface; i.e.,

$$\int_\pi da = \int_\Pi J \, dA \quad \text{for all} \quad \Pi \subset \Omega. \tag{37}$$

To express the right-hand side of (36) in terms of the fixed-coordinate parametrization, we combine $\dot{J}/J = \frac{1}{2}a^{\alpha\beta}\dot{a}_{\alpha\beta}$ (Steigmann et al. 2003) with (33), obtaining

$$\dot{J}/J = v^\alpha_{;\alpha} - 2Hw. \tag{38}$$

For example, mass conservation is expressed by

$$\frac{d}{dt} \int_\pi \rho \, da = 0. \tag{39}$$

Using (36) with $f = \rho$ and invoking the arbitrariness of π then yields the local conservation law

$$0 = \dot{\rho} + \rho \dot{J}/J = \rho_t + v^\alpha \rho_{,\alpha} + \rho(v^\alpha_{;\alpha} - 2Hw). \tag{40}$$

2.6 Area incompressibility

It is well known that lipid membranes are relatively stiff against areal dilation in comparison with bending or shearing in the tangent plane (Evans and Skalak 1980; Secomb and Skalak 1982). To model this, we impose $J = 1$ as a local constraint at material points. Accordingly, $\dot{J}$ vanishes and (40) simplifies to

$$0 = \dot{\rho} = \rho_t + v^\alpha \rho_{,\alpha}, \tag{41}$$

the first of which implies that ρ is independent of t in the convected-coordinate description; that is, ρ is independent of t when expressed as a function of ξ^μ and t. Its value at a particular material point is thus invariant in time and hence given by the density in the fixed configuration associated with Ω.

The constraint on J is seen to be equivalent to the invariance of ρ at any material point. To accommodate it, we replace F in Sect. 2.3 by

$$F(\rho, H, K; \xi^\mu, t) = \bar{F}(H, K; \xi^\mu) - \gamma(\xi^\mu, t)/\rho, \tag{42}$$

where γ is a constitutively indeterminate Lagrange-multiplier field. The latter takes whatever values that may be required by the equations of equilibrium and any subsidiary conditions in the particular problem at hand and may thus depend on the coordinates and time. This yields the formula $\gamma = \rho^2 F_\rho$ and hence the mechanical interpretation of the Lagrange-multiplier field γ as a surface pressure. However, it is *not* a material property and so its a priori specification in terms of the known surface tension of a particular liquid—commonplace in works concerned with incompressible lipid membranes—is logically inconsistent. This point appears to be a source of considerable misunderstanding in the literature.

2.7 Areal free-energy density

It is customary in the literature to formulate the mechanics of lipid membranes in terms of the free energy per unit area of the surface ω currently occupied by the material; namely

$$W = \rho\bar{F}. \tag{43}$$

In terms of this, we have

$$\sigma^{\alpha\beta} = (\lambda + W)a^{\alpha\beta} - (2HW_H + 2KW_K)a^{\alpha\beta} + W_H\tilde{b}^{\alpha\beta},$$
$$M^{\alpha\beta} = \frac{1}{2}W_H a^{\alpha\beta} + W_K\tilde{b}^{\alpha\beta},$$
$$N^{\alpha\beta} = (\lambda + W)a^{\alpha\beta} - (HW_H + KW_K)a^{\alpha\beta} + \frac{1}{2}W_H\tilde{b}^{\alpha\beta},$$
$$-S^\alpha = \frac{1}{2}(W_H)_{,\beta}a^{\alpha\beta} + (W_K)_{,\beta}\tilde{b}^{\alpha\beta}, \tag{44}$$

where

$$\lambda = -(\gamma + W). \tag{45}$$

The shape equation (20) is then seen to be equivalent to

$$p = \Delta\left(\frac{1}{2}W_H\right) + (W_K)_{;\alpha\beta}\tilde{b}^{\alpha\beta} + W_H(2H^2 - K)$$
$$+ 2H(KW_K - W) - 2\lambda H, \tag{46}$$

whereas

$$N^{\beta\alpha}_{;\alpha} - S^\alpha b^\beta_\alpha = -(\gamma_{,\alpha} + W_K K_{,\alpha} + W_H H_{,\alpha})a^{\beta\alpha}$$
$$= (\partial W/\partial\theta^\alpha_{|\exp} + \lambda_{,\alpha})a^{\beta\alpha}, \tag{47}$$

wherein the derivative $\partial W/\partial\theta^\alpha_{|\exp}$ accounts for any explicit coordinate dependence of the material properties arising in non-uniform membranes. Further, the Lagrange multiplier is presumed, by virtue of the one-to-one relation between the convected and fixed coordinates at any t, to be expressible in terms of θ^α; that is, $\gamma_{,\alpha} = \partial\gamma/\partial\theta^\alpha$. The tangential equations of equilibrium are then equivalent to Agrawal and Steigmann (2009a,b)

$$\lambda_{,\alpha} = -\partial W/\partial\theta^\alpha_{|\exp}. \tag{48}$$

The specializations of the edge conditions follow by substituting (44) into (3), (4) and (11). The involved calculations are found to yield results which coincide exactly with those derived in detail elsewhere (Agrawal and Steigmann 2009a,b) by a variational method. These are

$$M = \frac{1}{2}W_H + \kappa_\tau W_K \quad \text{and} \quad \mathbf{f} = f_\nu\boldsymbol{\nu} + f_\tau\boldsymbol{\tau} + f_n\mathbf{n}, \tag{49}$$

where

$$\begin{aligned} f_\nu &= W + \lambda - \kappa_\nu M \\ f_\tau &= -\tau M, \\ f_n &= (\tau W_K)' - \left(\frac{1}{2}W_H\right)_{,\nu} - (W_K)_{,\beta}\tilde{b}^{\alpha\beta}\nu_\alpha, \end{aligned} \tag{50}$$

and

$$\mathbf{f}_i = W_K[\tau]_i\mathbf{n}. \tag{51}$$

Here,

$$\kappa_\nu = b_{\alpha\beta}\nu^\alpha\nu^\beta, \quad \kappa_\tau = b_{\alpha\beta}\tau^\alpha\tau^\beta, \quad \tau = b_{\alpha\beta}\nu^\alpha\tau^\beta, \tag{52}$$

respectively, are the normal curvatures of ω in the directions $\boldsymbol{\nu}$ and $\boldsymbol{\tau}$ and the twist of ω on the $(\boldsymbol{\nu}, \boldsymbol{\tau})$ - axes. Use has also been made of the identities $H = \frac{1}{2}(\kappa_\nu + \kappa_\tau)$ and $K = \kappa_\nu \kappa_\tau - \tau^2$.

Evidently the actual surface tension is given by f_ν. This is the projection onto $\boldsymbol{\nu}$ of the force per unit length transmitted across a curve with unit normal $\boldsymbol{\nu}$. The net force reduces to $\mathbf{f} = \lambda\boldsymbol{\nu}$ $(= -\gamma\boldsymbol{\nu})$ only in the absence of bending effects.

Suppose the membrane is such that the mass density is uniformly distributed on the fixed surface Ω used in the definition of convected coordinates. The presumed existence of such a configuration, even if it is never actually occupied in the course of the motion, constitutes part of the definition of a uniform film. By the chain rule we then have $0 = \partial\rho/\partial\xi^\alpha = \rho_{,\beta}\partial\theta^\beta/\partial\xi^\alpha$. The presumed invertibility of the relation between the fixed and convected coordinates implies that the matrix $(\partial\theta^\beta/\partial\xi^\alpha)$ is non-singular and hence that $\rho_{,\beta} = 0$. The mass-conservation law (41) then yields

$\rho_t = 0$, implying that ρ is a fixed constant on the surface ω, independent of θ^α and t.

If the film is uniform in the sense described, then its response to H and K should be the same at all material points. There is then no explicit coordinate dependence in the specific energy density $\bar{F}$, and hence neither in the areal energy density W. Thus $\partial W/\partial\theta^\alpha_{|\exp}$ vanishes in uniform membranes, implying (cf. (48)) that λ is uniformly distributed on ω. The latter result is modified in the case of films with non-uniform bending properties in which the non-uniformity is induced by a diffusing species, for example (Agrawal and Steigmann 2011). We show below that it is also modified by viscous flow in the surface.

This framework incorporates the well-known shape equation

$$k[\Delta H + 2H(H^2 - K)] - 2\lambda H = p \tag{53}$$

for uniform Helfrich membranes, defined by

$$W = kH^2 + \bar{k}K, \tag{54}$$

in which λ is uniform and k and $\bar{k}$ are the (constant) bending moduli.

3 Effect of intra-membrane viscosity

3.1 Conventional theory of intra-membrane viscosity

Extensions of classical capillarity theory to accommodate intra-surface viscous flow are well known (Scriven 1960; Aris 1989). They entail the addition of a conventional viscous term to the stress $\sigma^{\alpha\beta} = -(\rho^2 F_\rho)a^{\alpha\beta}$, while bending effects are of course suppressed in the classical theory. Typically the viscous stress is assumed to be the 2-D analog of that appearing in the 3-D Navier–Stokes theory. This stress arises in response to straining of the fluid, which is simply the time derivative of the evolving metric in the convected-coordinate description (Aris 1989). Accordingly, in the case of area incompressibility, we have

$$\sigma^{\alpha\beta} = -\gamma a^{\alpha\beta} + \pi^{\alpha\beta}, \quad \text{where} \quad \pi^{\alpha\beta} = \nu a^{\alpha\lambda}a^{\beta\mu}\dot{a}_{\lambda\mu} \tag{55}$$

and ν is the intra-membrane shear viscosity.

This form of the viscous stress is not appropriate for use with the θ^α - parametrization. To rectify this, we substitute (33), obtaining

$$\pi^{\alpha\beta} = \nu\left[a^{\alpha\lambda}a^{\beta\mu}(v_{\mu;\lambda} + v_{\lambda;\mu}) - 2wb^{\alpha\beta}\right], \tag{56}$$

which is equivalent to eqs. (10.23.3, 4) of Aris (1989) in the case of area incompressibility ($a^{\alpha\beta}\dot{a}_{\alpha\beta} = 0$).

3.2 Viscous lipid membranes

We suppose that viscous interaction among lipids arises mainly from their relative motion in the surface ω, while the effect of relative misalignment of neighboring lipids due to bending remains essentially elastic in nature. Accordingly, the required adjustment to the elastic theory of Sect. 2, to account for viscosity, is limited simply to the addition of a viscous stress $\pi^{\alpha\beta}$ of the form (56) to the expression (44)$_1$ for $\sigma^{\alpha\beta}$ and hence also to the expression (44)$_3$ for $N^{\alpha\beta}$ (see (4)). This has the effect of adding the terms

$$\pi^{\beta\alpha}_{;\alpha} \quad \text{and} \quad \pi^{\beta\alpha} b_{\beta\alpha} \tag{57}$$

to the left-hand sides of (46) and (47), respectively. To aid in the reduction of the first term, we recast (56), using (15), as

$$\pi^{\beta\alpha} = \nu \left[a^{\beta\lambda} a^{\alpha\mu} (v_{\mu;\lambda} + v_{\lambda;\mu}) - 4wH a^{\beta\alpha} + 2w\tilde{b}^{\beta\alpha} \right]. \tag{58}$$

From the Mainardi–Codazzi equations (16) and because the metric is covariant-constant (Sokolnikoff 1964), we then obtain

$$\pi^{\beta\alpha}_{;\alpha} = 2\nu (a^{\beta\lambda} a^{\alpha\mu} d_{\lambda\mu;\alpha} - w_{,\alpha} b^{\beta\alpha} - 2wH_{,\alpha} a^{\beta\alpha}), \tag{59}$$

where

$$d_{\lambda\mu} = \frac{1}{2} (v_{\mu;\lambda} + v_{\lambda;\mu}) \tag{60}$$

and

$$d_{\alpha\beta;\mu} = d_{\alpha\beta,\mu} - d_{\beta\lambda} \Gamma^\lambda_{\alpha\mu} - d_{\alpha\lambda} \Gamma^\lambda_{\beta\mu}. \tag{61}$$

Using (13)$_1$ and (17) we also derive

$$\pi^{\beta\alpha} b_{\beta\alpha} = 2\nu \left[b^{\alpha\beta} d_{\alpha\beta} - w(4H^2 - 2K) \right]. \tag{62}$$

The equations of motion for viscous membranes, replacing (46) and (48) respectively, are thus given in the case of *uniform* membranes of Helfrich type (cf. 54)) by

$$\lambda_{,\gamma} - 4\nu w H_{,\gamma} + 2\nu (a^{\alpha\mu} d_{\gamma\mu;\alpha} - w_{,\alpha} b^\alpha_\gamma) = 0, \quad v^\alpha_{;\alpha} - 2wH = 0, \tag{63}$$

and

$$k[\Delta H + 2H(H^2 - K)] - 2\lambda H + 2\nu[b^{\alpha\beta} d_{\alpha\beta} - w(4H^2 - 2K)] = p. \tag{64}$$

The constraint (63)$_2$ may be treated by introducing a function φ such that

$$\Delta \varphi = 2Hw, \quad \text{where} \quad \Delta \varphi = (\sqrt{a})^{-1} (\sqrt{a} a^{\alpha\beta} \varphi_{,\beta})_{,\alpha}, \tag{65}$$

and then defining a vector field ϖ^α such that

$$v^\alpha = a^{\alpha\beta} \varphi_{,\beta} + \varpi^\alpha. \tag{66}$$

From (63)$_2$ and (65)$_1$ we have $\varpi^\alpha_{;\alpha} = 0$ and it follows, for any simply connected patch of the surface, that

$$\varpi^\alpha = \varepsilon^{\alpha\beta} \psi_{,\beta} \tag{67}$$

for some scalar field ψ. However, we make no use of this decomposition in the present work.

3.3 Explicit edge conditions

The edge conditions for lipid membranes with viscosity are given by (3), (4) and (11) in which the viscous stress $\pi^{\alpha\beta}$ is added to the expression (44)$_1$ for $\sigma^{\alpha\beta}$. This results in the addition of the term $\pi^{\beta\alpha} v_\alpha \mathbf{a}_\beta$ to the right-hand side of the formula (49) for the edge force $\mathbf{f}$. The decomposition $\mathbf{a}_\beta = v_\beta \mathbf{v} + \tau_\beta \boldsymbol{\tau}$ facilitates the separation of terms into normal and tangential parts, yielding (44) with

$$\begin{aligned} f_\nu &= W + \lambda - \kappa_\nu M + \pi^{\beta\alpha} v_\beta v_\alpha \\ f_\tau &= \pi^{\beta\alpha} \tau_\beta v_\alpha - \tau M, \end{aligned} \tag{68}$$

while f_n remains unaltered. The expressions (49)$_1$ and (51) for the bending moment and corner forces are also unchanged.

The full set of edge conditions for uniform viscous Helfrich membranes is

$$\begin{aligned} M &= \frac{1}{2} k(\kappa_\nu + \kappa_\tau) + \bar{k}\tau \\ f_\nu &= \frac{1}{4} k(\kappa_\tau^2 - \kappa_\nu^2) - \bar{k}\tau^2 + \lambda + \pi^{\beta\alpha} v_\beta v_\alpha, \\ f_\tau &= \pi^{\beta\alpha} \tau_\beta v_\alpha - \frac{1}{2} k\tau\kappa_\nu - \left(\frac{1}{2}k + \bar{k} \right) \tau\kappa_\tau, \\ f_n &= \bar{k}\tau' - kH_{,\nu} \end{aligned} \tag{69}$$

with corner forces

$$\mathbf{f}_i = \bar{k}[\tau]_i \mathbf{n}. \tag{70}$$

3.4 Dissipation

The discrepancy between the power supplied to the membrane and rate at which energy is stored in it is the *dissipation* D, defined by

$$D = P - \frac{d}{dt} E, \tag{71}$$

and satisfying

$$D \geq 0 \tag{72}$$

in any dissipative process, this effectively serving as a surrogate for the second law of thermodynamics in the present, purely mechanical, setting.

In Steigmann (1999b), it is shown that the equations of mechanical equilibrium for surfaces yield the global balance equation

$$S = P, \tag{73}$$

where S is the *stress power*. In the purely elastic theory, this is given simply by $S = dE/dt$, yielding (7) and $D = 0$. In the presence of both elasticity and viscosity, the appropriate expression for S follows from eq. (2.18) of Steigmann (1999b) and is given by

$$S = \frac{d}{dt}E + \int_\omega \pi^{\beta\alpha}\mathbf{a}_\beta \cdot \dot{\mathbf{a}}_\alpha da, \tag{74}$$

where E is again given by (9). Accordingly,

$$D = (P - S) + \int_\omega \pi^{\beta\alpha}\mathbf{a}_\beta \cdot \dot{\mathbf{a}}_\alpha da, \tag{75}$$

yielding

$$D = \int_\omega \pi^{\beta\alpha}\mathbf{a}_\beta \cdot \dot{\mathbf{a}}_\alpha da \tag{76}$$

in all inertia-less motions of the surface. The argument is easily modified to accommodate inertia. In this case, the equations of motion of the film yield $dK/dt + S = P$ in place of (73), where K is the kinetic energy of the surface. The dissipation is now defined by $D = P - d(E + K)/dt$, while (74) and (76) are unaltered.

The integrand in (73) is $\pi^{\beta\alpha}\mathbf{a}_\beta \cdot \dot{\mathbf{a}}_\alpha = \frac{1}{2}\pi^{\beta\alpha}\dot{a}_{\beta\alpha}$, where

$$\pi^{\beta\alpha}\dot{a}_{\beta\alpha} = \nu a^{\beta\lambda}a^{\alpha\mu}\dot{a}_{\lambda\mu}\dot{a}_{\beta\alpha}. \tag{77}$$

The factor multiplying ν is the squared norm of the surface tensor with covariant components equal to $\dot{a}_{\alpha\beta}$. A positive value of the viscosity thus ensures that any motion involving straining of the surface is strictly dissipative in the sense of the strict inequality in (72). Further, the dissipation vanishes only if the straining vanishes pointwise.

If the loading on the membrane is conservative, then there exists L such that $P = \frac{d}{dt}L$. In this case, we have

$$D = -\frac{d}{dt}(E - L) \tag{78}$$

and hence

$$\frac{d}{dt}(E - L) \leq 0. \tag{79}$$

The *potential energy* $E - L$ then furnishes a Lyapunov function for the motion. In particular, it is minimized by asymptotically stable equilibria at which dissipation ceases and the response is purely elastic.

4 Monge parametrization

To illustrate the use of the theory we adopt the Monge representation

$$\mathbf{r}(\theta^\alpha, t) = \boldsymbol{\theta} + z(\boldsymbol{\theta}, t)\mathbf{k} \tag{80}$$

of ω, where $\boldsymbol{\theta}(\theta^\alpha)$ is position on a plane with unit normal $\mathbf{k}$. The shape of the membrane is then determined by the single function $z(\boldsymbol{\theta}, t)$. For example, the plane may be parametrized globally by a single system of Cartesian coordinates θ^α, in which case

$$\boldsymbol{\theta} = \theta^\alpha \mathbf{e}_\alpha, \tag{81}$$

where $\{\mathbf{e}_\alpha\}$ is an orthonormal basis for the plane. These then furnish a parametrization of ω via (80). We compute

$$\mathbf{r}_t = z_t\mathbf{k}, \quad \mathbf{a}_\alpha = \mathbf{e}_\alpha + z_{,\alpha}\mathbf{k}, \quad a_{\alpha\beta} = \delta_{\alpha\beta} + z_{,\alpha}z_{,\beta}$$
$$\text{and} \quad \mathbf{n} = (\mathbf{k} - \nabla z)/\sqrt{a}, \tag{82}$$

where $\delta_{\alpha\beta}$ is the Kronecker delta, $\nabla z = z_{,\alpha}\mathbf{e}_\alpha$ is the gradient on the plane, and

$$a = \det(a_{\alpha\beta}) = 1 + |\nabla z|^2. \tag{83}$$

Further, the integrand in (8) is

$$\mathbf{n} \cdot \mathbf{r} = (z - \theta^\alpha z_{,\alpha})/\sqrt{a}, \tag{84}$$

the covariant curvature components are

$$b_{\alpha\beta} = \mathbf{n} \cdot \mathbf{a}_{\alpha,\beta} = z_{,\alpha\beta}/\sqrt{a}, \tag{85}$$

and the curvature tensor is $\mathbf{b} = b_{\alpha\beta}\mathbf{a}^\alpha \otimes \mathbf{a}^\beta$, where

$$\mathbf{a}^1 = \frac{1}{a}\{[1 + (z_{,2})^2](\mathbf{e}_1 + z_{,1}\mathbf{k}) - z_{,1}z_{,2}(\mathbf{e}_2 + z_{,2}\mathbf{k})\}, \tag{86}$$

together with a similar formula for $\mathbf{a}^2$, obtained by interchanging the subscripts. These are derived by using $\mathbf{a}^\alpha = a^{\alpha\beta}\mathbf{a}_\beta$ with $(a^{\alpha\beta}) = (a_{\alpha\beta})^{-1}$.

The Christoffel symbols $\Gamma^\lambda_{\alpha\beta} = \mathbf{a}^\lambda \cdot \mathbf{a}_{\alpha,\beta}$ (Sokolnikoff 1964), required in (60) and (61), are

$$\Gamma^\lambda_{\alpha\beta} = z_{,\lambda}z_{,\alpha\beta}/a, \tag{87}$$

and the normal velocity is

$$w = \mathbf{n} \cdot \mathbf{r}_t = z_t/\sqrt{a}. \tag{88}$$

It is straightforward to use these formulas to cast the equations of motion (63), (64) in terms of $z(\theta^\alpha, t)$. However, the resulting system is quite complicated, and so we refrain from exhibiting it explicitly.

4.1 Example

Consider an example in which the membrane flows over a rectangular portion of the plane with sides of length a and b. The kinematic boundary conditions are

$$z = 0, \quad \mathbf{n} = \mathbf{k}. \tag{89}$$

The latter implies that $z_{,\alpha}$ vanish on the boundary and hence so too the normal derivative $z_{,\nu} = \nu^\alpha z_{,\alpha}$. From (88) we have

$$w = 0 \tag{90}$$

Table 1 Boundary conditions used in Figs. 1 and 2

Variable	Boundary 1	Boundary 2	Boundary 3	Boundary 4
z	0	0	0	0
z_x	0	–	–	0
z_y	–	0	0	–
v^α	0	0	0	–
f_v	–	–	–	λ
f_τ	–	–	–	0
λ	–	–	–	10^{-4} pN/nm

on the boundary, while the condition $z_{,v} = 0$ implies that $w_{,v}$ also vanishes there. We conclude that $\mathbf{n} \cdot \mathbf{u}_{,v}$ vanishes on the boundary. Because boundary conditions entail the specification of one or the other member of a pair of power-conjugate variables, it follows from (10) that the transverse shear force f_n and the bending moment M are *not* specified.

No-slip boundary conditions are used on three of the four boundaries for the tangential velocity components, i.e., $v^\alpha = 0$. On the fourth boundary, the traction boundary condition given in (68) is used to relate the velocity gradient to the surface pressure and the membrane shape. On the fourth boundary, we specify the left-hand sides of

$$f_v = \lambda - \frac{1}{4}k(v^\alpha v^\beta z_{,\alpha\beta})^2 + \pi^{\beta\alpha}v_\beta v_\alpha, \tag{91}$$
$$f_\tau = \pi^{\beta\alpha}\tau_\beta v_\alpha,$$

where use has been made of the fact that κ_τ and τ vanish on the boundary; the former following from the fact that the boundary is piecewise straight and the latter from the fact that ∇z vanishes identically on the boundary. In particular, there are no jumps in the twist τ at the corners and thus no corner forces. The boundary conditions are listed in Table 1.

To solve this highly nonlinear coupled system, we use the multiphysics code COMSOL. We study the evolution of membrane shape and flow in response to an applied uniform pressure, p, normal to the surface. It should be noted that when $p = 0$, $z = 0$ is a solution to the shape equation even when the tangential flow is not zero. The model was implemented in the equations-based PDE interface of COMSOL Multiphysics, using the backward difference formula solver for time dependent problems. The equations were solved on a domain of 1,000 nm square with an intramembrane surface viscosity of 10^{-4} pN · s/nm (Hochmuth 1987). The value of bending modulus of the membrane assumed here is 82 pN·nm (Derenyi et al. 2002).

We choose $f_v = \lambda$ on one boundary as shown in Table 1. In Fig. 1, we show the height of the membrane z at the center in response to the applied lateral pressure at different times. The lateral pressure is increased as a ramp function, as shown in Fig. 1a. In response to this increasing pressure,
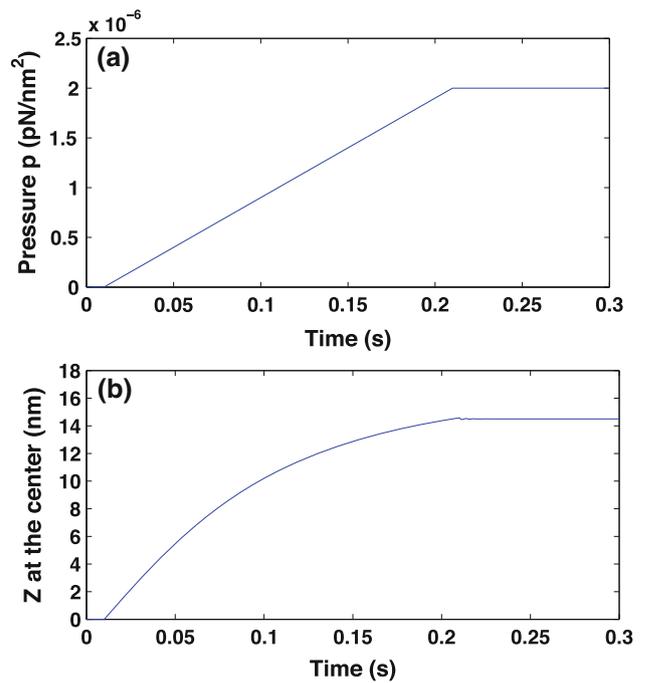

Fig. 1 **a** Lateral pressure as a function of time and **b** the height z at the center of the *square patch*

the height, z, at the center also increases with time and eventually attains a constant value when p is held constant (Fig. 1b), indicating that the system has attained a steady-state configuration.

The tangential velocity field (Fig. 2) also undergoes transient changes to accommodate the flow of lipids required to produce the change in shape. The fluid membrane flows in from the open boundary. At early times, the fluid flow rate is low because the lateral pressure is small and at later times the tangential velocity settles into a steady flow pattern in response to the steady surface pressure. This steady flow (Fig. 2b, c) is maintained even after the lateral pressure attains a constant value because the velocity gradient at that boundary is coupled to the function $z(\theta^\alpha, t)$.

The surface pressure λ varies spatially to accommodate the tangential flow of the lipids into the computational domain. Even though the lipid membrane is homogeneous, the change in the fluid flow pattern results in a change in the surface pressure in a region adjoining the open boundary. At early times, the change is the surface pressure is very small and not discernible. As the shape evolves in response to the lateral pressure p, the surface pressure λ develops a spatial inhomogeneity, which begins to appear around $t = 0.1$ s and intensifies as lateral pressure increases (Table 2).

The coupling between surface shape and surface flow is primarily through the behavior of λ. The choice of f_v at the boundary is also important in determining the flow of lipids and how membrane shape evolves. In this example, the
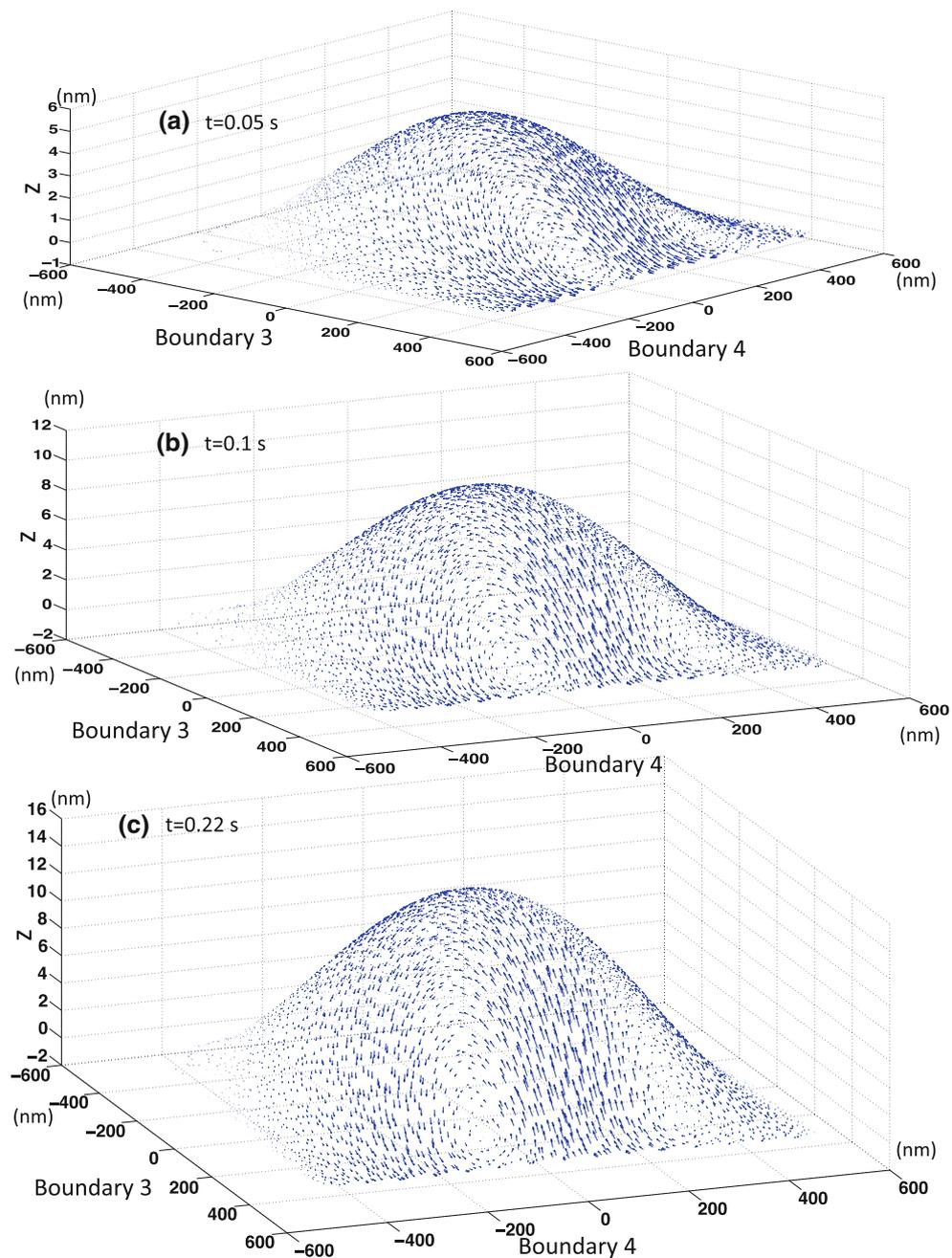
Fig. 2 The tangential velocity vector illustrates the flow of lipids on the surface of the membrane at times **a** $t = 0.05$ s, **b** $t = 0.1$ s, and **c** $t = 0.22$ s. The flow of lipids settles into a steady pattern in accordance with the prescribed boundary conditions and vortices can be seen near the open boundary where traction is assigned

choice $f_v = \lambda$ resulted in a boundary condition wherein the velocity gradient arises in response to surface shape alone and not surface pressure. To further understand how the system responds to different values of f_v, we conducted simulations with the same lateral pressure function but changed the value f_v/λ on boundary 4 (Table 3).

Figure 3 depicts the shape of the membrane along the center of the domain. When f_v/λ is less than unity, the height of the membrane increases. As the value of f_v/λ is increased,

not only does the membrane flatten; it also adopts an asymmetric shape. That is, the maximum height is not at the center of the domain but rather closer to the open boundary. This is due to the role played by λ as an effective surface pressure; larger values at a given pressure correlate roughly with smaller curvature and hence, in this example, smaller height (Table 3).

Another parameter that affects the relationship between surface deformation and flow is the number of boundaries

Table 2 Boundary conditions used in Fig. 4

Variable	Boundary 1	Boundary 2	Boundary 3	Boundary 4
z	0	0	0	0
z_x	0	–	–	0
z_y	–	0	0	–
v^α	0	0	–	–
f_ν	–	–	λ	λ
f_τ	–	–	0	0
λ	–	–	10^{-4} pN/nm	10^{-4} pN/nm

Table 3 Boundary conditions used in Fig. 5

Variable	Boundary 1	Boundary 2	Boundary 3	Boundary 4
z	0	0	0	0
z_x	0	–	–	0
z_y	–	0	0	–
v^α	0	–	–	–
f_ν	–	λ	λ	λ
f_τ	–	0	0	0
λ	–	10^{-4} pN/nm	10^{-4} pN/nm	10^{-4} pN/nm

that allow flow of lipids. In Fig. 4, we show cases where two adjacent boundaries, boundaries 3 and 4, allow flow with the boundary conditions $f_\nu = \lambda$ and $f_\tau = 0$. The shape is symmetric (Fig. 4) but the tangential flow field of lipids is very different from that of the previous example. In Fig. 4,

we observe the lipids entering and leaving the domain from the two adjacent walls. Similarly, when only one boundary is treated as a wall (Fig. 5), the lipids flow in and out of three boundaries (Fig. 5). Even though the shape is similar for these boundary conditions, the flow pattern of lipids on the surface

Fig. 3 A cross section of the domain at the *center line* shows the membrane shape in response to different values of f_ν/λ at the boundary. As f_ν/λ increases, the height decreases and the surface shape becomes increasingly asymmetric for the same value of pressure ($p = 2 \times 10^{-6}$ pN/nm^2)
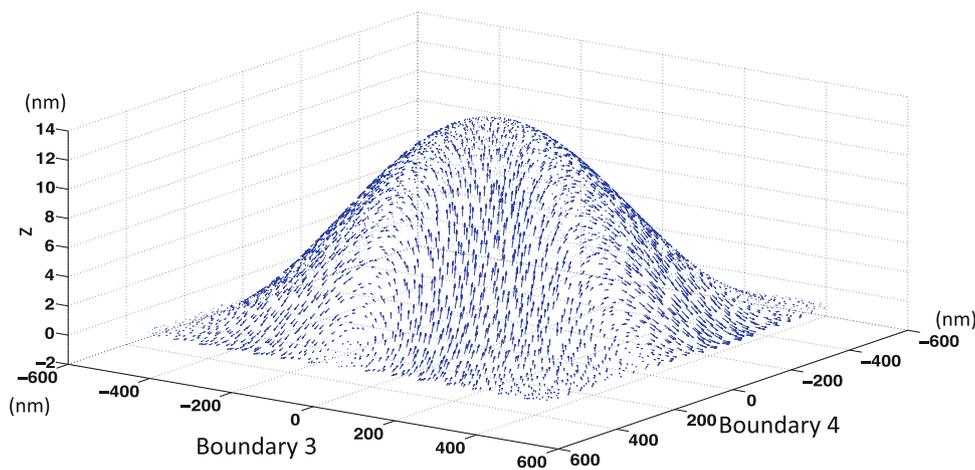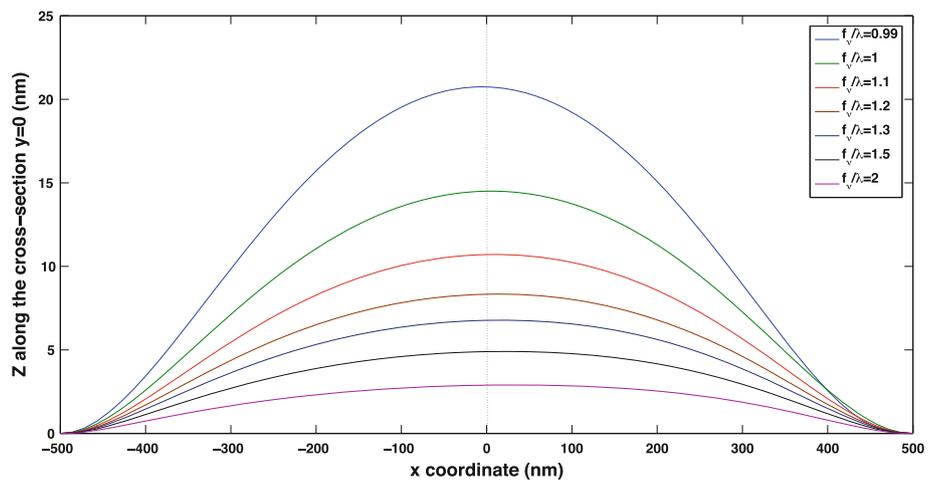
Fig. 4 In this example, two walls, boundaries 3 and 4 have the boundary condition $f_\nu = \lambda$. The characteristics of the membrane are shown at time $t = 0.1$ s, after shape evolution has ceased. The tangential velocity field shows the lipid flow in and out of two boundaries and the presence of vortices near these boundaries
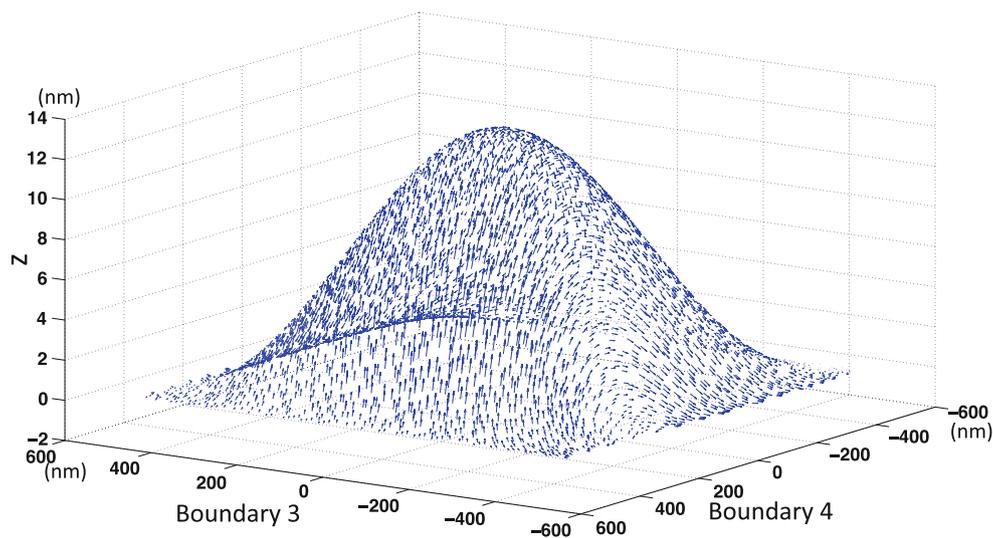
Fig. 5 In this example, three walls, boundaries 1, 3 and 4 have the boundary condition $f_v = \lambda$. The characteristics of the membrane are shown at time $t = 0.1$ s, after shape evolution has ceased. The tangential velocity field shows the lipid flow in and out of three boundaries and the presence of vortices near these boundaries

and the surface pressure pattern are quite different in each of these conditions.

We note that the shape equation (cf. (64)) involves λ algebraically, whereas the remaining equations of the model involve λ only through $\nabla \lambda$. Thus, if the membrane remains flat under the stated boundary conditions, λ affects the flow only through $\nabla \lambda$ and the specification of λ on the boundary is neither necessary nor consistent. In the general case in which membrane shape evolves, we specify a uniform value of λ on the traction part of the boundary and use the fully coupled system for flow and shape to obtain its distribution in the interior. In effect we are solving a one-parameter family of problems, parametrized by the specified boundary value of λ. To find the physiologically relevant value, it would be necessary in principle to assess the predictions obtained using different parameter values against some empirically determined aspect of membrane shape; the appropriate value of the parameter is then given by that which furnishes the best-fit simulation. This exercise is beyond our present scope, however, due to the paucity of available data consistent with our boundary conditions.

5 Conclusions

In this work, we have developed a model for the intra-surface flow of lipids on a bilayer membrane by coupling the elastic model for membrane bending with a model for flow on an evolving curved surface. This model is based entirely on balance laws and constitutive equations. Solutions to representative problems exhibit the coupled interplay between lipid flow and membrane shape. Potential applications to biophysics include the simulation of membrane nanotube formation and the cubic-to-lamellar transition. Our work on these problems is in progress and will be reported elsewhere.

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