Fluid flow and interface motion in gels: A finite-strain theory and its application to a channel flow problem

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ABSTRACT

We present a theory for coupled deformation and liquid permeation in polymer gels, allowing for large strains and rotations in conjunction with sharp interfaces separating regions of high and low polymer volume fraction. The theory is applied to pressure-driven liquid flow through a gel-filled rectangular channel, with the aim of investigating how the elastic and osmotic properties of the gel influence the resulting permeation process. Apart from streamwise flow in the direction of the applied pressure drop, the permeation process may involve transverse flow if the elastic response of the gel allows the normal stress acting on a plane parallel to the channel walls to be affected by the shear stress acting on the same plane. For certain elastic energy densities, including those of neo-Hookean and Mooney type, liquid permeation takes place exclusively in the direction of the applied pressure drop. Otherwise, spanwise permeation driven by the contribution to the normal stress distributed on planes parallel to the channel walls is also present. In contrast to the downstream flow, the transverse flow is transient and is accompanied by changes in composition leading to a nonuniform polymer distribution across the channel and thereby influencing the downstream flow profile. By analyzing the instantaneous response of the gel, we determine the driving force for the transverse flow and show that for strain-stiffening networks the transverse motion of the interstitial liquid is directed inward from the channel walls and that the polymer network motion spreads outward from the center of the channel toward the walls. The opposite occurs for strain-slackening networks. By analyzing the steady-state response, we demonstrate that the applied pressure gradient leads to nonuniform transverse distributions of polymer chains than can be continuous or discontinuous. Discontinuous distributions occur for an effective osmotic response involving attractive interactions and are accompanied by the formation of regions of high and low polymer volume fraction separated by sharp interfaces. Finally, we provide illustrative examples for a gel whose elastic and osmotic responses are modeled by the Gent elastic energy density and the Flory–Huggins mixing energy density, respectively.

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1. Introduction

A polymer gel is a two-component material composed of a cross-linked polymer network and a liquid that fills the interstitial space of the network. These materials display unusual and complex behavior due to the coupling between large network distortions and liquid permeation. This interplay gives rise to very interesting and complex behaviors, including volumetric phase transition. These and other phenomena are discussed elsewhere in the literature. See, for instance, the recent contributions by Dimitriyev et al. (2019), Hennessy et al. (2020), Dušek and Dušková-Smrčková (2020), and Sakai (2020), and the references therein.

In this paper, we present a theory for polymer gels that incorporates the coupling between large strains and rotations, liquid permeation, and interface motion. We begin by introducing the basic balances of the theory, namely standard force balance, network content conservation, liquid content balance, and configurational force balance. Thereafter, we introduce a constitutive theory consistent with a free-energy imbalance which — in a setting involving only deformation, liquid permeation, and interface motion — combines and ensures the satisfaction of the first and second laws of the thermodynamics.

The bulk constitutive theory developed here is suitable for network elasticity and is based on the classical assumption that the solid and liquid components of a polymer gel are incompressible, from which we derive the condition needed to ensure that local changes of volume and liquid content must accommodate each other. The deformation gradient and the liquid content density at a material point are consequently not independent but rather related by a condition that we call '*liquid-induced compressibility*'. Under this condition, a gel behaves like a compressible elastic solid whenever its liquid content is allowed to change and like an incompressible elastic solid otherwise. We treat the liquid-induced compressibility condition as an internal constraint that must be maintained by reactions with null dissipation rate. Accordingly, we find that the stress is constitutively assigned to within a pressure-like contribution that is proportional to the chemical potential of the liquid. Moreover, we find that the chemical potential is given by the sum of two parts, one reactive and other active. An equivalent result was obtained by the present authors (Duda et al., 2010) on the basis of a multiplicative decomposition of the deformation gradient into elastic and liquid-induced parts. A similar kind of internal constraint is provided by the temperature-dependent compressibility constraint in which a continuum body is incompressible under isothermal conditions and compressible otherwise. In this case, as Trapp (1971) and Gurtin and Podio-Guidugli (1973) show, the reactions necessary to maintain such internal constraint are characterized by an arbitrary pressure-like field determining the reactive parts of the stress and entropy. We also introduce the decomposition of the total stress in a gel into network and liquid components. The former component consists of elastic and osmotic contributions and the latter is determined by the interstitial liquid pressure, which is defined as the pressure that the liquid would have if the network was absent and it had the same chemical potential it has in the gel. We also find that the interstitial liquid flow is driven by the gradient of the interstitial liquid pressure.

The interfacial constitutive theory hinges on the assumption that the motion and chemical potential of the liquid are continuous. This leads to a localized version of the free-energy imbalance that involves the Eshelby stress tensor, which in turn can be decomposed into network and liquid contributions. Of these, only the former contribution enters the free-energy imbalance. Instead of discussing the most general constitutive theory compatible with the interfacial free-energy imbalance, we consider a theory appropriate to situations in which interface motion does not involve dissipation. In combination with the configuration force balance, this yields the well-known Maxwell coexistence condition.

The governing equations of the theory are obtained by combining the corresponding basic balances and the constitutive relations. These equations can be formulated in different but equivalent forms depending on the choice of the unknown quantities. As is suitable for isotropic gels, we adopt a formulation in which the basic unknowns determine the network motion, the interstitial liquid pressure, and the interface motion. The liquid pressure provides a two-way coupling between mechanics and permeation since it appears simultaneously in the mechanical force and liquid content balances. The governing equations are presented in both referential and spatial forms, with the latter applied to investigate liquid permeation through a gel-filled channel. Specifically, we study the liquid flow through a gel slab of uniform composition placed between two infinite parallel walls and driven by a pressure drop. We address this problem by adopting an approach similar in spirit to that employed by Coleman et al. (1966) to investigate the plane Poiseuille flow of incompressible liquids. Accordingly, we stipulate that the motion experienced by the gel network has the form of an inhomogeneous shearing in the direction of the applied pressure drop followed by an inhomogeneous stretching in the direction perpendicular to the channel walls. We also allow for the possibility that the state of the gel at a given time may involve the presence of a pair of planar interfaces symmetrically located with respect to the central plane of the channel. As a result, the governing equations of the theory simplify to a system involving time-dependent functions of the space variable along the spanwise direction, except for the pressure that also varies along the streamwise direction, and interface locations that vary only in time. In accord with these equations, liquid permeation occurs in response to inhomogeneities in the shear and normal network stress distributed on the planes parallel to the channel walls and, hence, can take place along two directions only, one parallel and other perpendicular to the direction of the imposed pressure drop. Contrary to the parallel component of the flow, which is proportional to the applied pressure drop and, hence, is always present, any transverse flow that arises has a transient nature and depends sensitively on the constitutive response of the network. Moreover, it must be accompanied by network motion to ensure the satisfaction of the liquid-induced compressibility constraint, thereby triggering changes in the composition of gel until steady state is reached and the transverse flow ceases. Using these equations, we address aspects of the instantaneous and steady-state responses of the gel to a suddenly applied pressure drop.

We investigate the instantaneous response with the purpose of establishing the conditions under which transverse liquid flow tends to occur, as described by the component of the pressure gradient in the direction normal to the channel walls. Since there is insufficient time for liquid permeation to occur in this regime, we show that the gel behaves like an incompressible elastic body

undergoing an inhomogeneous shear deformation compatible with the distribution of the amount of shear across the channel. It transpires that the instantaneous response of the gel is determined by its elastic properties and involves simple shear only. We obtain the amount of shear by the solving, at each shearing plane, the constitutive equation giving the shear stress, which is known, as a function of the amount of shear. After introducing sufficient conditions for the existence and uniqueness of solutions to this problem, we conclude that whenever the normal network stress increases with the amount of shear the interstitial liquid is directed from the walls toward the center of the channel. The opposite occurs when the normal network stress decreases with the amount of shear. If the shear stress is unaffected by the amount of shear, there is no tendency for the liquid to flow. This is the case for neo-Hookean and Mooney models for elastic networks.

Regarding the steady-state response, where both elastic and osmotic properties are active, our purpose is to describe the profile reached by the only nonvanishing component of the liquid flux, namely the component in the direction of, and proportional to, the applied pressure drop. Bearing in mind that the proportionality factor is determined by the liquid mobility response, which in turn is an increasing function of the stretch in the direction perpendicular to the channel walls, the problem amounts to obtaining the profile for that stretch. Assuming that the constitutive relation giving the shear stress as a function of the stretch and amount of shear can be inverted for the amount of shear, we arrive at a set of equations to be solved for the stretch distribution. These equations include the extra interfacial condition equivalent to the Maxwell equal area rule expressed in terms of the partial Legendre transform of the free-energy density with respect to the shear stress. We establish a necessary condition for a solution to present interfaces across which the stretch exhibits a jump discontinuity. This condition hinges upon the nonmonotonicity of the constitutive expression that determines the normal network stress. This leads us to address two cases separately, with and without interfaces, according to whether the profile for the stretch is or is not smooth. We also provide illustrative examples by using the Gent and Flory–Huggins models for elastic and mixing contributions to the free-energy density.

The problem of liquid flow through a gel-filled channel under steady-state conditions was investigated by Cogan and Keener (2005) using a description of the gel system distinct from ours. In fact, treating a gel as a mixture of two immiscible components, namely an elastic polymer network and a Newtonian fluid, Cogan & Keener used mixture theory to derive a set of equations that accounted for network deformation, fluid motion, and polymer diffusion within the gel. In their investigation, interfaces emerged by analyzing the mentioned equations in the singular limit as the polymer "diffusion coefficient" tends to zero. In contrast, we conceive a gel as a single continuum body consisting of an elastic polymer network hosting an interstitial liquid and used the framework of modern continuum mechanics to derive a set of equations accounting for network deformation, fluid motion, and interface motion within the gel. To analyze the short-term transition response, we apply methods similar to those used by Zhang and Rajagopal (1992) and Saccomandi (2004) in studies of how an applied pressure drop may induce an inhomogeneous state of shear in an incompressible and isotropic elastic slab. Given the prominence of the pressure-induced channel flow problem for understanding the behavior of liquids, we believe that this may also apply to gels. In a gel-filled channel, the pressure drop drives the flow of the interstitial liquid through the deformable polymer network, which otherwise adheres to the channel walls. This analogous problem has been addressed within the theory of porous media by several authors. See, for instance, Levick (1987), Yang et al. (2002), and Guy et al. (2011), whose works do not consider the deformability of the polymer network. As Nakagaki and Guy (2008) and Guy et al. (2011) observe, liquid flow through a gel-filled channel is a subject of considerable interest for various biological materials. See also the review by Mogilner and Manhart (2018). More recently, Loessberg-Zahl et al. (2019) used microfluidic devices to study flow through gels.

Recognizing the unique behavior of polymer gels stemming from the interplay of large deformation, fluid permeation, and volume transition, we believe that the present work may find applications in many fields. One such field is biology in view of two concurrent observations:

- the intracellular and extracellular spaces in living systems are generally occupied by gel-like materials such as the cytoplasm and the interstitium;
- fluid flows are important in a myriad of cellular processes, including the transport of ions and proteins, differentiation, proliferation, and gene expression.

See, for instance, the reviews of Mogre et al. (2020) and Wiig and Swartz (2012). Of course, these potential applications may require the extension of our framework to account for anisotropy and time-dependence of the network response, the multicomponent nature of the network and interstitial fluid, and other salient factors.

The remainder of this paper is organized as follows. After some essential preliminary material outlined in Section 2, the basic laws of the theory are formulated in Section 3 and the constitutive theory is presented Section 4. The referential and spatial versions of the final governing equations are set forth in Section 5 and Section 6, respectively. The problem of pressure-driven liquid flow through a gel-filled channel is formulated and solved in Section 7. A cursory discussion regarding the extension of the present work to account for network anisotropy and different flow configurations is presented in Section 8. Concluding remarks are contained in Section 9.

2. Preliminaries

We consider a material body $\mathcal{B}$ composed by a polymer gel and identified with the region it occupies in a fixed reference configuration. At time t, the body $\mathcal{B}$ is divided into complementary subregions $\mathcal{B}^-(t)$ and $\mathcal{B}^+(t)$ by a sharp interface $\mathcal{S}(t)$. At each point $\boldsymbol{X}$ of $\mathcal{S}(t)$, we write $U(\boldsymbol{X}, t)$ for the scalar normal-velocity of $\mathcal{S}(t)$ in the direction of the unit normal $\mathrm{n}(\boldsymbol{X}, t)$ directed outward from $\mathcal{B}^-(t)$.

To describe the shape and composition of $\mathcal{B}$ at time t, we introduce the fields y, C_p, and C, which assign to each material point X in $\mathcal{B}\backslash S(t)$ the corresponding spatial location $x = y(X,t)$, the polymer number density $C_p(X,t)$, and the liquid number density $C(X,t)$, with both densities being measured per unit reference volume. These fields are smooth away from and up to the interface $S(t)$ from either side. Whereas the field y is assumed to be continuous across the interface, the fields C_p and C may display jump discontinuities denoted by $[\![C_p]\!]$ and $[\![C]\!]$, where, here and henceforth, $[\![\varphi]\!]$ designates the jump in the field φ across the interface, namely

$$[\![\varphi]\!] = \overset{+}{\varphi} - \overset{-}{\varphi}, \tag{1}$$

with $\overset{-}{\varphi}$ and $\overset{+}{\varphi}$ denoting the respective interfacial limits of φ on the sides of S out of and into which n points. For later reference, we also introduce the average of the interfacial limits of φ, namely

$$\langle\!\langle \varphi \rangle\!\rangle = \tfrac{1}{2}(\overset{+}{\varphi} + \overset{-}{\varphi}). \tag{2}$$

Writing $F = \nabla y$ for the deformation gradient, we recall that the continuity of y across the interface yields the compatibility conditions (see, for instance, Gurtin et al., 2010 and Šilhavỳ, 1997)

$$[\![\dot{y}]\!] + U[\![F]\!]\mathrm{n} = 0, \qquad [\![F]\!](I - \mathrm{n} \otimes \mathrm{n}) = \mathbf{0}, \qquad [\![F^C]\!]\mathrm{n} = \mathbf{0}. \tag{3}$$

where a superposed dot denotes material time differentiation and F^C is the cofactor of F:

$$F^C = (\det F)F^{-\top}. \tag{4}$$

The composition of a polymer gel is often described in terms of spatial fields ϕ and φ representing, respectively, the network and liquid volume fractions. The relation among the variables ϕ, φ, C_p and C will be discussed later. Throughout this paper, the words liquid, fluid, and solvent will be treated as synonymous.

3. Balance laws

Whereas changes of shape and composition sustained by $\mathcal{B}$ must be consistent with laws of balance for standard forces, network content, and liquid content, interface motion must be consistent with an extra condition — often called the configurational force balance — which provides a means for incorporating the microphysical processes that govern growth of one phase at the expense of another. Together with the free-energy imbalance, these laws comprise the kinetic foundation of the theory and are postulated for arbitrary parts of $\mathcal{B}$. A part $\mathcal{P}$ of $\mathcal{B}$ is a bounded subregion of $\mathcal{B}$ with outward unit normal to $\partial\mathcal{P}$ denoted by m. For any such part $\mathcal{P}$ of $\mathcal{B}$, the standard force balance reads

$$\int_{\partial\mathcal{P}} Sm \, \mathrm{d}A + \int_{\mathcal{P}} b_0 \, \mathrm{d}V = \mathbf{0}, \tag{5}$$

where S is the first Piola stress and b_0 is the external body force density, while the network and liquid content balances read

$$\frac{\mathrm{d}}{\mathrm{d}t}\int_{\mathcal{P}} C_p \, \mathrm{d}V = 0 \qquad \text{and} \qquad \frac{\mathrm{d}}{\mathrm{d}t}\int_{\mathcal{P}} C \, \mathrm{d}V = -\int_{\partial\mathcal{P}} J \cdot m \, \mathrm{d}A + \int_{\mathcal{P}} Q \, \mathrm{d}V, \tag{6}$$

where J is the Piola liquid flux, measured relative to the network, and Q is the external supply of liquid content. Furthermore, the configurational force balance for a part $\mathcal{P}$ of $\mathcal{B}$ reads

$$\int_{\mathcal{P}\cap S(t)} (f_i + f_e) \, \mathrm{d}A = 0, \tag{7}$$

where f_i and f_e are interfacial measures of internal and external configurational force densities. Finally, the law of free-energy imbalance for $\mathcal{P}$ reads

$$\frac{\mathrm{d}}{\mathrm{d}t}\int_{\mathcal{P}} \psi \, \mathrm{d}V \leq \mathcal{W}_s(\mathcal{P}) + \mathcal{W}_c(\mathcal{P}) + \mathcal{T}(\mathcal{P}), \tag{8}$$

where ψ is the free-energy density, $\mathcal{W}_s$ and $\mathcal{W}_c$ denote the standard and configurational contributions to the power expended on $\mathcal{P}$ by external agencies, as given by

$$\mathcal{W}_s(\mathcal{P}) = \int_{\partial\mathcal{P}} Sm \cdot \dot{y} \, \mathrm{d}A + \int_{\mathcal{P}} b_0 \cdot \dot{y} \, \mathrm{d}V \qquad \text{and} \qquad \mathcal{W}_c(\mathcal{P}) = \int_{\mathcal{P}\cap S(t)} f_e U \, \mathrm{d}A, \tag{9}$$

and $\mathcal{T}$ denotes the energy flow that accompanies liquid transport, as given by

$$\mathcal{T}(\mathcal{P}) = -\int_{\partial\mathcal{P}} \mu J \cdot m \, \mathrm{d}A + \int_{\mathcal{P}} \mu Q \, \mathrm{d}V, \tag{10}$$

where μ is the chemical potential of the interstitial liquid. As a reminder, we recall that the quantity U appearing in $\mathcal{W}_c$ is the scalar normal velocity of the interface S.

The following remarks are in order:

- As is evident from (5), we assume from the outset that inertia is negligible. This is consistent with most applications involving polymer gels.

- The interfacial configurational force densities are power-conjugate to the normal interface velocity and the balance involving those quantities is postulated for a body part that contains an interface. In Gurtin's (Gurtin, 1999) treatment of configurational forces (which is extended by Fried and Gurtin (1999) for problems involving species migration), the configurational force system involves additional quantities, notably the configurational stress, and the configurational force balance is postulated for migrating control volumes that contain the interface.
- The basic balances can be derived as consequences of the principle of virtual power following arguments used previously by the present authors (Duda et al., 2010). To streamline the presentation, we opt here for the standard approach (see, for instance, Gurtin et al., 2010) and postulate the basic laws in integral forms.

3.1. Bulk versions of the basic laws

The basic laws (5)–(8) are now localized to an arbitrary point which, at a given time t, is away from the interface $S(t)$. This can be done by using standard arguments for parts $\mathcal{P}$ consistent with $\mathcal{P} \cap S(t) = \emptyset$. In particular, the standard force balance (5), network content balance $(6)_1$, liquid content balance $(6)_2$ localize to

$$\text{Div}\boldsymbol{S} + \boldsymbol{b}_0 = \boldsymbol{0}, \qquad \dot{C}_p = 0, \qquad \text{and} \qquad \dot{C} = -\text{Div}\boldsymbol{J} + Q, \tag{11}$$

where "Div" is the referential divergence operator, while the free-energy imbalance (8) localizes, with reference to $(11)_{1,3}$, to

$$\dot{\psi} - \boldsymbol{S} \cdot \dot{\boldsymbol{F}} - \mu\dot{C} + \boldsymbol{J} \cdot \boldsymbol{\xi} \leq 0, \tag{12}$$

where the symbol $\boldsymbol{\xi}$ has been introduced to denote the gradient of the chemical potential:

$$\boldsymbol{\xi} = \nabla\mu. \tag{13}$$

3.2. Interfacial versions of the basic laws

The basic laws are now localized to an arbitrary point which, at a given time t, is located on the interface $S(t)$. This can be done by using standard arguments for parts $\mathcal{P}$ such that $\mathcal{P} \cap S(t) \neq \emptyset$. The interested reader is referred to Gurtin et al. (2010) and Šilhavỳ (1997) for details.

In particular, while the standard force balance, network content balance, and liquid content balance localize to

$$[\![\boldsymbol{S}]\!]\mathbb{n} = \boldsymbol{0}, \qquad [\![C_p]\!]U = 0, \qquad \text{and} \qquad [\![C]\!]U = [\![\boldsymbol{J}]\!] \cdot \mathbb{n}, \tag{14}$$

the configurational force balance (7) localizes to

$$f_i + f_e = 0. \tag{15}$$

In view of $(14)_2$, we henceforth take C_p to be continuous across the interface S.

Furthermore, the free-energy imbalance (8) localizes to

$$-([\![\psi]\!] + f_e)U - [\![\boldsymbol{S}^\top\dot{\boldsymbol{y}}]\!] \cdot \mathbb{n} + [\![\mu\boldsymbol{J}]\!] \cdot \mathbb{n} \leq 0. \tag{16}$$

Invoking $(3)_1$, $(14)_{2,3}$, and (15) and introducing the configurational stress (or Eshelby) tensor

$$\boldsymbol{\Sigma} = (\psi - \mu C)\boldsymbol{I} - \boldsymbol{F}^\top\boldsymbol{S}, \tag{17}$$

we arrive at a convenient alternative to (16):

$$-(\mathbb{n} \cdot [\![\boldsymbol{\Sigma}]\!]\mathbb{n} + [\![\mu]\!]\langle\!\langle C\rangle\!\rangle - f_i)U + [\![\mu]\!]\langle\!\langle\boldsymbol{J}\rangle\!\rangle \cdot \mathbb{n} \leq 0. \tag{18}$$

3.3. Spatial versions of the bulk and interfacial balances

For later reference, we recall that the spatial counterparts of the local balances (11) can be written as

$$\text{div}\boldsymbol{T} + \boldsymbol{b} = \boldsymbol{0}, \qquad \frac{\partial c_p}{\partial t} + \text{div}(c_p\boldsymbol{v}) = 0, \qquad \text{and} \qquad \frac{\partial c}{\partial t} + \text{div}(c\boldsymbol{v}) = -\text{div}\boldsymbol{J} + q, \tag{19}$$

where "div" is the spatial divergence operator and $\boldsymbol{T}$, $\boldsymbol{b}$, c_p, $\boldsymbol{v}$, c, $\boldsymbol{J}$, and q are spatial fields representing, respectively, the Cauchy stress, spatial body force density, spatial polymer density, spatial velocity, spatial liquid content density, Cauchy liquid flux, and external supply of liquid. Furthermore, the following relations between referential and spatial quantities hold

$$\boldsymbol{S} = \boldsymbol{T}\boldsymbol{F}^c, \qquad \boldsymbol{b}_0 = (\det\boldsymbol{F})\boldsymbol{b}, \qquad \boldsymbol{v} = \dot{\boldsymbol{y}}, \qquad C_p = (\det\boldsymbol{F})c_p, \qquad C = (\det\boldsymbol{F})c, \qquad \boldsymbol{J} = (\boldsymbol{F}^c)^\top\boldsymbol{J}, \qquad Q = (\det\boldsymbol{F})q. \tag{20}$$

On using s to denote the spatial interface, defined for each time t by

$$s(t) = \{\boldsymbol{y}(\boldsymbol{X}, t), \boldsymbol{X} \in S(t)\}, \tag{21}$$

the spatial counterparts of (14) for an arbitrary point on s can, moreover, be written as

$$[\![\boldsymbol{T}]\!]\mathbb{m} = \boldsymbol{0}, \qquad [\![c_p]\!]u = [\![c_p\boldsymbol{v}]\!] \cdot \mathbb{m}, \qquad \text{and} \qquad [\![c]\!]u = [\![c\boldsymbol{v}]\!] \cdot \mathbb{m} + [\![\boldsymbol{J}]\!] \cdot \mathbb{m}, \tag{22}$$

where m and u, the spatial counterparts of the unit normal n and scalar normal velocity U of the referential interface S, are given by

$$\mathrm{m} = \frac{\langle\!\langle \boldsymbol{F}^{\mathrm{C}} \rangle\!\rangle \mathrm{n}}{|\langle\!\langle \boldsymbol{F}^{\mathrm{C}} \rangle\!\rangle \mathrm{n}|} \qquad \text{and} \qquad u - \langle\!\langle \boldsymbol{v} \rangle\!\rangle \cdot \mathrm{m} = U \langle\!\langle |\boldsymbol{F}^{\top} \mathrm{m}| \rangle\!\rangle. \tag{23}$$

The condition $(3)_3$ requiring that $\boldsymbol{F}^{\mathrm{C}} \mathrm{n}$ be continuous across the interface has been used to express m in terms of n and $u - \langle\!\langle \boldsymbol{v} \rangle\!\rangle \cdot \mathrm{m}$ in terms of U. To see this, it suffices to show that the standard relation

$$\mathrm{m} = \frac{\overset{\pm}{\boldsymbol{F}^{\mathrm{C}}} \mathrm{n}}{|\overset{\pm}{\boldsymbol{F}^{\mathrm{C}}} \mathrm{n}|} \tag{24}$$

between n and m is equivalent to $(23)_1$. Specifically, as a consequence of $(3)_3$, we have that

$$\overset{+}{\boldsymbol{F}^{\mathrm{C}}} \mathrm{n} = \overset{-}{\boldsymbol{F}^{\mathrm{C}}} \mathrm{n} = \overset{\pm}{\boldsymbol{F}^{\mathrm{C}}} \mathrm{n} = \langle\!\langle \boldsymbol{F}^{\mathrm{C}} \rangle\!\rangle \mathrm{n} \tag{25}$$

and, thus, that

$$|\overset{+}{\boldsymbol{F}^{\mathrm{C}}} \mathrm{n}| = |\overset{-}{\boldsymbol{F}^{\mathrm{C}}} \mathrm{n}| = |\overset{\pm}{\boldsymbol{F}^{\mathrm{C}}} \mathrm{n}| = |\langle\!\langle \boldsymbol{F}^{\mathrm{C}} \rangle\!\rangle \mathrm{n}|. \tag{26}$$

Hence the equivalence between $(23)_1$ and (24) follows. On the other hand, $(23)_2$ is a direct consequence of the standard relation

$$u = U |\overset{\pm}{\boldsymbol{F}^{\top}} \mathrm{m}| + \overset{\pm}{\boldsymbol{v}} \cdot \mathrm{m} \tag{27}$$

between u and U. The interested reader is again referred to Gurtin et al. (2010) and Šilhavỳ (1997) for a detailed presentation of the arguments involved in the foregoing derivations.

4. Constitutive theory

The basic laws introduced previously hold for any material body composed of two components, one solid-like that is conserved and other liquid-like that is free to move through solid component. We next introduce specific constitutive assumptions often used to describe polymer gels, in which the solid- and liquid-like components are given by the polymer network and interstitial liquid, respectively. We present a constitutive theory compatible with the local consequences (12) and (18) of the free-energy imbalance (8), considering first bulk quantities and next interfacial quantities.

4.1. Bulk constitutive theory

Bearing in mind that our aim is to develop a coupled theory between shape and composition changes appropriate for gels whose polymer network is elastic, we assume that the constitutive response at a given material point of a gel is determined by the deformation gradient, liquid content density, and liquid flux at the same material point. We also adopt the classical assumption that both components of a gel are incompressible and on the basis this assumption derive the standard condition stating that local volume variation must be accompanied by local liquid-content variation and, hence, that the gel exhibits liquid-induced compressibility. In other words, the gel behaves like a compressible solid whenever its liquid content is allowed to change and like an incompressible solid otherwise. The deformation gradient and the liquid content density at a material point are therefore not independent but rather are related by a requirement that we refer to as the liquid-induced compressibility condition. We treat the said condition as an internal constraint that must be maintained by reactions whose associated dissipation rate is null. Accordingly, we show that the stress is constitutively assigned to within a pressure-like contribution determined by the chemical potential of the interstitial liquid, which in turn is given by the sum of two parts, one reactive and other active. Whereas the reactive contribution to the chemical potential is arbitrary, a constitutive equation for the active part must be prescribed and two such prescriptions are discussed: one for which the active part of the chemical potential is identically zero and other for which it is prescribed in manner similar to the unconstrained case, namely as if the deformation gradient and liquid content density were independent variables. An equivalent result was obtained by the present authors (Duda et al., 2010) using the multiplicative decomposition of the deformation gradient into elastic and liquid-induced components. A similar kind of internal constraint is provided by the temperature-dependent compressibility constraint in which a continuum body is incompressible under isothermal conditions and compressible otherwise. As shown by Trapp (1971) and Gurtin and Podio-Guidugli (1973), the reactions necessary to maintain such internal constraint are under such conditions characterized by an arbitrary pressure-like field determining the reactive components of the Cauchy stress and the specific entropy.

4.1.1. Internal constraint

Given a bulk part $\mathcal{P}$ of $\mathcal{B}$, we write $\mathrm{vol}(\mathcal{P}, t)$ for the volume of its deformed image $\mathcal{P}_t = \boldsymbol{y}(\mathcal{P}, t)$ at time t:

$$\mathrm{vol}(\mathcal{P}, t) = \int_{\mathcal{P}_t} \mathrm{d}v, \tag{28}$$

We now introduce constitutive assumptions concerning to the composition of $\mathcal{B}$:

1. The volume $\text{vol}(\mathcal{P}, t)$ occupied by an arbitrary bulk part $\mathcal{P}$ at time is equal to the sum

$$\text{vol}(\mathcal{P}, t) = \text{vol}_\text{p}(\mathcal{P}, t) + \text{vol}_\text{f}(\mathcal{P}, t), \tag{29}$$

of the volumes

$$\text{vol}_\text{p}(\mathcal{P}, t) = \int_{\mathcal{P}_t} \phi(\boldsymbol{x}, t)\,\mathrm{d}v \quad \text{and} \quad \text{vol}_\text{f}(\mathcal{P}, t) = \int_{\mathcal{P}_t} \varphi(\boldsymbol{x}, t)\,\mathrm{d}v \tag{30}$$

occupied by the polymer network and the interstitial liquid, respectively.

2. The contributions $\text{vol}_\text{p}(\mathcal{P}, t)$ and $\text{vol}_\text{f}(\mathcal{P}, t)$ to $\text{vol}(\mathcal{P}, t)$ can be written as

$$\text{vol}_\text{p}(\mathcal{P}, t) = \int_{\mathcal{P}_t} v_p c_p(\boldsymbol{x}, t)\,\mathrm{d}v \quad \text{and} \quad \text{vol}_\text{f}(\mathcal{P}, t) = \int_{\mathcal{P}_t} v c(\boldsymbol{x}, t)\,\mathrm{d}v, \tag{31}$$

where $v_p > 0$ and $v > 0$ are given constants.

Since $\mathcal{P}$ is an arbitrary part of B, we see from (29)–(31) that

$$\phi + \varphi = 1, \qquad \phi = v_p c_p, \qquad \text{and} \qquad \varphi = vc. \tag{32}$$

The first of (32), which is often called the saturation condition, expresses the assumption that the network is maximally impregnated by liquid.

We assume hereinafter that $B = \boldsymbol{y}(B, 0)$, namely that the reference configuration is the configuration occupied by the body at the instant $t = 0$. This being the case, we find from (11)$_2$, (20)$_4$, and (32)$_2$ that ϕ is determined by ϕ_0 and $\det \boldsymbol{F}$ by

$$\phi = \frac{\phi_0}{\det \boldsymbol{F}}, \tag{33}$$

where $\phi_0(\boldsymbol{X}) := v_p C_p(\boldsymbol{X}, 0)$ is the polymer volume fraction in the reference configuration. On the other hand, we infer from (20)$_5$, (32)$_3$, and (33) that the alternative condition

$$\det \boldsymbol{F} = \phi_0 + vC \tag{34}$$

holds. On combining (33) and (34), we obtain a relation between the compositional variables ϕ and C:

$$\phi = \frac{\phi_0}{\phi_0 + vC}. \tag{35}$$

The relation (35) allows us to choose either C or ϕ to measure composition according to convenience. Due to our decision to identify the reference configuration with the initial configuration, evaluating (35) at $t = 0$ yields

$$\phi_0 + vC_0 = 1, \tag{36}$$

where C_0 is the liquid content density at time $t = 0$; hence, (34) can be written as

$$\det \boldsymbol{F} = 1 + v(C - C_0). \tag{37}$$

Under the constitutive assumptions adopted thus far, (33), (34), and (37) are equivalent to the network content conservation (11)$_2$. Further, (33), (34), and (37) provide a useful connection between the local volume change as measured by $\det \boldsymbol{F}$ to the composition as measured by φ and C. In particular, (34) shows that local volume changes undergone by a gel are due to changes in the liquid content only. We treat (34) as an internal constraint, which we call the mechanical incompressibility constraint or the 'liquid-induced compressibility condition'.

4.1.2. Constitutive variables and dissipation principle

Using the consequence

$$\boldsymbol{F}^\text{C} \cdot \dot{\boldsymbol{F}} = v\dot{C} \tag{38}$$

of computing the material time derivative of the mechanical incompressibility constraint (34) in (12), we find that the local free-energy inequality can be expressed as

$$\dot{\psi} - \boldsymbol{P} \cdot \dot{\boldsymbol{F}} + \boldsymbol{\xi} \cdot \boldsymbol{J} \leq 0, \tag{39}$$

where

$$\boldsymbol{P} = \boldsymbol{S} + \frac{\mu}{v} \boldsymbol{F}^\text{C} \tag{40}$$

is the effective measure of Piola stress that is power-conjugate to the rate $\dot{\boldsymbol{F}}$ at which the deformation gradient $\boldsymbol{F}$ changes with respect to time.

Guided by the inequality (39), we select $(\boldsymbol{F}, \boldsymbol{J})$ and $(\psi, \boldsymbol{P}, \boldsymbol{\xi})$ as lists of independent and dependent constitutive variables, respectively, and prescribe constitutive equations for ψ, $\boldsymbol{\xi}$, and $\boldsymbol{P}$ through

$$\psi = \bar{\psi}(\boldsymbol{F}, \boldsymbol{J}), \qquad \boldsymbol{P} = \bar{\boldsymbol{P}}(\boldsymbol{F}, \boldsymbol{J}), \qquad \text{and} \qquad \boldsymbol{\xi} = \bar{\boldsymbol{\xi}}(\boldsymbol{F}, \boldsymbol{J}), \tag{41}$$

with $\bar{\psi}$, $\bar{P}$, and $\bar{\xi}$ being the constitutive response functions for ψ, P, and ξ. These functions must be consistent with the free-energy inequality (39). Following the Coleman–Noll procedure, it can thus be shown that the following conditions must hold:

- The constitutive response functions $\bar{\psi}$ and $\bar{P}$ are independent of J, and satisfy

$$\frac{\partial \bar{\psi}(F, J)}{\partial J} = 0 \qquad \text{and} \qquad \bar{P}(F) = \frac{\partial \bar{\psi}(F)}{\partial F}. \tag{42}$$

- The constitutive response function $\bar{\xi}$ must obey the internal dissipation inequality

$$\bar{\xi}(F, J) \cdot J \leq 0 \tag{43}$$

for all choices of F and J.

Notice that according to (40), (41)$_2$ and (42)$_2$, the Piola stress S is given by

$$S = \frac{\partial \bar{\psi}(F)}{\partial F} - \frac{\mu}{v} F^C \tag{44}$$

and is, thus, the sum of two parts, the effective stress P that arises from the free-energy of the gel and a contribution involving the pressure-like factor μ/v that arises from the mechanical incompressibility constraint (34).

4.1.3. Active and reactive components of S and μ

The foregoing development does not recognize the presence of the reactions needed to maintain the mechanical incompressibility constraint (34). To address this issue, we adopt a treatment that parallels the one provided by Capriz (1989) within the context of continua with microstructure.

Specifically we assume that ψ, S, μ, and ξ admit additive decompositions

$$\psi = \psi_r + \psi_a, \qquad S = S_r + S_a, \qquad \mu = \mu_r + \mu_a, \qquad \xi = \xi_r + \xi_a, \tag{45}$$

into reactions ψ_r, S_r, μ_r, and ξ_r and actions ψ_a, S_a, μ_a, and ξ_a. Using (45)$_{2,3}$ in (40), we see that the effective stress P also admits such a decomposition, namely

$$P = P_r + P_a, \tag{46}$$

with P_r and P_a being given by

$$P_r = S_r + \frac{\mu_r}{v} F^C \qquad \text{and} \qquad P_a = S_a + \frac{\mu_a}{v} F^C. \tag{47}$$

Whereas the reactions ψ_r, S_r, μ_r, and ξ_r must be powerless in that sense that they obey

$$\dot{\psi}_r - P_r \cdot \dot{F} + \xi_r \cdot J = 0 \tag{48}$$

for all choices of $\dot{F}$ and J, the actions ψ_a, S_a, μ_a, and ξ_a can serve as dependent constitutive variables to which constitutive relations must be assigned. Moreover, the actions must satisfy the reduced inequality

$$\dot{\psi}_a - P_a \cdot \dot{F} + \xi_a \cdot J \leq 0 \tag{49}$$

that follows from (39), (45), and (48).

We now explore the consequences of (48). Taking into account that $\dot{F}$ and J can be varied independently, we see from (48) that

$$\dot{\psi}_r \equiv 0, \qquad P_r \equiv 0, \qquad \xi_r \equiv 0. \tag{50}$$

With the recognition that the free-energy density ψ can be determined only up to an additive constant and dropping the subscript "a" on ψ_a, P_a, and ξ_a, we deduce that (39) and (49) are equivalent and, hence, that (42) and (43) hold. Furthermore, we see from (47)$_1$ and (50)$_2$ that the reactive components S_r and μ_r of the Piola stress S and the chemical potential μ are given by

$$S_r = -\varpi F^C \qquad \text{and} \qquad \mu_r = v\varpi, \tag{51}$$

where ϖ is a scalar field that serves to ensure that the constraint (34) of mechanical incompressibility is maintained.

Referring to (42)$_2$, (46) and (47)$_2$, we find that

$$S_a + \frac{\mu_a}{v} F^C = \frac{\partial \bar{\psi}(F)}{\partial F} \tag{52}$$

and thus conclude that the constitutive equations determining S_a and μ_a must be assigned in conformity with (52). Two possible choices, along with the corresponding representations for S and μ, are discussed next.

4.1.4. Representations for S and μ

We are free to choose any constitutive relations for S_a and μ_a that do not violate (52). Here, we describe two simple alternatives:

1. Consistent with (52), our first choice for S_a and μ_a is

$$S_a = \frac{\partial \bar{\psi}(F)}{\partial F} \qquad \text{and} \qquad \mu_a \equiv 0. \tag{53}$$

Combining $(45)_2$, (51), (52), and (53), we conclude that the Piola stress S and the chemical potential μ of the interstitial liquid are given by

$$S = \frac{\partial \bar{\psi}(F)}{\partial F} - \varpi F^C \qquad \text{and} \qquad \mu = v\varpi. \tag{54}$$

2. We next suppose that the free-energy density ψ is provided constitutively as a function of F and C. Then, we can use the mechanical incompressibility constraint (34) to infer that

$$\bar{\psi}(F) = \hat{\psi}(F, C) \tag{55}$$

and, consequently, that the relation $(42)_2$ between the response functions $\bar{\psi}$ and $\bar{S}$ determining the free-energy density ψ and the effective stress P can be written as

$$P = \frac{\partial \bar{\psi}(F)}{\partial F} = \frac{\partial \hat{\psi}(F, C)}{\partial F} + \frac{1}{v} \frac{\partial \hat{\psi}(F, C)}{\partial C} F^C, \tag{56}$$

which suggests our second choice for S_a and μ_a, namely

$$S_a = \frac{\partial \hat{\psi}(F, C)}{\partial F} \qquad \text{and} \qquad \mu_a = \frac{\partial \hat{\psi}(F, C)}{\partial C}. \tag{57}$$

Combining $(45)_{2,3}$, (51), and (57), we conclude that for our second choice the Piola stress S and the chemical potential μ of the interstitial liquid are given by

$$S = \frac{\partial \hat{\psi}(F, C)}{\partial F} - \varpi F^C \qquad \text{and} \qquad \mu = \frac{\partial \hat{\psi}(F, C)}{\partial C} + v\varpi. \tag{58}$$

With reference to (54) and (58), we emphasize that ϖ is a problem dependent field that enters the theory to ensure the constraint (34), a role that is completely analogous to that of the pressure in an incompressible solid or liquid. Notice that $(58)_2$ shows that the chemical potential μ of the solvent in the gel is given by the sum of an active part obtained by differentiating the free energy $\hat{\psi}$ with respect to C, as if F and C were independent, plus a reactive part that comes from the fact that F and C are constrained by the liquid-induced compressibility condition. This second term can be evaluated by using $(58)_1$ and hence accounts for mechanical effects on the chemical potential μ of the interstitial liquid.

It is worth mentioning that the free-energy response of a gel is often, from the outset, written as a function of the deformation variable F and a measure, whether C or ϕ, of liquid content. Thus, bearing (34) in mind, the alternatives presented above can be used to arrive at the same final expressions for S and μ. The difference between the two alternatives relies on the interpretation of μ. For the remainder of this paper we will adopt the alternative leading to the choice (57) for the active parts of the Piola stress and chemical potential and hence to the representation (58) for the total Piola stress and chemical potential.

4.1.5. Fluid and osmotic pressures and network stress

The response function $\hat{\psi}$ determining the free-energy density ψ as a function of the deformation gradient F and the liquid concentration C must properly incorporate contributions due to the "unmixed" solid- and liquid-like constituents, the mixing of those constituents, and elastic deformation of the solid-like constituent. We therefore assume that $\hat{\psi}$ is of the form

$$\hat{\psi}(F, C) = \mu_0^L C + \hat{\Psi}(F, C), \tag{59}$$

where μ_0^L is the chemical potential of the pure solvent in a reference liquid state and $\hat{\Psi}$ incorporates the contributions to the energetic response due to mixing and network deformation. Notice that since the network content is conserved, the contribution due to the "unmixed" network strands gives rise to inconsequential additive constant that has accordingly been omitted.

We now explore some consequences of the decomposition (59). By (57), it follows that the active parts of the Piola stress S and chemical potential μ of the interstitial liquid are such that

$$S_a = \frac{\partial \hat{\Psi}(F, C)}{\partial F} \qquad \text{and} \qquad \mu_a - \mu_0^L = \frac{\partial \hat{\Psi}(F, C)}{\partial C}. \tag{60}$$

Further, by $(58)_2$, the chemical potential of the liquid in the gel relative to its chemical potential in the absence of the network, namely

$$\mu - \mu_0^L = \frac{\partial \hat{\Psi}(F, C)}{\partial C} + v\varpi, \tag{61}$$

is the sum of a "chemical" term obtained by differentiating the free energy $\hat{\Psi}$ with respect to C plus a reactive term originated from the fluid-induced compressibility constraint. By $(58)_1$, the reactive contribution to the chemical potential accounts for mechanical effects on the relative chemical potential $\mu - \mu_0^L$.

From the relative chemical potential $\mu - \mu_0^L$ and its active part $\mu_a - \mu_0^L$ we define quantities p and π by the relations

$$p := \frac{\mu - \mu_0^L}{v} \qquad \text{and} \qquad \pi := -\frac{\mu_a - \mu_0^L}{v} = -\frac{1}{v}\frac{\partial \hat{\Psi}(F, C)}{\partial C}. \tag{62}$$

Granted that the chemical potential μ_L of a liquid at pressure p_L in the absence of the network is equal to $\mu_L = \mu_0^L + v p_L$, we may interpret p and $-\pi$ as the pressures the interstitial liquid alone would have in states with $\mu_L = \mu$ and $\mu_L = \mu_a$, respectively. A related quantity is the activity a of the interstitial liquid in the gel defined by the standard relation $\ln a = (\mu - \mu_0^L)/(k_B T)$, where k_B is the Boltzmann constant and T the absolute temperature. The definition $(62)_2$ of π also resembles the definition of osmotic pressure in the context of polymer solutions, as discussed for instance by Flory (1953). Motivated by these considerations, here and henceforth we refer to p as the liquid pressure and to π as the osmotic pressure.

By (61) and (62), we write the reaction ϖ as

$$\varpi = p + \pi, \tag{63}$$

which together with $(58)_1$ allows us to express the Piola stress S as

$$S = S_{\mathrm{n}} - p F^{\mathrm{C}}, \tag{64}$$

where S_{n} has the form

$$S_{\mathrm{n}} = \frac{\partial \hat{\Psi}(F, C)}{\partial F} - \pi F^{\mathrm{C}}. \tag{65}$$

From $(20)_1$, (64), and (65), we see that the Cauchy stress T takes the form

$$T = T_{\mathrm{n}} - p I, \tag{66}$$

where $T_{\mathrm{n}} = S_{\mathrm{n}}(F^{\mathrm{C}})^{-1}$. Building upon the interpretation of p as the interstitial fluid pressure, (62)–(64) show that the Piola stress tensor S can be decomposed as the sum of two contributions, one due to the network other due to the fluid. Whereas S_{n} is due to distortions of the network, $-pF^{\mathrm{C}}$ is due to pressurization of the interstitial fluid. Moreover, the network stress S_{n} has two origins, one elastic and other due to mixing. A similar interpretation holds true for the decomposition (66) of the Cauchy stress T.

Strictly speaking, the quantity π introduced above is the chemical part of the osmotic pressure. Indeed, as observed by the present authors (Duda et al., 2010), the osmotic pressure Π can be seen as the difference between the hydrostatic stress $p_G = -(\mathrm{tr}T)/3$ in the gel and the liquid pressure p. Thus, by (65) and (66),

$$\Pi = -\frac{1}{3}\mathrm{tr}T_{\mathrm{n}} = -\frac{1}{3}\mathrm{tr}T_{\mathrm{a}} + \pi, \tag{67}$$

where the relation $T_{\mathrm{n}} = T_{\mathrm{a}} - \pi I$ between the network and active parts, T_{n} and T_{a}, of the Cauchy stress T has been used. This shows that $-\mathrm{tr}T_{\mathrm{a}}/3$ and π are, respectively, the "mechanical" and "chemical" contributions to the osmotic pressure. Another way to see this is to consider a gel in equilibrium when immersed in a pure liquid solution. Under these conditions, the chemical potential μ of the liquid is constant and equal to the ambient chemical potential μ_a, which for an environment composed of a pure liquid solution is equal $\mu_0^L + v p_a$, where p_a is the ambient liquid pressure and, hence, we see from $(62)_1$ that $p = p_a$. If the equilibrium deformation of the gel is prescribed, $\Pi = p_G - p_a$ is the extra pressure that must be applied at the boundary of the gel to ensure that the system is in a state of mechanical equilibrium.

For later reference, we notice with reference to (34) and $(62)_2$ that the osmotic pressure π can be expressed as a function of F through

$$\pi = \hat{\pi}(F) := -\frac{\partial \hat{\Psi}(F, \zeta/v)}{\partial \zeta}\bigg|_{\zeta = \det F - \phi_0}. \tag{68}$$

Moreover, using the mechanical incompressibility constraint (34) to write

$$\bar{\Psi}(F) := \hat{\Psi}(F, (\det F - \phi_0)/v), \tag{69}$$

we see from (65) and (68) that the network stress S_{n} can be expressed as

$$S_{\mathrm{n}} = \frac{\partial \bar{\Psi}(F)}{\partial F}. \tag{70}$$

4.1.6. A particular constitutive theory

We now present a constitutive theory based on specific choices for the response functions $\hat{\Psi}$ and $\bar{\xi}$. Following Flory and Rehner (1943), we assume that $\hat{\Psi}$, which characterizes the energetic response due to mixing and network elastic deformation is additive and separable. Accordingly, we write $\hat{\Psi}$ as

$$\hat{\Psi}(F, C) = \hat{\psi}_e(F) + \hat{\psi}_m(C), \tag{71}$$

where $\hat{\psi}_e$ accounts for the deformation of the network and $\hat{\psi}_m$ is the free energy of mixing.[1]

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[1] Earlier discussion in support of the decomposition (71) of the free-energy density can be found in the classical treatises by Flory (1953) and Treloar (1975). For more recent discussions we refer the reader to Horkay and McKenna (2007), McKenna (2012, 2018), and references cited therein.

For an isotropic elastic response, $\hat{\psi}_e$ can depend on F only through the principal invariants

$$I_1 = \text{tr } B, \qquad I_2 = \tfrac{1}{2}(I_1^2 - \text{tr}(B^2)), \qquad I_3 = \det B, \tag{72}$$

of the left Cauchy–Green tensor $B = FF^\top$. Throughout the literature, two families of elastic models for swollen polymer networks can be found according to which the third invariant I_3 is accounted for or not, with Flory (1953) and Treloar (1975) being pioneering representatives of these families. For our purposes, it is enough to assume that $\hat{\psi}_e$ has the type

$$\hat{\psi}_e(F) = \tilde{\psi}_e(I_1, I_2). \tag{73}$$

For elastic models described by (73), we see from (65) and (71) that the network contribution (65) to the Piola stress has the explicit form

$$S_n = 2\alpha_1 F + 2\alpha_2(I_1 I - B)F - \pi F^C, \tag{74}$$

where α_1 and α_2 are defined by

$$\alpha_1 = \frac{\partial \tilde{\psi}_e(I_1, I_2)}{\partial I_1} \qquad \text{and} \qquad \alpha_2 = \frac{\partial \tilde{\psi}_e(I_1, I_2)}{\partial I_2} \tag{75}$$

and, thus, generally depend on the scalar invariants I_1 and I_2. This implies, together with $(20)_1$, that the network Cauchy T_n can be written as

$$T_n = S_n F^{-C} = \frac{1}{\det F} S_n F^\top = \frac{2}{\det F}\Big(\alpha_1 B + \alpha_2(I_1 I - B)B\Big) - \pi I. \tag{76}$$

We now turn to the constitutive relation $(41)_3$ determining $\xi = \nabla\mu$ in terms of F and J and hereinafter assume that it can be inverted and written as

$$J = \bar{J}(F, \xi) = -\hat{M}(F)\nabla\mu, \tag{77}$$

where $\hat{M}$ is a positive semidefinite tensor-valued function related to the mobility of the interstitial liquid. The particular choice

$$\hat{M}(F) = \hat{m}(\det F)F^{-1}F^C \tag{78}$$

for $\hat{M}$ and the attendant relation

$$J = -\hat{m}(\det F)F^{-1}F^C\nabla\mu \tag{79}$$

for J are suitable for an isotropic gel. Indeed, granted that (78) holds, we see from $(20)_6$ and $\nabla\mu = F^\top \text{grad}\,\mu$ that the Cauchy fluid flux $\jmath$ is given by.[2]

$$\jmath = -\hat{m}(\det F)\text{grad}\,\mu. \tag{80}$$

For later reference, we use $(62)_1$ and rewrite (79) and (80) as

$$J = -\hat{m}(\det F)F^{-1}F^C\nabla p \tag{81}$$

and

$$\jmath = -v\hat{m}(\det F)\text{grad}\,p. \tag{82}$$

4.2. Interfacial constitutive theory

In what amounts to a basic constitutive assumption for the interface, we restrict attention to processes in which the chemical potential μ of the interstitial liquid is continuous across the interface S, so that

$$[\![\mu]\!] = 0. \tag{83}$$

Using (83) in (18), we find that the interfacial free-energy imbalance simplifies to

$$(\mathtt{n} \cdot [\![\Sigma]\!]\mathtt{n} - f_i)U \geq 0. \tag{84}$$

Guided by (84), we consider a simple interfacial constitutive theory in which the quantity f defined by

$$f = \mathtt{n} \cdot [\![\Sigma]\!]\mathtt{n} - f_i \tag{85}$$

serves as a dependent constitutive variable and the scalar normal velocity U, to which f is power-conjugate, serves as the independent constitutive variable. Thus, we consider the class of interfacial constitutive relations of the form $f = \hat{f}(U)$. For the

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[2] It can be shown that the most general constitutive representation for $\jmath$ as function of $(F, \text{grad}\,\mu)$ in the isotropic case is $\jmath = (m_0 I + m_1 B + m_2 B^2)\text{grad}\,\mu$, where m_0, m_1, and m_2 can be expressed as functions of $(I_1, I_2, I_3, |\text{grad}\,\mu|, |\sqrt{B}\text{grad}\,\mu|, |B\text{grad}\,\mu|)$.

purposes of this work, we will consider that interfacial motion occurs without dissipation and hence choose $\hat{f}(U) \equiv 0$. This choice assures that (84) holds in all processes and allows us to write (85) as

$$f_i = \mathbb{n} \cdot [\![\boldsymbol{\Sigma}]\!]\mathbb{n}. \tag{86}$$

If the external configurational force density f_e vanishes, we may supplement the configurational force balance (15) with the definition (17) of the configurational stress tensor $\boldsymbol{\Sigma}$ and (86) to find that

$$[\![\psi - \mu C]\!] - \boldsymbol{S}\mathbb{n} \cdot [\![\boldsymbol{F}]\!]\mathbb{n} = 0. \tag{87}$$

Then again, combining the mechanical incompressibility condition (34), the defining relation $(62)_1$ for the liquid pressure p, and the decomposition (64) of the Piola stress $\boldsymbol{S}$, we find that $\boldsymbol{\Sigma}$ can be written

$$\boldsymbol{\Sigma} = (\psi - \mu_0^L C)\boldsymbol{I} - \boldsymbol{F}^\top \boldsymbol{S}_{\mathrm{n}}. \tag{88}$$

From (64), the interfacial force balance $(14)_1$, the requirement $(3)_3$ that $\boldsymbol{F}^C\mathbb{n}$ be continuous across $\mathcal{S}$, and the consequence

$$[\![p]\!] = 0 \tag{89}$$

of $(62)_1$ and (83), we find that the network stress $\boldsymbol{S}_{\mathrm{n}}$ must be consistent with the traction continuity condition

$$[\![\boldsymbol{S}_{\mathrm{n}}]\!]\mathbb{n} = \boldsymbol{0}. \tag{90}$$

Using (88) and (90) in (86), we see that (87) takes the alternative form

$$[\![\psi - \mu_0^L C]\!] - \boldsymbol{S}_{\mathrm{n}}\mathbb{n} \cdot [\![\boldsymbol{F}]\!]\mathbb{n} = 0. \tag{91}$$

Several remarks are now in order:

- Like the Piola stress $\boldsymbol{S}$, the configurational stress $\boldsymbol{\Sigma}$, may be decomposed into contributions associated with the network and the interstitial liquid. Indeed, taking (34), (59), $(62)_1$, and (64) into consideration, we see from (17) that

$$\boldsymbol{\Sigma} = \boldsymbol{\Sigma}_{\mathrm{n}} + \boldsymbol{\Sigma}_{\mathrm{f}}, \tag{92}$$

  where $\boldsymbol{\Sigma}_{\mathrm{n}}$ and $\boldsymbol{\Sigma}_{\mathrm{f}}$ defined by

$$\boldsymbol{\Sigma}_{\mathrm{n}} = (\psi - \mu_0^L C)\boldsymbol{I} - \boldsymbol{F}^\top \boldsymbol{S}_{\mathrm{n}} \qquad \text{and} \qquad \boldsymbol{\Sigma}_{\mathrm{f}} = -p\phi_0\boldsymbol{I}, \tag{93}$$

  are the contributions the configurational stress $\boldsymbol{\Sigma}$ associated with the network and the interstitial liquid. From the representation

$$f_i = [\![\psi - \mu_0^L C]\!] - \boldsymbol{S}_{\mathrm{n}}\mathbb{n} \cdot [\![\boldsymbol{F}]\!]\mathbb{n} \tag{94}$$

  that ensues from using the decomposition (92)–(93) in (86), it is evident that the jump in the normal configurational stress acting on the plane perpendicular to $\mathbb{n}$ is determined by the network configurational stress.
- Early derivations of (87), suitable for equilibrium situations and using variational arguments, can be traced back to Robin (1974) and Larche and Cahn (1978). The condition (87), augmented by the presence of surface tension, was used by Dolbow et al. (2004) in a study of the kinetics of phase transition in hydrogels. See also Ji et al. (2006), who accounted for thermal effects and linear transition kinetics.
- Using (59), (69), and (70) in (91) yields a condition,

$$[\![\bar{\psi}(\boldsymbol{F})]\!] - \frac{\partial \bar{\psi}(\boldsymbol{F})}{\partial \boldsymbol{F}}\mathbb{n} \cdot [\![\boldsymbol{F}]\!]\mathbb{n} = 0, \tag{95}$$

  which is formally identical to the wildly imposed Maxwell relation for two-phase elastic bodies, as discussed, for instance, by Gurtin (1999), Šilhavý (1997), and the references therein. It is well known that the Maxwell relation appears as a consequence of one of the following assumptions: stationarity of the first variation of the total potential energy with respect to variations of the interface position, stability of equilibria, and dissipation-free interfacial motion.
- To the best of our knowledge, the first derivation of (95) for polymer gels was provided by Sekimoto and Kawasaki (1989) as a consequence of the stationarity of the first variation of the total free-energy of a two-phase gel body with respect to variations of the interface position. Implicit in their treatment was the use of the constraint (34) allowing a two-phase gel body to be treated as a two-phase elastic body. Although the derivation of Sekimoto & Kawasaki was performed under equilibrium conditions, the relation (95) has subsequently been used even outside equilibrium and in that context has been referred to as the "local equilibrium coexistence condition". See, for instance, Doi (2009).
- With reference to compatibility condition $(3)_3$ for $\boldsymbol{F}^C\mathbb{n}$, the mechanical incompressibility constraint (34), the decomposition (59) of the response function $\hat{\psi}$, the relation (63) determining the reaction ϖ to (34), and the expressions (64) and (65) for the Piola and network stresses $\boldsymbol{S}$ and $\boldsymbol{S}_{\mathrm{n}}$, we obtain an alternative to the Maxwell relation (95) in which the dependence upon the liquid content C remains explicit and the influence of incompressibility is evident:

$$[\![\hat{\Psi}(\boldsymbol{F}, C)]\!] - \frac{\partial \hat{\Psi}(\boldsymbol{F}, C)}{\partial \boldsymbol{F}}\mathbb{n} \cdot [\![\boldsymbol{F}]\!]\mathbb{n} - \upsilon\varpi[\![C]\!] = 0. \tag{96}$$

- The representation (17) for the Eshelby stress $\boldsymbol{\Sigma}$ may be seen as a finite-strain extension of that obtained by Gurtin and Voorhees (1993) (see also Fried and Gurtin, 1999), when specialized to interfaces without structure.

5. Referential versions of the governing equations

The governing equations of the theory are obtained by combining the basic balances and constitutive relations, in bulk and on the interface. In this section, we present those equations for the particular constitutive theory, pertaining to isotropic gels, introduced in Section 4.1.6. For simplicity, external supplies of standard body force, liquid, and configurational forces are neglected:

$$\boldsymbol{b}_0 = \boldsymbol{0}, \qquad Q = 0, \qquad \text{and} \qquad f_e = 0. \tag{97}$$

Along with the bulk and interface equations, boundary conditions describing mechanical and permeation interactions between the solid and the environment must also be formulated and applied. Here, we suppose that for a material point located at the boundary $\partial \mathcal{B}$, with outward unit normal $\partial \mathcal{B}$, between the solid and the environment:

- Mechanical conditions: either the Piola traction $\boldsymbol{S}\boldsymbol{n}$ or the motion $\boldsymbol{y}$ is prescribed;
- Permeation conditions: either the Piola normal flux $\boldsymbol{J} \cdot \boldsymbol{n}$ or the chemical potential μ — or, by virtue of $(62)_1$, the fluid pressure p — is prescribed.

We choose the motion $\boldsymbol{y}$, polymer volume fraction ϕ, fluid pressure p, and interface location S as the unknown variables and write the governing equations in a manner that reflects this choice. This set of unknown variables is often used to formulate the equations of gel dynamics; see, for instance, Doi (2013). Provided some identifications are carried out, the final equations so obtained may be interpreted as describing fluid permeation in finite-strained poroelastic bodies, an idea explored in detail by Anand (2015) in a context where the strains and rotations are infinitesimal. It is worth mentioning that polymer gels have often been described as poroelastic bodies, irrespective of the physical differences between these two type of materials. The issue of coupling large deformation and fluid permeation with the context of poroelasticity remains a topic of intense research given its importance for both scientific and technological applications. For a recent discussion on the subject matter see MacMinn et al. (2016).

5.1. Bulk equations

The bulk equations of theory consist of the standard force balance, the network content balance, and the liquid content balance as presented in (11) in conjunction with the expression (64) determining the Piola stress $\boldsymbol{S}$ in terms of the network stress $\boldsymbol{S}_n$ and the liquid pressure p, as defined by $(62)_1$, and the expression (81) determining the Piola flux $\boldsymbol{J}$. Recalling that $(11)_2$ and (33) are equivalent and using (34) to eliminate C from $(11)_3$, these equations can be expressed as

$$\left. \begin{aligned} \text{Div}(\boldsymbol{S}_n - p\boldsymbol{F}^C) &= \boldsymbol{0}, \\ \phi \det \boldsymbol{F} &= \phi_0, \\ \overline{\det \boldsymbol{F}} &= -v\text{Div}\boldsymbol{J}, \end{aligned} \right\} \tag{98}$$

where, with reference to (74) and (81), the network Piola stress $\boldsymbol{S}_n$ and Piola fluid flux $\boldsymbol{J}$ are given by

$$\boldsymbol{S}_n = 2\alpha_1 \boldsymbol{F} + 2\alpha_2 (I_1 \boldsymbol{I} - \boldsymbol{B})\boldsymbol{F} - \pi \boldsymbol{F}^C \qquad \text{and} \qquad \boldsymbol{J} = -v\hat{m}(\det \boldsymbol{F})\boldsymbol{F}^{-1}\boldsymbol{F}^C \nabla p. \tag{99}$$

In writing $(99)_1$, we recall that α_1 and α_2 are determined by the deformation gradient $\boldsymbol{F}$ through (75). Additionally, while π is most primitively a function of C, the mechanical incompressibility constraint (33) allows us to treat it as a function of $\boldsymbol{F}$, as demonstrated in (68). Keeping in mind that $v\boldsymbol{J}$ is the referential volumetric flux of interstitial fluid, $(98)_3$ states, in referential terms, that local volumes changes are caused by the volumetric flux of the interstitial fluid.

5.2. Interfacial equations

From (14), (33), (59), and (91), the equations at the interface S take the form

$$\left. \begin{aligned} [\![\boldsymbol{S}_n]\!]\mathbb{n} &= \boldsymbol{0}, \\ [\![\det \boldsymbol{F}]\!]U - [\![v\boldsymbol{J}]\!] \cdot \mathbb{n} &= 0, \\ [\![\Psi]\!] - \boldsymbol{S}_n\mathbb{n} \cdot [\![\boldsymbol{F}]\!]\mathbb{n} &= 0, \end{aligned} \right\} \tag{100}$$

where $\boldsymbol{S}_n$ and $\boldsymbol{J}$ are given by (99) and, with reference to (59) and (71), Ψ is given by

$$\Psi = \tilde{\psi}_e(I_1, I_2) + \hat{\psi}_m((\det \boldsymbol{F} - \phi_0)/v). \tag{101}$$

In writing $(100)_1$, we have used the continuity of $\boldsymbol{F}^C\mathbb{n}$ (see $(3)_3$) and p across S to eliminate the pressure contribution. In this regard, we recall that the continuity of p stems from the assumption that the chemical potential μ is continuous across S and the relation $(62)_1$ determining p in terms of μ. We have also used (34) to replace C by $\det \boldsymbol{F}$ in $(100)_2$ and (101).

6. Spatial versions of the governing equations

6.1. Bulk equations

The spatial counterparts of the referential governing equations (98)–(99) are obtained on combining (19), (20), and (66). We may write these equations as

$$
\left.
\begin{aligned}
\operatorname{div}(\boldsymbol{T}_n - p\boldsymbol{I}) &= \boldsymbol{0}, \\
\phi \det \boldsymbol{F} &= \phi_0, \\
\operatorname{div}(v\boldsymbol{J} + \boldsymbol{v}) &= 0,
\end{aligned}
\right\}
\tag{102}
$$

where, with reference to (76) and (82), the network Cauchy stress $\boldsymbol{T}_n$ and the Cauchy fluid flux $\boldsymbol{J}$ are given by

$$
\boldsymbol{T}_n = \frac{2}{\det \boldsymbol{F}}\left(\alpha_1 \boldsymbol{B} + \alpha_2(I_1 \boldsymbol{I} - \boldsymbol{B})\boldsymbol{B}\right) - \pi \boldsymbol{I}
\quad\text{and}\quad
\boldsymbol{J} = -v\hat{m}(\det \boldsymbol{F})\operatorname{grad} p.
\tag{103}
$$

The equations of gel dynamics presented by Doi (2009) can be obtained from (102) and (103). Specifically, by standard arguments of continuum mechanics, $(102)_2$ is equivalent to the evolution equation

$$
\frac{\partial \phi}{\partial t} + \operatorname{div}(\phi \boldsymbol{v}) = 0
\tag{104}
$$

for the polymer volume fraction ϕ. Additionally, upon introducing the solvent velocity $\boldsymbol{v}_s$ through the relation

$$
\boldsymbol{J} = c(\boldsymbol{v}_s - \boldsymbol{v})
\tag{105}
$$

and recalling that, by (32), $cv = 1 - \phi$, $(102)_3$ and $(103)_2$ can be recast as

$$
\operatorname{div}((1-\phi)\boldsymbol{v}_s + \phi\boldsymbol{v}) = 0
\quad\text{and}\quad
\boldsymbol{v}_s - \boldsymbol{v} = -\kappa \operatorname{grad} p,
\tag{106}
$$

where κ is defined by

$$
\kappa = \hat{\kappa}(\phi) := \frac{\hat{m}(\phi_0/\phi)}{1-\phi}.
\tag{107}
$$

From $(106)_2$ it is evident that permeation velocity $\boldsymbol{v}_s - \boldsymbol{v}$ is proportional to the pressure gradient $\operatorname{grad} p$, a relation that is known within the theory of porous media as Darcy's law. The quantity κ given in (107) is a non-negative function of ϕ which is related to the permeability. Finally, the equations of gel dynamics presented by Doi (2009) are obtained from (102) and (103) by replacing $(102)_2$, $(102)_3$, and $(103)_2$ by (104), $(106)_1$, and $(106)_2$, respectively. Supplementing these equations by the purely kinematical evolution equation

$$
\frac{\partial \boldsymbol{B}}{\partial t} + (\operatorname{grad} \boldsymbol{B})\boldsymbol{v} = (\operatorname{grad} \boldsymbol{v})\boldsymbol{B} + \boldsymbol{B}(\operatorname{grad} \boldsymbol{v})^{\top}
\tag{108}
$$

for $\boldsymbol{B}$ yields a system of equations for ϕ, $\boldsymbol{v}$, $\boldsymbol{v}_s$, and p.

6.2. Interface equations

The spatial counterparts of the interfacial conditions (100), which hold on the spatial image s of S obtain on combining (22), $(23)_1$, (66), and (91). We may write these equations as

$$
\left.
\begin{aligned}
[\![\boldsymbol{T}_n]\!]\mathrm{m} &= \boldsymbol{0}, \\
[\![\phi]\!]u - [\![\phi\boldsymbol{v}]\!]\cdot\mathrm{m} &= 0, \\
[\![v\boldsymbol{J} + \boldsymbol{v}]\!]\cdot\mathrm{m} &= 0, \\
[\![\boldsymbol{\Psi}]\!] - \boldsymbol{T}_n\mathrm{m}\cdot\left[\!\!\left[\frac{\phi_0 \boldsymbol{B}}{\phi\mathrm{m}\cdot\boldsymbol{B}\mathrm{m}}\right]\!\!\right]\mathrm{m} &= 0,
\end{aligned}
\right\}
\tag{109}
$$

where m and u, as defined by (23), represent the unit normal to m, the spatial scalar normal velocity of s and $\boldsymbol{T}_n$ and $\boldsymbol{J}$ are given by (103), and we have used the consequences

$$
\frac{[\![\boldsymbol{F}]\!]\mathrm{n}}{|\boldsymbol{F}^{\mathrm{C}}\mathrm{n}|} = \left[\!\!\left[\frac{\boldsymbol{B}}{\det \boldsymbol{F}}\frac{\boldsymbol{F}^{\mathrm{C}}\mathrm{n}}{|\boldsymbol{F}^{\mathrm{C}}\mathrm{n}|}\right]\!\!\right] = \left[\!\!\left[\frac{\phi \boldsymbol{B}}{\phi_0}\right]\!\!\right]\frac{\langle\!\langle \boldsymbol{F}^{\mathrm{C}}\rangle\!\rangle \mathrm{n}}{|\langle\!\langle \boldsymbol{F}^{\mathrm{C}}\rangle\!\rangle\mathrm{n}|} + \left\langle\!\!\left\langle\frac{\phi \boldsymbol{B}}{\phi_0}\right\rangle\!\!\right\rangle\frac{[\![\boldsymbol{F}^{\mathrm{C}}]\!]\mathrm{n}}{|\boldsymbol{F}^{\mathrm{C}}\mathrm{n}|} = \left[\!\!\left[\frac{\phi \boldsymbol{B}}{\phi_0}\right]\!\!\right]\mathrm{m}
\tag{110}
$$

and

$$
\mathrm{m}\cdot\boldsymbol{B}\mathrm{m} = |\boldsymbol{F}^{\top}\mathrm{m}|^2 = \frac{\boldsymbol{F}^{\top}\boldsymbol{F}^{\mathrm{C}}\mathrm{n}}{|\boldsymbol{F}^{\mathrm{C}}\mathrm{n}|}\cdot\frac{\boldsymbol{F}^{\top}\boldsymbol{F}^{\mathrm{C}}\mathrm{n}}{|\boldsymbol{F}^{\mathrm{C}}\mathrm{n}|} = \frac{\det \boldsymbol{B}}{|\boldsymbol{F}^{\mathrm{C}}\mathrm{n}|^2} = \frac{\phi_0^2}{\phi^2|\boldsymbol{F}^{\mathrm{C}}\mathrm{n}|^2}
\tag{111}
$$

of $(3)_3$, $(23)_1$, and (33) to derive $(109)_4$ from the referential form $(100)_3$ of configurational force balance. The latter derivation makes no use of the constitutive relations (71) and (76) determining the free-energy density $\boldsymbol{\Psi}$ and the Cauchy network stress $\boldsymbol{T}_n$ and, hence, holds independent of the elastic properties of the gel.
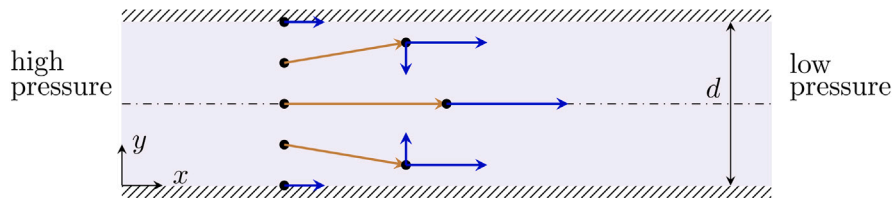
Fig. 1. Schematic of the channel flow problem: A gel placed between two infinitely long, stationary, impermeable, and parallel walls separated by a gap d is forced by a pressure drop in the streamwise direction. Material points identified with their locations before the application of the pressure difference move as indicated by the corresponding (brown) displacement vectors and liquid permeation at their corresponding spatial locations takes place as indicated by the components of the (blue) flux vectors. The outermost materials points do not move since the gel is presumed to be fixed to the walls. Material points symmetrically located with respect to the central line remain so after the application of the pressure drop, and liquid flow may take place in both streamwise and spanwise directions.

7. Pressure-driven flow through a gel-filled channel

We next apply the system of equations comprised by (102), (103), and (109) to study the behavior of a gel of uniform composition placed between two flat walls and subjected to a pressure drop, as depicted in Fig. 1. Specifically, we consider the space between two infinitely long impermeable parallel walls separated by a gap d and completely filled by a gel that adheres to the channel walls. The gel is taken to be initially in a stress-free state with polymer volume fraction constant and equal to ϕ_0. A fixed pressure drop is then applied to the system, causing the gel to deform and the interstitial liquid to flow through the network. Our primary objective is to investigate how the elastic and osmotic responses of the gel interact to determine its transient and state–state behaviors.

Our investigation will proceed in steps, in a manner similar to that used by Coleman et al. (1966) in their study of the channel flow of a general incompressible fluid. We begin by introducing an Ansatz concerning the form of the deformation field and interface location and orientation. Stipulating that our Ansatz satisfies the governing equations, we next arrive at a set of equations to be solved for the various unknown quantities that are needed to completely specify a solution to our problem.

7.1. Ansatz

Considering a gel situated between and fixed to two infinite parallel walls and subjected to an applied pressure drop, we introduce the following kinematical assumptions:

1. Each material plane parallel to the channel walls undergoes a rigid motion characterized by the composition of two translations, one along the direction of the applied pressure drop and other in the direction orthogonal to the channel walls. The magnitudes of these translations are allowed to vary from material plane to material plane. In particular, the material planes in contact with the channel walls remain fixed.
2. Each pair of material planes that is symmetrically located with respect to the central plane between the channel walls experience the same displacement in the direction of the applied pressure drop but opposite displacements in the direction orthogonal to the walls.
3. The central material plane translates only along the direction of the applied pressure drop.

To expresses the foregoing assumptions mathematically, we introduce a right-handed Cartesian coordinate system with the x-axis being parallel to the direction of decreasing pressure, the y-axis orthogonal to the channel walls, and z-axis orthogonal to the (x, y)-plane. We suppose that all fields are independent of z. Additionally, we take the channel walls to be located at $y = 0$ and $y = d$. With this choice of coordinates, the state reached by the gel at time t due to an imposed pressure drop is such that:

- The components of the inverse deformation mapping $\mathbf{y}^{-1}$ take the form

$$X = x + f(y, t), \qquad Y = y + g(y, t), \qquad Z = z, \tag{112}$$

where (X, Y, Z) are the referential coordinates of a gel particle, and f and g entering $(112)_1$ and $(112)_2$ are to be determined.
- Consistent with the requirement that the gel is fixed to the channel walls at $y = 0$ and $y = d$, f and g obey the conditions

$$f(0, t) = f(d, t) = 0 \qquad \text{and} \qquad g(0, t) = g(d, t) = 0. \tag{113}$$

- The functions f and g entering (112) obey the symmetry relations with respect to the central line $y = d/2$ between the channel walls:

$$f(y, t) = f(d - y, t) \qquad \text{and} \qquad g(y, t) = -g(d - y, t), \qquad 0 \le y \le d. \tag{114}$$

It is evident that the requirements itemized above express the foregoing assumptions on the motion undergone by the gel due to an applied pressure drop.[3]

We also allow for the possibility that the state of the gel may involve the presence of planar interfaces symmetrically located with respect to the central plane of the channel. Specifically, if such planar interfaces are present at time t, we denote their positions relative to the plane $y = d/2$ by

$$y = w(t) \quad \text{and} \quad y = d - w(t), \quad \text{with} \quad 0 < w(t) < d/2. \tag{115}$$

In absence of interfaces, the continuous functions f and g are smooth on the entire interval $(0, d)$. Otherwise, they are piecewise smooth, meaning that they are smooth on the sub-intervals $(0, w)$, $(w, d/2)$, and $(d - w, d)$.

We next explore the implications of our assumptions on relevant kinematic quantities as well as on the network stress.

7.1.1. Kinematic quantities

Suppose that t is fixed and consider that $y \in [0, d]$, with $y \neq w(t)$ and $y \neq d - w(t)$. A direct calculation using (112) yields that the matrix representation of F relative to the underlying coordinate frame is given by

$$[F] = \begin{bmatrix} 1 & \gamma & 0 \\ 0 & \lambda & 0 \\ 0 & 0 & 1 \end{bmatrix}, \tag{116}$$

where γ and λ are given by

$$\gamma := -\frac{\partial f}{\partial y}\left(1 + \frac{\partial g}{\partial y}\right)^{-1} \quad \text{and} \quad \lambda := \left(1 + \frac{\partial g}{\partial y}\right)^{-1}. \tag{117}$$

The matrix representations of $B = FF^{\top}$ and F^{C} relative to the underlying coordinate frame are thus

$$[B] = \begin{bmatrix} 1 + \gamma^2 & \gamma\lambda & 0 \\ \gamma\lambda & \lambda^2 & 0 \\ 0 & 0 & 1 \end{bmatrix} \quad \text{and} \quad [F^{C}] = \begin{bmatrix} \lambda & 0 & 0 \\ -\gamma & 1 & 0 \\ 0 & 0 & \lambda \end{bmatrix}. \tag{118}$$

From $(118)_1$, the principal scalar invariants of B are given by

$$I_1 = 2 + \gamma^2 + \lambda^2, \qquad I_2 = 1 + \gamma^2 + 2\lambda^2, \qquad I_3 = \lambda^2. \tag{119}$$

With reference to (116), we infer that F has the form of a superposition of a simple shear of amount γ and a uniaxial extension with stretch λ, which, by (114), satisfy the conditions

$$\gamma(y, t) = -\gamma(d - y, t) \quad \text{and} \quad \lambda(y, t) = \lambda(d - y, t). \tag{120}$$

From $(120)_1$, we see that the amount of shear vanishes at $y = d/2$, that is,

$$\gamma(d/2, t) = 0. \tag{121}$$

Referring to the boundary conditions in $(113)_2$, which stem from the assumption that the gel adheres to the channel walls at $y = 0$ and $y = d$, and using $(117)_1$, we find that the stretch λ must satisfy the integral condition

$$\int_0^d \frac{\mathrm{d}y}{\lambda(y, t)} = d, \tag{122}$$

which, by symmetry, is equivalent to

$$\int_0^{d/2} \frac{\mathrm{d}y}{\lambda(y, t)} = \frac{d}{2}. \tag{123}$$

We now obtain the Cartesian components of v away from any interfaces that may be present. Using the classical relation $v = -F(\partial y^{-1}/\partial t)$, in conjunction with (116) and (117), we find that

$$v_x = -\gamma\frac{\partial g}{\partial t} - \frac{\partial f}{\partial t}, \qquad v_y = -\lambda\frac{\partial g}{\partial t}, \qquad \text{and} \qquad v_z = 0, \tag{124}$$

---

[3] To verify the stated assertion, let $\mathcal{M}(y_0, t)$ be the set of material points occupying the plane $y = y_0 \in [0, d]$ in the deformed configuration at time t. According to (112), this set occupies the plane $Y = Y_0$, with $Y_0 = y_0 + g(y_0, t)$, in the reference configuration. Moreover, $\mathcal{M}(y_0, t)$ undergoes a plane and rigid displacement, with X and Y components given by $-f(y_0, t)$ and $-g(y_0, t)$, respectively. This observation lead us to conclude that: (i) the set $\mathcal{M}(d/2, t)$ is displaced along the X-direction only since, by $(114)_2$, $g(d/2, t) = 0$; (ii) the material sets $\mathcal{M}(0, t)$ and $\mathcal{M}(d, t)$ remain unaltered since, by (113), f and g vanish at $y = 0$ and $y = d$; (iii) the material sets $\mathcal{M}(y_1, t)$ and $\mathcal{M}(y_2, t)$, with $0 < y_1 < y_2 < d$, slide with respect to each other without fixing distance between them; iv) the material sets $\mathcal{M}(y_0, t)$ and $\mathcal{M}(d - y_0, t)$ undergo the same displacement in the X-direction but opposite displacements in the Y-direction, as (114) shows. The latter implies that the planes occupied by $\mathcal{M}(y_0, t)$ and $\mathcal{M}(d - y_0, t)$ in the reference configuration are equidistant from the central plane. Thus, $\mathcal{M}(y_0, t)$ and $\mathcal{M}(d - y_0, t)$ occupy planes in the reference configuration and the deformed configuration, respectively, that are symmetrically located with the respect to the central plane, while the corresponding separation may change.

where v_x, v_y, and v_z are, respectively, the components of the velocity field $\boldsymbol{v}$ in the directions of increasing x, y, and z. Using (114) and (120), we conclude that

$$v_x(y,t) = v_x(d-y,t) \qquad \text{and} \qquad v_y(y,t) = -v_y(d-y,t). \tag{125}$$

Further, using the relation $\text{div}\,\boldsymbol{v} = \dot{\boldsymbol{F}} \cdot \boldsymbol{F}^{-\top}$, we find that

$$\text{div}\,\boldsymbol{v} = \frac{\partial v_y}{\partial y} = \overline{\ln \lambda} \tag{126}$$

Keeping in mind our assumption that inertia is of negligible importance, we do not present the acceleration field.

If interfaces are present at $y = w(t)$ and $y = d - w(t)$ as described in (115), we use the unit normal $\mathbf{m}$ to (x,z)-plane, oriented in the direction of increasing y, to denote their common unit. In view of this convention, we observe for later reference that the scalar normal velocities of the interfaces at $y = w(t)$ and $y = d - w(t)$ are given by

$$u(w(t),t) = \dot{w}(t) \qquad \text{and} \qquad u(d-w(t),t) = -\dot{w}(t), \tag{127}$$

respectively. Thus, the layers of the gel closest to the walls bounding the channel therefore thicken at the expense of the inner layer surrounding the central plane of the channel if $\dot{w} > 0$ and the opposite occurs when $\dot{w} < 0$.

7.1.2. The network stress

Combining $(103)_1$, $(118)_1$, and (119), we conclude that the matrix representation relative to the underlying coordinate frame of the Cauchy network stress $\boldsymbol{T}_\text{n}$ reads

$$[\boldsymbol{T}_\text{n}] = \begin{bmatrix} \sigma_x & \tau_{xy} & 0 \\ \tau_{xy} & \sigma_y & 0 \\ 0 & 0 & \sigma_z \end{bmatrix}, \tag{128}$$

with σ_x, σ_y, σ_z, and τ_{xy} given by

$$\left. \begin{aligned} \sigma_x &= \frac{2}{\lambda}(\alpha_1(1+\gamma^2) + \alpha_2(1+\gamma^2+\lambda^2)) - \hat{\pi}(\lambda), \\ \sigma_y &= 2\lambda(\alpha_1 + 2\alpha_2) - \hat{\pi}(\lambda), \\ \sigma_z &= \frac{2}{\lambda}(\alpha_1 + \alpha_2(1+\lambda^2+\gamma^2)) - \hat{\pi}(\lambda), \\ \tau_{xy} &= 2\gamma(\alpha_1 + \alpha_2), \end{aligned} \right\} \tag{129}$$

where, recalling (75), α_1 and α_2 are given by

$$\alpha_1 = \hat{\alpha}_1(\gamma,\lambda) := \frac{\partial \tilde{\psi}_e(I_1, I_2)}{\partial I_1} \qquad \text{and} \qquad \alpha_2 = \hat{\alpha}_2(\gamma,\lambda) := \frac{\partial \tilde{\psi}_e(I_1, I_2)}{\partial I_2}, \tag{130}$$

with I_1 and I_2 determined in terms of γ and λ by $(119)_1$ and $(119)_2$. Since $\hat{\pi}(\lambda)$ appears only in the expressions $(129)_{1-3}$, we see the osmotic pressure affects only the normal stresses and that the shear stress is completely determined by the elastic response of the network and the same goes for the normal stress differences.

Anticipating that the constitutive equations for σ_y and τ_{xy} will be of prominent importance in the subsequent development, we find it convenient to recast $(129)_{2,4}$ as

$$\sigma_y = \hat{\sigma}(\gamma,\lambda) := 2(\hat{\alpha}_1(\gamma,\lambda) + 2\hat{\alpha}_2(\gamma,\lambda))\lambda - \hat{\pi}(\lambda), \qquad \tau_{xy} = \hat{\tau}(\gamma,\lambda) := \hat{G}(\gamma,\lambda)\gamma, \tag{131}$$

where $\hat{G}$ defined in accord with

$$\hat{G}(\gamma,\lambda) = 2(\hat{\alpha}_1(\gamma,\lambda) + \hat{\alpha}_2(\gamma,\lambda)) \tag{132}$$

is the generalized shear modulus at (γ,λ). Further, defining $\hat{\varphi}$ by

$$\hat{\varphi}(\gamma,\lambda) = \tilde{\psi}_e(2+\gamma^2+\lambda^2, 1+\gamma^2+2\lambda^2) + \hat{\psi}_m((\lambda-\phi_0)/v), \tag{133}$$

we note that $\hat{\sigma}$ and $\hat{\tau}$ are given by

$$\hat{\sigma}(\gamma,\lambda) = \frac{\partial\hat{\varphi}(\gamma,\lambda)}{\partial\lambda} \qquad \text{and} \qquad \hat{\tau}(\gamma,\lambda) = \frac{\partial\hat{\varphi}(\gamma,\lambda)}{\partial\gamma}. \tag{134}$$

Henceforth, we will require that the elastic response of the gel complies with the Baker–Ericksen inequality, which implies that $\hat{G}(\gamma,\lambda) > 0$ for $\gamma \neq 0$ (see Appendix A), and that the infinitesimal generalized shear modulus $\hat{G}(0,\lambda)$ is positive. As a consequence of these assumptions, the generalized shear modulus $\hat{G}(\gamma,\lambda) > 0$ must always be positive, so that

$$\hat{G}(\gamma,\lambda) > 0 \tag{135}$$

for any (γ,λ) of interest.

We finalize this subsection with two remarks to which we will later return:

- In conjunction with $(131)_2$, (135) implies that for each λ, $\partial\hat{\tau}(\cdot,\lambda)/\partial\gamma > 0$ in a neighborhood of $(0,\lambda)$, and, hence, that $\hat{\tau}(\cdot,\lambda)$ is invertible in that same neighborhood. However, if we consider only situations in which interfaces arise due to volume transitions (see, for instance, Dušek and Dušková-Smrčková, 2020 and Doi, 2013), we find (see Appendix B) that $\hat{\tau}(\cdot,\lambda)$ is invertible for all choices of γ rather than only in a neighborhood of $\gamma = 0$. This makes it permissible to replace γ by τ_{xy} as the independent constitutive variable, a possibility that will be explored in several occasions throughout this paper.

- By (134), $\partial\hat{\sigma}/\partial\gamma \equiv 0$ if and only if $\partial\hat{\tau}/\partial\lambda \equiv 0$, which means that the normal stress response $\hat{\sigma}$ is unaffected by the amount of shear γ whenever shear stress response $\hat{\tau}$ is unaffected by the stretch λ, and vice versa. For this condition to hold, it is necessary and sufficient that $\hat{\varphi}$ be an additive and separable function of γ and λ, that is, that $\hat{\varphi}$ can be written in the form $\hat{\varphi}(\gamma,\lambda) = \hat{\varphi}_\gamma(\gamma) + \hat{\varphi}_\lambda(\lambda)$ for single-valued functions $\hat{\varphi}_\gamma$ and $\hat{\varphi}_\lambda$. Equivalently, by (133), the response for the free energy density $\check{\psi}_e$ must have the form

$$\check{\psi}_e(I_1, I_2) = c_1 I_1 + c_2 I_2 + \hat{q}_1(I_2 - 2I_1) + \hat{q}_2(I_2 - I_1), \tag{136}$$

for some constants c_1 and c_2, and functions $\hat{q}_1$ and $\hat{q}_2$.[4] It is noteworthy that both neo-Hookean and Mooney models for nonlinear elastic networks comply with (136).

7.2. Governing equations

We next specialize the spatial versions (102), (103), and (109) of the bulk and interfacial governing equations in accord with the assumptions described in Section 7.1, the ultimate goal being to obtain a system of equations for f, g, p, and w.

We consider separately each of the basic equations augmented with the pertinent constitutive information and boundary conditions. Whenever necessary, we make use of our choice of $\mathtt{m}$ as the unit vector oriented in the direction of increasing y.

7.2.1. Polymer content conservation

From (116), we see that $\det \mathbf{F} = \lambda$ and hence $(102)_2$ can be replaced by a relation between the stretch λ and polymer volume fraction ϕ of the form

$$\lambda\phi = \phi_0, \tag{137}$$

from which we infer that ϕ, like λ, depends on y and t alone. As a consequence of (137), we see that the composition of the gel, as described by the polymer volume fraction ϕ, is fully determined by the stretch λ. Turning to the interface, we see that, after taking (125) and (127) into account, the condition $(109)_2$ takes the form

$$[\![\phi]\!]\dot{w} + [\![\phi v_y]\!] = 0. \tag{138}$$

7.2.2. Mechanical force balance

According to (128) and (129), the components of the network stress $\mathbf{T}_\mathrm{n}$ are determined by γ and λ and hence are functions of y and t only. In this case, $(102)_1$ reduces to

$$\frac{\partial\sigma_y}{\partial y} - \frac{\partial p}{\partial y} = 0, \qquad \frac{\partial\tau_{xy}}{\partial y} - \frac{\partial p}{\partial x} = 0, \qquad \frac{\partial p}{\partial z} = 0. \tag{139}$$

Next, bearing in mind that $\mathtt{m}$ is oriented in the direction of increasing y and using (128), we next deduce that $(109)_1$ requires that normal stress σ_y the shear stress τ_{xy} satisfy

$$[\![\sigma_y]\!] = 0 \qquad \text{and} \qquad [\![\tau_{xy}]\!] = 0 \tag{140}$$

and, thus, must be continuous at $y = w$ and $y = d - w$.

Conditions (139) and (140) have important implications: from $(139)_{1,3}$, we see that the pressure p must be linear in x, independent of z, and continuous at $y = w$ and $y = d - w$, from which we conclude that the difference $\sigma_y - p$ must be of the form

$$\sigma_y(y,t) - p(x,y,t) = k_1(t)x + k_2(t), \qquad -\infty < x < \infty, \qquad 0 \le y \le d, \qquad t \ge 0 \tag{141}$$

for some choice of time-dependent functions k_1 and k_2; moreover, using (141) in $(139)_1$ while recognizing from (121) and $(129)_4$ that τ_{xy} vanishes at $y = d/2$, we see that it must be of the form

$$\tau_{xy}(y,t) = k_1(t)\left(\frac{d}{2} - y\right), \qquad 0 \le y \le d, \qquad t \ge 0. \tag{142}$$

The following remarks are in order:

---

[4] The condition $\partial\hat{\sigma}/\partial\gamma \equiv 0$ is equivalent to the second order partial differential equation

$$\frac{\partial^2\check{\psi}_e(I_1, I_2)}{\partial I_1^2} + 3\frac{\partial^2\check{\psi}_e(I_1, I_2)}{\partial I_1 \partial I_2} + 2\frac{\partial^2\check{\psi}_e(I_1, I_2)}{\partial I_2^2} = 0$$

for $\check{\psi}_e$, the most general solution of which has the form (136).

- By (141), the total normal stress $\sigma_y - p$ acting on planes parallel to the channel walls does not vary from plane to plane. Any variation of the network part of that normal stress must therefore be compensated by a variation of the corresponding fluid part.
- If $k_1(t) > 0$ for a given time t, then, by (142), the total shear stress τ_{xy} acting on planes parallel to the channel walls varies linearly with the plane position, attaining its maximum and minimum at the planes in contact with the lower and upper walls, respectively. The locations of the maxima and minima switch if $k_1(t) < 0$.

Following Coleman et al. (1966), we now discuss how the time-dependent functions k_1 and k_2 can be obtained and show that k_1 can be identified with the applied pressure drop per unit length. To this end, we consider a gel slab lying between the planes $x = x_1$ and $x = x_2$, with $x_2 > x_1$, and observe that k_1 obeys the relation

$$k_1 = \frac{1}{d(x_2 - x_1)} \int_0^d ((\sigma_x - p)|_{x=x_2} - (\sigma_x - p)|_{x=x_1}) \, dy \tag{143}$$

obtained by using (141) and recalling that $\sigma_x = (\sigma_x - \sigma_y) + \sigma_y$ and that σ_x and σ_y are independent of x. The integral term of (143) represents total applied force, per unit length in the direction normal to the (x, y)-plane, at time t, in the x-direction, imposed on the gel slab. Thus, (143) shows that k_1 is the time varying applied force per unit volume in the direction of the interstitial fluid flow, that is, the driving force that produces the flow. In view of (141) one can see that $k_1(t)$, the negative of the pressure gradient in the direction of the fluid flow at time t, is given by $(p(x_1, y, t) - p(x_2, y, t))/(x_2 - x_1)$ and hence can be identified with the pressure drop per unit length at that time. Otherwise, k_2 can be obtained if the normal thrust per unit area at a point of one of the bounding walls, which is equal to the normal stress $\sigma_y - p$ at that point, is known as function of t.

We finalize the discussion regarding the mechanical force balance by adding the following remarks:

- In view of (141) and (142), we find from the mechanical force balance that the components $\sigma_y - p$ and τ_{xy} of the total stress $\boldsymbol{T}_n - p\boldsymbol{I}$ are determined by the time-dependent functions k_1 and k_2. Moreover, we see from (141) that

$$\left.\begin{array}{l} \sigma_x(y, t) - p(x, y, t) = \sigma_x(y, t) - \sigma_y(y, t) + k_1(t)x + k_2(t), \\ \sigma_z(y, t) - p(x, y, t) = \sigma_z(y, t) - \sigma_y(y, t) + k_1(t)x + k_2(t), \end{array}\right\} \tag{144}$$

for $-\infty < x < \infty$, $0 \le y \le d$, $t \ge 0$, and, thus, that to determine $\sigma_x - p$ and $\sigma_z - p$ requires knowledge of k_1 and k_2 and, in addition, of the normal stress differences $\sigma_x - \sigma_y$ and $\sigma_z - \sigma_y$. The situation is completely analogous to that encountered by Coleman et al. (1966) within the context of the channel flow of incompressible fluids.
- By (129), the normal stress differences $\sigma_x - \sigma_y$ and $\sigma_z - \sigma_x$ can be expressed as

$$\sigma_x - \sigma_y = \left(\frac{1 + \gamma^2 - \lambda^2}{\gamma \lambda}\right)\tau_{xy} \qquad \text{and} \qquad \sigma_x - \sigma_z = \frac{2\gamma^2 \hat{\alpha}_1(\gamma, \lambda)}{\lambda}. \tag{145}$$

We show in Appendix A that $(145)_1$ and $(145)_2$ hold for any isotropic gel of type considered here and therefore comprise universal relations for such gels.

7.2.3. Fluid content balance

We begin by obtaining the components of the solvent flux $\boldsymbol{J}$ in the underlying Cartesian frame. Recalling from $(139)_3$ that p is independent of z, and from (116) that $\det \boldsymbol{F} = \lambda$, we see from $(103)_2$ that the streamwise, spanwise, and transverse components of the solvent flux $\boldsymbol{J}$ are given by

$$J_x = -\upsilon \hat{m}(\lambda)\frac{\partial p}{\partial x}, \qquad J_y = -\upsilon \hat{m}(\lambda)\frac{\partial p}{\partial y}, \qquad \text{and} \qquad J_z = 0. \tag{146}$$

However, from (141), we see that the conditions in (146) can be expressed alternatively as

$$J_x = \upsilon \hat{m}(\lambda)k_1, \qquad J_y = -\upsilon \hat{m}(\lambda)\frac{\partial \sigma_y}{\partial y}, \qquad \text{and} \qquad J_z = 0. \tag{147}$$

Since λ and σ_y depend at most on y and t, we infer from (147) that the nontrivial components J_x and J_y of $\boldsymbol{J}$ can also depend at most on y and t. The analogous finding applies moreover to the streamwise and spanwise components of $\boldsymbol{v}$, as is evident from (124), and on this basis, we infer that $(102)_3$ reduces to

$$\frac{\partial(\upsilon J_y + \upsilon_y)}{\partial y} = 0, \tag{148}$$

which in combination with (126) leads to the requirement that

$$\overline{\ln \lambda} = -\frac{\partial(\upsilon J_y)}{\partial y} \tag{149}$$

hold throughout the channel, excepting the interfaces at $y = w$ and $y = d - w$. Turning to those interfaces and recalling that m is oriented in the direction of increasing y, we deduce from (109) and $(124)_3$ that the condition

$$[\![\upsilon J_y + \upsilon_y]\!] = 0 \tag{150}$$

is met at $y = w$ and $y = d - w$, which in conjunction with (124) leads to the finding that the condition

$$\llbracket v j_y \rrbracket = \left\llbracket \lambda \frac{\partial g}{\partial t} \right\rrbracket \tag{151}$$

is also met at $y = w$ and $y = d - w$. Integrating (148) and using the continuity condition (150), we thus conclude that $v j_y + v_y$ depends at most on t. Since the channel walls are impermeable and the gel adheres to them, we infer that $j_y = 0$ and $v_y = 0$ at $y = 0$ and $y = d$ and, hence, that the condition

$$v j_y + v_y = 0 \tag{152}$$

must apply throughout the channel. Thus, we deduce that fluid flow in the y-direction must be accompanied by the motion of the network in the opposing direction.

We finalize this subsection by emphasizing the most salient results obtained thus far:

- According to (147)$_1$, the pressure drop forces the interstitial fluid to flow along its direction of application. Moreover, the corresponding flow depends on the stretch λ through the response function $\hat{m}(\lambda)$ determining the mobility of the liquid. It is natural to expect that $\hat{m}(\lambda)$ decreases with the polymer volume fraction and hence, by (137), $\hat{m}(\lambda)$ increases with λ. Thus, the higher the stretch, the higher the fluid flow in the direction of the pressure drop.

- As (147)$_2$ shows, the applied pressure drop can also drive interstitial fluid flow in the direction perpendicular to the channel walls. However, this occurs indirectly thorough the gradient of the network normal stress σ_y, with the consequence that the interstitial fluid flows from higher to lower values of σ_y. By (152), any such flow must be accompanied by the network motion in the opposite direction and ceases to exist when the steady state is reached since $v_y = 0$ vanishes at that stage of the process. We therefore infer that σ_y must be homogeneous under steady-state conditions.

- When a pressure drop is applied to a gel placed between two fixed walls, mechanical equilibrium, which is attained instantaneously if inertial effects are negligible assumed herein, imposes the linear profile for τ_{xy} and the relation between σ_y and p presented in (141)–(142). In particular, (141)–(142) show that for all time: τ_{xy} and is completely specified by the applied pressure drop, as is $\partial p / \partial x$, which provides the driving force for fluid flow in the x-direction; $\partial p / \partial y$, which provides the driving force for fluid flow in the y-direction, is equal to $\partial \sigma_y / \partial y$. When the pressure drop is applied suddenly, τ_{xy} and $\partial p / \partial x$ are time-independent whereas σ_y and p generally evolve with time.

7.2.4. Configurational force balance

Referring to (118)$_1$, (128), and (137), we find that the interface condition (109)$_4$ simplifies to

$$\llbracket \Psi \rrbracket - \sigma_y \llbracket \lambda \rrbracket - \tau_{xy} \llbracket \gamma \rrbracket = 0, \tag{153}$$

where, recalling (71), (73), (101), (131), and (134), Ψ, σ_y, and τ_{xy} are given by

$$\Psi = \hat{\varphi}(\gamma, \lambda), \qquad \sigma_y = \hat{\sigma}(\gamma, \lambda) = \frac{\partial \hat{\varphi}(\gamma, \lambda)}{\partial \lambda}, \qquad \text{and} \qquad \tau_{xy} = \hat{\tau}(\gamma, \lambda) = \frac{\partial \hat{\varphi}(\gamma, \lambda)}{\partial \gamma}, \tag{154}$$

with $\hat{\varphi}$ defined according to (133).

7.2.5. Summary of the governing equations

We now gather the equations we will use to describe the behavior of a gel placed between two fixed walls under the action of an applied pressure drop. These consist of (117), (124)$_2$, (137), (138), (141), (142), (152), and (153), augmented by the constitutive relations (146) and (154)$_2$ for σ_y, τ_{xy}, and Ψ and j_y, respectively. Due to symmetry, only the lower half $0 \le y \le d/2$ of the channel need be considered. Specifically, we consider

$$\left. \begin{aligned} \sigma_y &= p + k_1 x + k_2, \\ \tau_{xy} &= k_1 \left(\frac{d}{2} - y \right), \\ v_y + v j_y &= 0, \end{aligned} \right\} \tag{155}$$

for all $0 < y \le d/2$ and $t > 0$,

$$\left. \begin{aligned} \lambda \left(1 + \frac{\partial g}{\partial y} \right) &= 1, \\ \gamma \left(1 + \frac{\partial g}{\partial y} \right) &= -\frac{\partial f}{\partial y}, \\ v_y &= -\lambda \frac{\partial g}{\partial t}, \\ \lambda \phi &= \phi_0, \\ \sigma_y &= \hat{\sigma}(\gamma, \lambda), \\ \tau_{xy} &= \hat{\tau}(\gamma, \lambda), \\ j_y &= -v \hat{m}(\lambda) \frac{\partial p}{\partial y}, \end{aligned} \right\} \tag{156}$$

for all $0 < y < w$ and $w < y < d/2$ and $t > 0$, and

$$\left.\begin{aligned}
[\![\phi]\!]\dot{w} + [\![\phi v_y]\!] &= 0, \\
[\![\Psi]\!] - \tau_{xy}[\![\gamma]\!] - \sigma_y[\![\lambda]\!] &= 0,
\end{aligned}\right\} \tag{157}$$

for $y = w$ and $t > 0$.

The system (155)–(157) must be supplemented with appropriate boundary and initial conditions. The first set of conditions, which follow from the boundary and symmetry conditions (113) and (114), are given by

$$\left.\begin{aligned}
f(0, t) &= 0, \\
\frac{\partial f(y, t)}{\partial y}\bigg|_{y=\frac{d}{2}} &= 0, \\
g(0, t) &= 0, \\
g\left(\frac{d}{2}, t\right) &= 0,
\end{aligned}\right\} \tag{158}$$

for all $t \geq 0$. As for the second set of conditions, we assume that the initial state of the gel is taken as the state prior to the application of the pressure drop, in which case the gel is assumed to be characterized by a uniform composition described by the constant value ϕ_0 of the polymer volume fraction. Thus, the initial conditions are given by

$$\left.\begin{aligned}
g(y, 0) &= 0, \\
w(0) &= w_0,
\end{aligned}\right\} \tag{159}$$

for all $0 \leq y \leq d/2$, with $0 \leq w \leq d/2$. Since interfaces are of course absent in the initial state of the gel, it seems sensible to assume that a new phase may initiate at either $w_0 = 0$ or $w_0 = d/2$. This means that a new phase may appear in two privileged locations, namely at the gel surface which is contact to the channel wall or inside the gel in the center of the channel.[5] It is therefore possible to use the system of equations described above to investigate the kinetics of phase transition by assuming that the interface may appear either at $y = 0$ or at $y = d/2$. An analysis of this kind is outside the scope of the present work but will be addressed elsewhere.

Given the applied pressure drop per unit length k_1, the polymer volume fraction ϕ_0 in the reference state, the volume v occupied by a fluid molecule, and the constitutive responses $\hat{\varphi}$, from which $\hat{\tau}$, $\hat{\sigma}$, and Ψ are obtained, and $\hat{m}$, the system (155)–(157), supplemented with the boundary and initial conditions (158) and (159), yield a system of equations to be solved for f, g, ϕ, and p, all of which vary with position and time, and w, which varies only on time. Additionally, the pressure p is determined to within the time-dependent function k_2.

At this juncture, it is worth highlighting some features depicted by the system (155)–(157) that are associated with the response functions $\hat{\tau}$ and $\hat{\sigma}$:

(i) An immediate calculation using (155) and (156) allows us to conclude that g must obey the equation

$$\lambda \frac{\partial g}{\partial t} = -v^2 \hat{m}(\lambda)\left(\frac{\partial \hat{\sigma}(\gamma, \lambda)}{\partial \gamma}\frac{\partial \gamma}{\partial y} + \frac{\partial \hat{\sigma}(\gamma, \lambda)}{\partial \lambda}\frac{\partial \lambda}{\partial y}\right) \tag{160}$$

for all $0 < y < w$ and $w < y < d/2$ and $t > 0$. This implies that the system (155)–(157) admits the trivial solution characterized by $g \equiv 0$ only if the constitutive response function $\hat{\sigma}$ determining the network stress σ_y is unaffected by the amount of shear γ, that is, whenever $\partial \hat{\sigma}/\partial \gamma \equiv 0$. In this case, (155)–(157) reduces to a system for the quantities f and w. This is the situation found for the family of elastic energy densities of the form (136), which includes both neo-Hookean and Mooney models for nonlinear elastic networks. In this case, the gel undergoes a inhomogeneous shear deformation while the fluid permeates exclusively along the direction of the applied pressure drop. Further, that trivial solution is unstable whenever $\partial \hat{\sigma}/\partial \lambda < 0$ at $\lambda = 1$, since in this case a small and smooth perturbation of $g \equiv 0$ grows without limit over time, as shown in Appendix E.

(ii) Under the proviso that τ_{xy} can be treated as an independent constitutive variable, the system (155)–(157) can be reformulated in such way that g, ϕ, p, and w can be solved for independent of f, which in turn is obtained from the knowledge of g and w, as shown in Appendix E. Consistent with previous remarks, this is exactly the situation encountered in the present paper because here interfaces emerge exclusively due of volume transitions. Consequently, the mapping $\hat{\tau}(\cdot, \lambda)$ can be inverted for each λ, thereby allowing the replacement of γ by τ_{xy} as the independent constitutive variable, as shown in Appendix B.

7.3. Instantaneous and steady-state responses

From here onward, we suppose that k_1 satisfies

$$k_1 = \text{constant} > 0 \tag{161}$$

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[5] This kind of consideration is often found in both theoretical and experimental studies on the kinetics of volume phase transition in gels. See, for instance, Hirotsu (1993), Tomari and Doi (1995), and Suzuki and Ishii (1999), among others.

and is applied instantaneously at $t = 0$. We then next investigate two limiting responses of the gel, namely the instantaneous and steady responses exhibited in the limits as time goes to zero and infinity, respectively.

We assume that the instantaneous response is characterized by the insufficient time for the fluid to flow in the direction perpendicular to plates and hence replace $(156)_3$ and $(157)_1$ by $v_y \equiv 0$ and $\dot{w} \equiv 0$, respectively. On the other hand, the steady response corresponds to time-independent solutions of (155)–(157), which in turn imply that $v_y \equiv 0$ and $\dot{w} \equiv 0$. Notice that although the conditions on v_y and w defining the instantaneous and steady responses are the same, they are distinct in nature and consequently have different implications. As a matter of fact:

- The conditions $v_y = -v j_y \equiv 0$ and $\dot{w} \equiv 0$ defining the instantaneous response originate from the assumption that immediately after the application of the pressure drop fluid flow in the direction perpendicular to the plates is insignificant regardless the driving force $\partial p / \partial y$ that is instantaneously generated. Under this condition, there is not enough time for the initial composition of the gel to change, with the consequence that the gel behaves like an incompressible elastic solid undergoing an inhomogeneous shear deformation.
- On the other hand, the conditions $v_y \equiv 0$ and $\dot{w} \equiv 0$ defining the steady response come from the assumption that, with the passage of time, $\partial p / \partial y$ decays until it eventually vanishes and, thus, fluid flow ceases. Under this condition, the gel behaves like a compressible elastic solid undergoing an inhomogeneous shear deformation upon which an inhomogeneous stretch is superimposed.

In Appendix F, we show that the instantaneous response can be seen as a first order approximation of the gel response for very short times, wherein the observation time scale t_o is much smaller than t_*, the intrinsic time scale associated with fluid transport. The latter can be estimated by

$$t_* = \frac{\lambda_c d}{2v^2 m_c a_c}, \tag{162}$$

where λ_c, m_c, and a_c are characteristic sizes for λ, $\hat{m}$, and $\partial p / \partial y$. On the other hand, the steady response can be viewed as a first order approximation of the gel response for very long times, wherein the observation time scale t_o is much greater than the intrinsic time scale t_*.

7.3.1. Instantaneous response

We now seek to describe the behavior of the gel immediately after the sudden application of pressure drop per unit length $k_1 > 0$, that is, in the limit $t \to 0^+$, by using (155)–(156). We assume that the instantaneous response does not involve interfaces. The conditions in (157) are therefore not germane under this limit.

For very short times, there is insufficient time for the fluid to flow and hence the gel composition is static. Hence, $\phi(\cdot, 0^+) \equiv \phi_0$ and, by $(156)_1$, $\lambda(\cdot, 0^+) \equiv 1$, from which, with reference to $(117)_2$, it follows that $g(\cdot, 0^+) \equiv 0$. The instantaneous response of the gel therefore involves an inhomogeneous shear deformation described by the unknown function $f(\cdot, 0^+)$, which in view of $(117)_2$ and $(113)_1$, can be expressed in terms of the function $\gamma(\cdot, 0^+)$ giving the amount of shear for very short times. To obtain $\gamma = \gamma(\cdot, 0^+)$, we must invert the equation

$$\hat{\tau}(\gamma(y), 1) = k_1 \left(\frac{d}{2} - y \right), \qquad 0 \leq y \leq \frac{d}{2}, \tag{163}$$

obtained by combining $(155)_2$ and $(156)_2$. In this connection, it is useful to notice that since the right-hand side of (163) take values on the interval $[0, k_1 d/2]$ and $\hat{\tau}(\gamma, 1) = \hat{G}(\gamma, 1)\gamma$, with $\hat{G}(\gamma, 1) > 0$, $\gamma(y, 0^+)$ takes values on the interval $[0, \gamma_m)$, where $\gamma_m > 0$ denotes the liming value of the amount of shear.[6]

We now discuss the existence and uniqueness of solutions $\gamma = \gamma(\cdot, 0^+)$ to (163). For each $0 \leq y \leq d/2$, (163) has a solution $\gamma(y, 0^+) \geq 0$ provided that the interval $[0, k_1 d/2]$ be contained in the image set of $\hat{\tau}(\cdot, 1)$. Bearing in mind that $\hat{\tau}(0, 1) = 0$, we see that a condition sufficient to ensure that this occurs for any $k_1 > 0$ is that

$$\lim_{\gamma \to \gamma_m} \hat{\tau}(\gamma, 1) = \infty. \tag{164}$$

The corresponding solution will be unique if $\hat{\tau}(\gamma, 1) = \hat{G}(\gamma, 1)\gamma$ depends monotonically on γ, which is equivalent, since $\hat{G}(\gamma, 1) > 0$, to stipulating that the inequality

$$\frac{\partial \hat{\tau}(\gamma, 1)}{\partial \gamma} > 0 \tag{165}$$

holds for all $\gamma \in [0, \gamma_m)$. In this case, for each $0 \leq y \leq d/2$, the solution $\gamma(y, 0^+)$ of (163) can be written as

$$\gamma(y, 0^+) = \tilde{\gamma}(\tau(y), 1), \qquad \text{with} \qquad \tau(y) = k_1 \left(\frac{d}{2} - y \right), \tag{166}$$

where $\tilde{\gamma}(\cdot, 1)$ is the inverse of $\hat{\tau}(\cdot, 1)$. Notice that (165) and (166) imply that $\gamma(y, 0^+)$ decreases with y on the interval $[0, d/2]$, with its maximum and minimum values being attained at $y = 0$ and $y = d/2$, respectively. The following remarks are in order:

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[6] We allow γ_m to be finite to account for elastic models that incorporate limiting chain extensibility effects. Otherwise, γ_m may tend to infinity.

- Conditions (164) and (165) were introduced by Horgan et al. (2002) and Saccomandi (2004), who showed that they are satisfied by the power-law elastic model introduced by Knowles (1977) with the provision that the stiffening parameter n be strictly greater than 1/2, and by the limiting-chain extensibility elastic model proposed by Gent (1996). See Appendix A for details.
- The existence and uniqueness conditions (164) and (165) are equivalent, respectively, to the conditions

$$\lim_{\gamma \to \gamma_m} \hat{G}(\gamma, 1) > 0 \quad \text{and} \quad \frac{\partial \hat{G}(\gamma, 1)}{\partial \gamma} \gamma + \hat{G}(\gamma, 1) > 0, \qquad 0 \le \gamma \le \gamma_m. \tag{167}$$

These conditions are satisfied for elastic materials for which $\partial \hat{G}(\gamma, 1)/\partial \gamma > 0$, that is, for strain-stiffening elastic materials.

We now turn to the calculation of the instantaneous pressure field $p(\cdot, 0^+)$. First, we observe that, by (156)$_5$, the normal stress σ_y must satisfy the condition

$$\sigma_y(y, 0^+) = \hat{\sigma}(\gamma(y, 0^+), 1), \tag{168}$$

with $\gamma(\cdot, 0^+)$ given by (163). By (155)$_1$, this implies that $p(\cdot, 0^+)$ must be such that

$$p(x, y, 0^+) = \hat{\sigma}(\gamma(y, 0^+), 1) - k_1 x - k_2(0^+). \tag{169}$$

Under the validity of conditions (164) and (165), we have shown that the instantaneous gel response to a suddenly-applied pressure drop is such that the gel undergoes an inhomogeneous shear deformation with amount of shear $\gamma = \gamma(\cdot, 0^+)$ given by (163), which by its turn determines the network normal stress σ_y and pressure p by (168) and (169), respectively.

The above-mentioned expressions for σ_y and p can next be used to show that

$$\frac{\partial p}{\partial y} = \frac{\partial \sigma_y}{\partial y} = -k_1 \frac{\partial \hat{\sigma}(\gamma, 1)}{\partial \gamma} \frac{\partial \tilde{\gamma}(\tau, 1)}{\partial \tau}, \tag{170}$$

from which, bearing in mind that $k_1 > 0$ and $\partial \tilde{\gamma}/\partial \tau > 0$, we arrive at the relation

$$\text{sgn}\left(\frac{\partial p}{\partial y}\right) = -\text{sgn}\left(\frac{\partial \hat{\sigma}(\gamma, 1)}{\partial \gamma}\right), \tag{171}$$

where "sgn" defined such that $\text{sgn}(u) = u/|u|$ for any $u \ne 0$ is the sign function. Bearing in mind that $-\partial p/\partial y$ is the driving force for liquid flow in the y-direction, we thus conclude that:

- If $\partial \hat{\sigma}(\gamma, 1)/\partial \gamma > 0$ for each γ satisfying $0 \le \gamma < \gamma_m$, then the normal network stress σ_y decreases monotonically from its maximum value at $y = 0$ to its minimum value at $y = d/2$. In this case, the flow of liquid in the spanwise direction is directed from the boundary walls toward the center of the channel.
- If $\partial \hat{\sigma}(\gamma, 1)/\partial \gamma = 0$ for each γ satisfying $0 \le \gamma < \gamma_m$, then the normal network stress σ_y is constant. In this case, there is no flow of liquid in the spanwise direction.
- If $\partial \hat{\sigma}(\gamma, 1)/\partial \gamma < 0$ for each γ satisfying $0 \le \gamma < \gamma_m$, then the normal network stress σ_y increases monotonically from its minimum value at $y = 0$ to its maximum value at $y = d/2$. In this case, the flow of liquid in the spanwise direction is directed from the center of the channel toward the boundary walls.

For elastic energies that depend on I_1 only, which define the class of the generalized neo-Hookean elastic materials, it follows from (131)$_1$ and (132) that

$$\hat{\sigma}(\gamma, 1) = \hat{G}(\gamma, 1) - \hat{\pi}(1). \tag{172}$$

and, thus, that $\partial \hat{\sigma}(\gamma, 1)/\partial \gamma = \partial \hat{G}(\gamma, 1)/\partial \gamma$. For such materials, the discussion above can therefore be given in terms of the response function $\hat{G}(\cdot, 1)$ that determines the generalized shear modulus.

The essential results concerning the short-time response of the gel can be summarized as follows:

1. The shear stress τ_{xy} requires the generation of an inhomogeneous profile for the shear amount γ which, like τ_{xy}, decreases monotonically on the lower half of the channel, with maximum and minimum values attained at $y = 0$ and $y = d/2$, respectively.
2. The normal network stress σ_y is determined, modulo a constant, by the amount of shear γ via the elastic response of the gel. For instance, if the elastic response is such σ_y increases monotonically with γ, the corresponding profile is such that σ_y decreases monotonically on the interval $[0, d/2]$. In this case, liquid flow in the y-direction will be driven towards the center of the channel. Otherwise, if σ_y decreases monotonically with γ, the corresponding profile is such σ_y increases monotonically on the interval $[0, d/2]$ and liquid flow in the y-direction will be driven towards the fixed walls of the channel. If σ_y is unaffected by γ, the σ_y profile is constant and hence there will be no driving force for liquid flow in the y-direction.
3. For generalized neo-Hookean elastic materials, the flow of liquid in the spanwise direction is directed towards the channel center and towards the channel walls for strain-stiffening and strain-slackening elastic materials, respectively.

7.3.2. Steady-state response

We now use (155)–(157) to investigate the steady-state behavior of the gel. In this limit, which is achieved as $t \to \infty$, all quantities are time-independent. Thus, in particular, the time derivative of g vanishes in steady state and we see from (156)$_3$ that $v_y = 0$. Furthermore, we see from (155)$_3$ that $J_y = 0$. Hence, with reference to (156)$_7$ and (155)$_1$, we find that $\partial p / \partial y = \partial \sigma_y / \partial y = 0$ away from the interface. Recalling the continuity condition (140)$_2$, we conclude from (155)$_{1,2}$ that at steady state σ_y and τ_{xy} must obey

$$\sigma_y(y) = \sigma_* \qquad \text{and} \qquad \tau_{xy}(y) = k_1\left(\frac{d}{2} - y\right), \qquad 0 \le y \le \frac{d}{2}, \tag{173}$$

where σ_*, like k_1, is constant. Thus, to characterize the steady state in the smooth case, we must obtain the normal network stress σ_* and, for each $y \in [0, d/2]$, the shear strain $\gamma(y)$ and the stretch $\lambda(y)$ from the conditions

$$\hat{\sigma}(\gamma(y), \lambda(y)) = \sigma_* \qquad \text{and} \qquad \hat{\tau}(\gamma(y), \lambda(y)) = k_1\left(\frac{d}{2} - y\right) \tag{174}$$

arising from (156)$_{5,6}$, (173), and the previously derived integral condition (123) for the stretch, which we repeat here for convenience:

$$\int_0^{d/2} \frac{\mathrm{d}y}{\lambda(y,t)} = \frac{d}{2}. \tag{175}$$

Additionally, if interfaces are present, it is necessary to determine their locations, as well as the limiting values of γ and λ from each side of the interfaces, and to ensure satisfaction of the continuity conditions

$$[\![\hat{\sigma}(\gamma(w), \lambda(w))]\!] = 0, \qquad [\![\hat{\tau}(\gamma(w), \lambda(w))]\!] = 0, \tag{176}$$

which follow (156)$_{5,6}$, (173), and (174), and the configurational force balance

$$[\![\hat{\varphi}(\gamma(w), \lambda(w))]\!] - \hat{\sigma}(\gamma(w), \lambda(w))[\![\lambda(w)]\!] - \hat{\tau}(\gamma(w), \lambda(w))[\![\gamma(w)]\!] = 0, \tag{177}$$

which follows from (157)$_2$, (173), and (174). Therefore, when interfaces are present, the system (173)–(177) consists of eight conditions for determining γ and λ as functions of the spanwise coordinate y, the normal stress σ_*, the location w of the interface in the lower half channel (and, thus, the location $d - w$ of the interface in the upper half channel), and the corresponding limiting values,

$$\overset{\pm}{\gamma} \qquad \text{and} \qquad \overset{\pm}{\lambda}, \tag{178}$$

of γ and λ. Otherwise, when interfaces are not present, (173)–(175) provide three conditions for determining λ, γ, and σ_*. In either case, having determined γ and λ, (113) and (117) can be used to evaluate f and g.

We now specialize the system (174)–(177) to situations in which the shear stress response $\hat{\tau}(\cdot, \lambda)$ is invertible as a function of γ for each admissible choice of λ, in which case the shear stress τ_{xy} can be treated as an independent constitutive variable. See Appendix E for additional discussion. Recalling that both τ_{xy} and γ are non-negative in the lower half of the channel, we accordingly assume that the response function $\hat{\tau}(\cdot, \lambda)$ obeys the conditions

$$\lim_{\gamma \to \gamma_m} \hat{\tau}(\gamma, \lambda) = \infty \qquad \text{and} \qquad \frac{\partial \hat{\tau}(\gamma, \lambda)}{\partial \gamma} > 0 \tag{179}$$

for all $\gamma \in [0, \gamma_m)$ each admissible λ, with the limiting value γ_m possibly depending on λ. Under these conditions, the relation $\tau_{xy} = \hat{\tau}(\gamma, \lambda)$ can be inverted for γ for any $\tau_{xy} \ge 0$, and, thus, in particular, for any τ_{xy} satisfying $0 \le \tau_{xy} \le k_1 d/2$. Introducing the partial inverse $\tilde{\gamma}$ of $\hat{\tau}$, we can therefore write

$$\gamma = \tilde{\gamma}(\tau_{xy}, \lambda) \tag{180}$$

for any $\tau_{xy} \ge 0$. We can thus consider τ_{xy} as an independent constitutive variable and define functions $\tilde{\varphi}$ and $\tilde{\sigma}$ of τ_{xy} and λ through

$$\tilde{\varphi}(\tau_{xy}, \lambda) := \hat{\varphi}(\tilde{\gamma}(\tau_{xy}, \lambda), \lambda) \qquad \text{and} \qquad \tilde{\sigma}(\tau_{xy}, \lambda) := \hat{\sigma}(\tilde{\gamma}(\tau_{xy}, \lambda), \lambda). \tag{181}$$

It is also convenient to introduce a function $\tilde{\Phi}$ of τ_{xy} and λ through the relation

$$\tilde{\Phi}(\tau_{xy}, \lambda) = \tilde{\varphi}(\tau_{xy}, \lambda) - \tau_{xy}\tilde{\gamma}(\tau_{xy}, \lambda), \tag{182}$$

which can be identified as the negative of the partial Legendre transformation of $\tilde{\varphi}$ with respect to τ_{xy}. Direct calculations then show $\tilde{\Phi}$ serves as a potential for the functions $\tilde{\sigma}$ and $\tilde{\gamma}$ defined in (181):

$$\tilde{\sigma}(\tau_{xy}, \lambda) = \frac{\partial \tilde{\Phi}(\tau_{xy}, \lambda)}{\partial \lambda}, \qquad \tilde{\gamma}(\tau_{xy}, \lambda) = \frac{\partial \tilde{\Phi}(\tau_{xy}, \lambda)}{\partial \tau_{xy}}. \tag{183}$$

With these definitions, we replace (173) and (174) by

$$\tilde{\sigma}(\tau_{xy}(y), \lambda(y)) = \sigma_* \qquad \text{and} \qquad \tau_{xy}(y) = k_1\left(\frac{d}{2} - y\right), \tag{184}$$

with y belonging to $(0, d/2] \setminus \{w\}$. Additionally, we may use (180)–(182) to express (176)–(177) in the form

$$\tilde{\sigma}\left(k_1\left(\frac{d}{2} - w\right), \overset{+}{\lambda}\right) = \sigma_*, \qquad \tilde{\sigma}\left(k_1\left(\frac{d}{2} - w\right), \overset{-}{\lambda}\right) = \sigma_*, \qquad \tilde{\Phi}\left(k_1\left(\frac{d}{2} - w\right), \overset{+}{\lambda}\right) - \tilde{\Phi}\left(k_1\left(\frac{d}{2} - w\right), \overset{-}{\lambda}\right) = \sigma_*(\overset{+}{\lambda} - \overset{-}{\lambda}). \tag{185}$$

Granted knowledge of the field λ, we may use (180) to determine the field γ. The following remarks are in order:

- Using (A.8) and (A.9), we find that the conditions appearing in (179) are satisfied for the power-law model of Knowles (1977) for any choice of the power n satisfying $n > 1/2$ and also for the limiting-chain extensibility elastic model of Gent (1996).
- Recalling from (156)$_4$ that $\lambda = \phi_0/\phi$, we see that λ must take values in the interval (ϕ_0, ∞), with $\lambda \to \phi_0$ as $\phi \to 1$ and $\lambda \to \infty$ as $\phi \to 0$. For a model such as Gent's (Gent, 1996) that incorporate limiting chain extensibility, λ takes values in the interval (ϕ_0, λ_m) instead, where the limiting value $\lambda_m < \infty$ for the stretch λ may depend on γ and hence on the shear stress τ_{xy}.
- From (185)$_{1,2}$, we find that interfaces cannot be present if $\tilde{\sigma}(\tau_{xy}, \cdot)$ depends monotonically on its argument for all choices of τ_{xy}.
- From (183)$_1$, we find that (185)$_3$ can be written as

$$\int_{\lambda}^{\overset{+}{\lambda}} \tilde{\sigma}\left(k_1\left(\frac{d}{2} - w\right), \lambda\right) d\lambda = \sigma_*(\overset{+}{\lambda} - \overset{-}{\lambda}), \tag{186}$$

from which we deduce that the configurational force balance yields the Maxwell equal area rule and identify σ_* as the Maxwell stress. See Appendix D.
- By the chain rule, (181)$_2$, and (184), we obtain the identity

$$\frac{\partial \tilde{\sigma}(\tau_{xy}(y), \lambda)}{\partial \lambda}\Bigg|_{\lambda = \lambda(y)} \frac{\partial \lambda(y)}{\partial y} = k_1 \frac{\partial \tilde{\sigma}(\tau, \lambda)}{\partial \tau}\Bigg|_{\tau = \tau_{xy}(y)}, \qquad y \neq w. \tag{187}$$

Granted that $\partial \tilde{\sigma}(\tau_{xy}, \lambda)/\partial \lambda > 0$ for $0 \leq y < w$ and $w < y \leq d/2$, we thus see that the sign of $\partial \lambda/\partial y$ is determined by the sign of $\partial \tilde{\sigma}(\tau_{xy}, \lambda)/\partial \tau_{xy}$. In particular, if $\tilde{\sigma}$ is independent of τ_{xy}, so that $\partial \tilde{\sigma}/\partial \tau_{xy} \equiv 0$, then λ is such that $\partial \lambda/\partial y \equiv 0$. As already remarked, this condition holds for the family of elastic energy densities of the type (136), a family that includes both neo-Hookean and Mooney models for nonlinear elastic networks.

From the foregoing discussion, it is evident that the properties of the response function $\tilde{\sigma}$ are central to the nature of the solutions of (185). If, for instance, $\tilde{\sigma}(\tau_{xy}, \cdot)$ is a monotonic function of λ, the interface conditions in (185) are vacuous and, hence, can be disregard altogether. Under these circumstances, the stretch profile is smooth. Otherwise, a stretch profile exhibits a discontinuity corresponding to the presence of an interface at $y = w$ only if $\tilde{\sigma}(\tau_{xy}, \cdot)$ is a nonmonotonic function of λ for some admissible choice of τ_{xy}. This lead to the need to consider two situations according to whether or not the stretch profile is continuous.

Henceforth, we assume that $\tilde{\sigma}(\tau_{xy}, \cdot)$ complies the conditions

$$\lim_{\lambda \to \phi_0} \tilde{\sigma}(\tau_{xy}, \lambda) = -\infty \qquad \text{and} \qquad \lim_{\lambda \to \lambda_m} \tilde{\sigma}(\tau_{xy}, \lambda) = \infty, \tag{188}$$

which may be seen as a result of the following provisions involving the osmotic $\hat{\pi}$ and elastic $\tilde{\sigma}_e$ contributions to $\tilde{\sigma}$:

- The osmotic contribution $\hat{\pi}$ to $\tilde{\sigma}$ tends to ∞ as λ tends to ϕ_0 and remains bounded as λ tends to λ_m.
- For each admissible value of τ_{xy}, the elastic contribution $\tilde{\sigma}_e(\tau_{xy}, \cdot)$ to $\tilde{\sigma}$ tends to ∞ as λ tends to λ_m and remains bounded as λ tends to ϕ_0.

These stipulations assure that, given τ_{xy}, the equation $\hat{\sigma}(\tau_{xy}, \lambda) = \sigma_*$ has a solution λ, generally dependent on τ_{xy}, for any choice of σ_*. A detailed discussion of this matter is provided in Appendix C.

Further, we assume that (179) holds and hence base the subsequent development on (175), (184), and (185). We consider continuous and discontinuous solutions of those equations. Solutions are continuous only if $\tilde{\sigma}(\tau_{xy}, \cdot)$ is monotonic for each τ_{xy} satisfying $0 \leq \tau_{xy} \leq k_1 d/2$ but are discontinuous in the absence of that monotonicity requirement.

7.3.2.1. Continuous solutions. For smooth solutions, the stretch λ is completely determined by solving (184) subject to (175). To obtain such a solution, we begin by observing that, since $\tilde{\sigma}(\tau_{xy}, \cdot)$ is monotonic for each τ_{xy} satisfying $0 \leq \tau_{xy} \leq k_1 d/2$ and (188) holds, the equation $\tilde{\sigma}(\tau_{xy}, \lambda) = k$ be inverted to give λ as a function of τ_{xy} for any choice of k. In other words, there exists a function $\tilde{\lambda}$ such that

$$\lambda = \tilde{\lambda}(\tau_{xy}, k) \tag{189}$$

for any choice of k. For each τ_{xy} in $0 \leq \tau_{xy} \leq k_1 d/2$, the function $\tilde{\lambda}(\tau_{xy}, \cdot)$ is the inverse of $\tilde{\sigma}(\tau_{xy}, \cdot)$. This being the case, λ and σ_* must be found by solving the integral equation

$$\int_0^{d/2} \frac{dy}{\tilde{\lambda}(\tau_{xy}(y), k)} = \frac{d}{2}, \tag{190}$$

with τ_{xy} being given by (184)$_2$, for σ_* and then using (184)$_2$ and (189) to determine λ at each y in the interval $[0, d/2]$. Observe that left-hand side of (190) can be considered as a function, say h, of k, defined such that

$$h(k) := \int_0^{d/2} \frac{dy}{\tilde{\lambda}(\tau_{xy}(y), k)}. \tag{191}$$

The function h is positive since the integrand is positive. Further, it has following properties:

1. Since, by virtue of the condition $\partial \tilde{\sigma}(\tau_{xy}, \lambda)/\partial \lambda > 0$ that is met for all λ of interest (see Appendices B and C), $\tilde{\lambda}(\tau_{xy}, \cdot)$ is an increasing function of its argument for each τ_{xy} satisfying $0 \le \tau_{xy} \le k_1 d/2$, and, hence, h decreases as k increases.
2. Since $\tilde{\lambda}(\tau_{xy}, k)$ tends to $\phi_0 < 1$ as k tends to $-\infty$, $h(k)$ tends to a limit that exceeds $d/2$ as k tends to $-\infty$.
3. Since, for models in which chain extensibility is not limited, $\tilde{\lambda}(\tau_{xy}, k)$ tends to ∞ as k tends to ∞, $h(k)$ tends to a limit that is less than $d/2$ as k tends to ∞.
4. Since, for models in which chain extensibility is limited, $\tilde{\lambda}(\tau_{xy}, k)$ tends to $\lambda_m(\tau_{xy}) < \lambda_m(0)$, with $\lambda_m(0) > 1$, as k tends to ∞, $h(k)$ tends to a limit that is less than $d/2$ as k tends to ∞.

These properties guarantee the existence and uniqueness of solutions to (190) and, using (173)$_2$, deliver a unique expression for λ through (189). With reference to (187), if $\partial \tilde{\sigma}(\tau_{xy}, \lambda)/\partial \tau_{xy} > 0$ everywhere, then λ increases with y, from its minimum close to the plate to its maximum in the center of the channel. From $\lambda = \phi_0/\phi$, this is accompanied by polymer accumulation near the channel walls and depletion near the center of the channel. The opposite takes place if $\partial \tilde{\sigma}(\tau_{xy}, \lambda)/\partial \tau_{xy} < 0$ everywhere. These results are consistent with the calculation of the driving force for very short times.

7.3.2.2. Discontinuous solutions. For a discontinuous solution, the unknown stretch λ, constant normal stress σ_*, and the position w of the interface in the lower half $0 \le y \le d/2$ of the channel are governed by (175), (184), and (185). Leaving a detailed analysis of the issues of existence and uniqueness of solutions to this system of equations to another work, we content ourselves with the following remarks:

- The interface at $y = w$ separates the gel occupying the lower half of the channel into two phases, the low-stretch phase within which λ takes values in the interval

$$\mathcal{L} := (\phi_0, \min\{\overset{-}{\lambda}, \overset{+}{\lambda}\}) \tag{192}$$

and the high-stretch phase within which λ takes values in the interval

$$\mathcal{H} := (\max\{\overset{-}{\lambda}, \overset{+}{\lambda}\}, \lambda_m). \tag{193}$$

In view of the relation $\lambda = \phi_0/\phi$, the low- and high-stretch phases correspond to the high- and low-polymer-content phases.
- If the jump $[\![\lambda]\!]$ of the stretch λ is such that $\overset{-}{\lambda} < \overset{+}{\lambda}$, then the subsets $0 \le y < w$ and $w < y \le d/2$ of the lower half of the channel are filled by the low- and high-stretches phases, respectively. The opposite occurs otherwise.
- If $\partial \tilde{\sigma}/\partial \lambda > 0$ and $\partial \tilde{\sigma}/\partial \tau_{xy} > 0$ for all λ in $\mathcal{L} \cup \mathcal{H}$ and τ_{xy} satisfies $0 \le \tau_{xy} \le k_1 d/2$, then (187) implies that the jump $[\![\lambda]\!]$ of the stretch λ is such that

$$\overset{+}{\lambda} > \overset{-}{\lambda} \tag{194}$$

and, thus, that the subsets $0 \le y < w$ and $w < y \le d/2$ of the lower half of the channel are occupied by the low- and high-stretches phases of the gel and that the stretch λ is monotonically increasing in both phases.
- If $\partial \tilde{\sigma}/\partial \lambda > 0$ and $\partial \tilde{\sigma}/\partial \tau_{xy} < 0$ for all λ in $\mathcal{L} \cup \mathcal{H}$ and τ_{xy} satisfies $0 \le \tau_{xy} \le k_1 d/2$, then (187) implies that the jump $[\![\lambda]\!]$ of the stretch λ is such that

$$\overset{+}{\lambda} < \overset{-}{\lambda} \tag{195}$$

and, thus, that the subsets $0 \le y < w$ and $w < y \le d/2$ of the lower half of the channel are occupied by the high- and low-stretches phases and that the stretch λ is monotonically decreasing in both phases.

These observations suggest an iterative procedure for determining λ, σ_*, and w. Granted that $\partial \tilde{\sigma}/\partial \tau_{xy}$ is everywhere either positive or negative and that $\tilde{\sigma}(\tau_{xy}, \cdot)$ is a nonmonotonic function of λ for all $\tau_{xy} \in [0, k_1 d/2]$, that procedure relies on the introduction of the implicit function $V : (0, d/2) \to \mathbb{R}$ whose value $V(\zeta)$ at $\zeta \in (0, d/2)$ is defined through the following steps:

1. Set $\tau = k_1(d/2 - \zeta)$ and solve

$$\tilde{\sigma}\left(\tau, \lambda_1^\zeta\right) = \sigma_*^\zeta, \qquad \tilde{\sigma}\left(\tau, \lambda_2^\zeta\right) = \sigma_*^\zeta, \qquad \tilde{\Phi}\left(\tau, \lambda_2^\zeta\right) - \tilde{\Phi}\left(\tau, \lambda_1^\zeta\right) = \sigma_*^\zeta(\lambda_2^\zeta - \lambda_1^\zeta), \tag{196}$$

for λ_1^ζ, λ_2^ζ, and σ_*^ζ;
2. Evaluate the limiting values $\overset{\pm}{\lambda}_\zeta$ according to one the following alternatives:

$$
\begin{aligned}
\frac{\partial \tilde{\sigma}}{\partial \tau_{xy}} > 0 \quad &\implies \quad \left\{ \begin{array}{l} \overset{-}{\lambda}_\zeta = \min\{\lambda_1^\zeta, \lambda_2^\zeta\}, \\ \overset{+}{\lambda}_\zeta = \max\{\lambda_1^\zeta, \lambda_2^\zeta\}, \end{array} \right. \\[2mm]
\frac{\partial \tilde{\sigma}}{\partial \tau_{xy}} < 0 \quad &\implies \quad \left\{ \begin{array}{l} \overset{-}{\lambda}_\zeta = \max\{\lambda_1^\zeta, \lambda_2^\zeta\}, \\ \overset{+}{\lambda}_\zeta = \min\{\lambda_1^\zeta, \lambda_2^\zeta\}. \end{array} \right.
\end{aligned}
\tag{197}
$$

3. Obtain the profile for λ_ζ by solving

$$\tilde{\sigma}(\tau_{xy}, \lambda_\zeta) = \sigma_*^\zeta, \quad \text{with} \quad \tau_{xy} = k_1\left(\frac{d}{2} - y\right), \tag{198}$$

subject to the following constraints:

$$
\left.
\begin{aligned}
\frac{\partial \tilde{\sigma}}{\partial \tau_{xy}} > 0 \quad &\Longrightarrow \quad
\begin{cases}
\phi_0 < \lambda_\zeta < \bar{\lambda}_\zeta, & 0 \le y < \zeta, \\
\overset{+}{\lambda}_\zeta < \lambda_\zeta < \lambda_m, & \zeta < y \le d/2,
\end{cases} \\
\frac{\partial \tilde{\sigma}}{\partial \tau_{xy}} < 0 \quad &\Longrightarrow \quad
\begin{cases}
\bar{\lambda}_\zeta < \lambda_\zeta < \lambda_m, & 0 \le y < \zeta, \\
\phi_0 < \lambda_\zeta < \overset{+}{\lambda}_\zeta, & \zeta < y \le d/2.
\end{cases}
\end{aligned}
\right\}
\tag{199}
$$

4. Evaluate $V(\zeta)$ by

$$V(\zeta) = d - 2\left(\int_0^\zeta \frac{\mathrm{d}y}{\lambda_\zeta(y)} + \int_\zeta^{d/2} \frac{\mathrm{d}y}{\lambda_\zeta(y)}\right). \tag{200}$$

These steps can be interpreted as follows. Suppose that the lower half of the deformed gel contains an interface located at $y = \zeta \in (0, d/2)$. In this case, the required normal network stress must be equal to σ_*^ζ as obtained in step 1. By considering steps 2 and 3, one get the information needed to obtain the stretch profile corresponding to ζ, namely its limiting values at and distribution away from the interface, respectively given by $\overset{\pm}{\lambda}_\zeta$ and λ_ζ. In step 4, we evaluate $V(\zeta)$, the difference of two contributions distance between the plates after and before the deformation. The latter is equal to d only if ζ is a zero of V. Therefore, a solution (λ, σ_*, w) of (173)–(175) and (185) corresponds to a zero of the function V.

7.4. An example

We hereinafter consider a gel with elastic properties described by the Gent (1996) model and osmotic properties described by the Flory–Huggins model, delineated, for instance, by Doi (2013):

$$
\left.
\begin{aligned}
\tilde{\psi}_e(I_1, I_2) &= -\frac{G_0 J_m}{2} \ln\left(1 - \frac{I_1 - 3}{J_m}\right), \\
\hat{\psi}_m(C) &= k_B T\, C\,(\ln(1 - \phi) + \chi\phi), \qquad \text{with} \qquad \phi = \frac{\phi_0}{\phi_0 + \upsilon C}.
\end{aligned}
\right\}
\tag{201}
$$

In (201), G_0 is the shear modulus at the reference state, J_m is the phenomenological parameter that accounts for the finite extensibility of polymer in the network, k_B the Boltzmann constant, T is the absolute temperature, υ is the volume occupied by a solvent molecule, and χ is the Flory–Huggins interaction parameter. As Gent (1996) showed, the neo-Hookean elastic model is recovered from $(201)_1$ on passing to the limit as $J_m \to \infty$.

We now obtain the constitutive responses $\hat{\tau}$ and σ for the shear stress τ_{xy} and the normal stress σ_y corresponding to elastic and mixing responses given by (201). On using (130), (131), and (201), we conclude that $\hat{\tau}$ and $\hat{\sigma}$ specialize to

$$\hat{\tau}(\gamma, \lambda) = \frac{G_0 J_m \gamma}{J_m - (\lambda^2 + \gamma^2 - 1)} \tag{202}$$

and

$$\hat{\sigma}(\gamma, \lambda) = \frac{G_0 J_m \lambda}{J_m - (\lambda^2 + \gamma^2 - 1)} - \hat{\pi}(\lambda), \tag{203}$$

where the mixing contribution $\hat{\pi}$ to the osmotic pressure is given by

$$\hat{\pi}(\lambda) = -\frac{k_B T}{\upsilon}\left.\left(\ln(1 - \phi) + \phi + \chi\phi^2\right)\right|_{\phi = \phi_0/\lambda}. \tag{204}$$

A direct calculation shows that (202) obeys the conditions (165) and (179). Thus, for each λ and τ_{xy}, we can invert $\tau_{xy} = \hat{\tau}(\gamma, \lambda)$ for γ and write

$$\frac{G_0 J_m \tilde{\gamma}(\tau_{xy}, \lambda)}{J_m - (\lambda^2 + \tilde{\gamma}^2(\tau_{xy}, \lambda) - 1)} = \tau_{xy} \tag{205}$$

as the implicit definition of $\tilde{\gamma}$. Another direct calculation shows that $\tilde{\sigma}(\tau_{xy}, \lambda) = \hat{\sigma}(\tilde{\gamma}(\tau_{xy}, \lambda), \lambda)$ complies with the limits

$$\lim_{\lambda \to \phi_0} \tilde{\sigma}(\tau_{xy}, \lambda) = -\infty, \qquad \lim_{\lambda \to \lambda_m} \tilde{\sigma}(\tau_{xy}, \lambda) = +\infty, \tag{206}$$

where λ_m is solution of the equation

$$\lambda_m^2 + \tilde{\gamma}^2(\tau_{xy}, \lambda_m) = J_m + 1 \tag{207}$$

and obeys

$$\phi_0 < \lambda_m < \infty. \tag{208}$$

7.4.1. Instantaneous response

We recall that the instantaneous response is such that $\lambda \equiv 1$ and the gel undergoes an inhomogeneous shear.

On using (202) and (203), we conclude that $\partial \hat{\tau}(\gamma, 1)/\partial \gamma > 0$ and $\partial \hat{\sigma}(\gamma, 1)/\partial \gamma > 0$. Hence, as already discussed, γ and σ_y are decreasing functions for $0 \leq y \leq d/2$. The driving force $\partial p/\partial y = \partial \sigma_y/\partial y$ for liquid flow in the spanwise direction is therefore negative, indicating a tendency for liquid to flow from the walls toward the center of the channel and, correspondingly, for polymer accumulation and depletion to occur close to and far from the channel walls, respectively.

Through the combination of (166) and (205), we find that instantaneous profile of the shear strain γ in the lower half of the channel is such that $\gamma(d/2, 0^+) = 0$ and

$$\gamma(y, 0^+) = \frac{-J_m + \sqrt{J_m^2 + \bar{k}_1^2(1 - 2\bar{y})J_m}}{\bar{k}_1(1 - 2\bar{y})} \qquad \text{for} \qquad 0 \leq y < \frac{d}{2}, \tag{209}$$

where $\bar{k}_1$ and $\bar{y}$ are defined by

$$\bar{k}_1 = \frac{k_1 d}{G_0} \qquad \text{and} \qquad \bar{y} = \frac{y}{d}. \tag{210}$$

From (209), we see that the maximum amount of shear $\gamma_{\max}$ is given by

$$\gamma_{\max} = \frac{-J_m + \sqrt{J_m^2 + \bar{k}_1^2 J_m}}{\bar{k}_1}, \tag{211}$$

which, as expected, tends to $\sqrt{J_m}$ as $\bar{k}_1$ tends to ∞. Furthermore, we find from (166), (202), and (203) that the driving force for transverse flow of the interstitial liquid can be expressed as

$$\frac{\partial p}{\partial y} = -\frac{2k_1 \gamma}{J_m + \gamma^2}. \tag{212}$$

From (209) and (212), we see that as $J_m \to \infty$ the profiles for the amount of shear γ becomes linear and the driving force $\partial p/\partial y$ vanishes identically; these results are consistent with the recovery from $(201)_1$ of the neo-Hookean elastic model as $J_m \to \infty$.

7.4.2. Steady-state response

In the presence of interfaces that separate the gel into low- and high-polymer content phases, the steady-state profiles for the stretch λ and amount of shear γ are discontinuous unless the response $\tilde{\sigma}(\tau_{xy}, \cdot)$ is monotonic for every τ_{xy} in the interval $[0, k_1 d/2]$.

We next discuss the ways in which the parameters ϕ_0, G, J_m, $k_B T$, v, and χ entering the definitions $(201)_1$ and $(201)_2$ of $\tilde{\psi}_e$ and $\hat{\psi}_m$ influence the monotonicity properties of $\tilde{\sigma}$. A closer examination of the partial derivative $\partial \tilde{\sigma}(\tau_{xy}, \lambda)/\partial \lambda$ allows us to introduce a function $\tilde{\chi}$ of τ_{xy} and λ which is parameterized by ϕ_0, J_m, $k_B T/vG$ and whose expression is a bit cumbersome and, hence, is omitted here. From the structure of $\tilde{\chi}$, we deduce that

$$\left.\begin{aligned} \chi < \tilde{\chi}(\tau_{xy}, \lambda) &\iff \frac{\partial \tilde{\sigma}(\tau_{xy}, \lambda)}{\partial \lambda} > 0, \\ \chi = \tilde{\chi}(\tau_{xy}, \lambda) &\iff \frac{\partial \tilde{\sigma}(\tau_{xy}, \lambda)}{\partial \lambda} = 0, \\ \chi > \tilde{\chi}(\tau_{xy}, \lambda) &\iff \frac{\partial \tilde{\sigma}(\tau_{xy}, \lambda)}{\partial \lambda} < 0. \end{aligned}\right\} \tag{213}$$

The conditions in (213) amount to the requirement that $\partial \tilde{\sigma}(\tau_{xy}, \lambda)/\partial \lambda$ and $\tilde{\chi}(\tau_{xy}, \lambda)$ be of opposite sign. Relevant aspects of the behavior of the function $\tilde{\chi}$ are discussed below. For convenience, the results are plotted and discussed in terms of the polymer volume fraction $\phi = \phi_0/\lambda$.

In Fig. 2, we exhibit the behavior of $\tilde{\chi}$ for increasing values of τ_{xy}, starting from $\tau_{xy} = 0$, $\phi_0 = 0.1$, $J_m = 10.$, and $k_B T/vG_0 = 100$. For each representative value of τ_{xy}, a corresponding solid curve, provided by the graph of $\tilde{\chi}$, separates the ϕ-χ plane into two complementary regions, below ($\chi < \tilde{\chi}(\tau_{xy}, \phi_0/\phi)$) and above ($\chi > \tilde{\chi}(\tau_{xy}, \phi_0/\phi)$) the curve, for which $\partial \tilde{\sigma}(\tau_{xy}, \phi_0/\phi)/\partial \phi < 0$ and $\partial \tilde{\sigma}(\tau_{xy}, \phi_0/\phi)/\partial \phi > 0$, respectively. For each τ_{xy}, a corresponding dashed curve indicates the existence of a critical value of χ, denoted by $\chi_c(\tau_{xy})$, below which $\partial \tilde{\sigma}(\tau_{xy}, \phi_0/\phi)/\partial \phi < 0$ for all admissible ϕ. Notice that $\chi_c(\tau_{xy})$ increases with τ_{xy} and that its minimum value χ_c is attained at $\tau_{xy} = 0$. Otherwise, for $\chi > \chi_c(\tau_{xy})$, the corresponding horizontal line intercepts the graph of $\tilde{\chi}(\tau_{xy}, \cdot)$ at two points, $(\phi_1(\tau_{xy}), \chi)$ and $(\phi_2(\tau_{xy}), \chi)$, with $\phi_1(\tau_{xy}) < \phi_2(\tau_{xy})$. The corresponding normal stresses are $\sigma_1(\tau_{xy}) = \tilde{\sigma}(\tau_{xy}, \phi_0/\phi_1(\tau_{xy}))$ and $\sigma_2(\tau_{xy}) = \tilde{\sigma}(\tau_{xy}, \phi_0/\phi_2(\tau_{xy}))$.

Recalling that $\lambda = \phi_0/\phi$, an immediate conclusion drawn the foregoing discussion is that the parameter χ, as expected, plays an important role in determining the behavior of $\tilde{\sigma}(\tau_{xy}, \cdot)$. In particular:

- If $\chi < \chi_c(\tau_{xy})$, $\tilde{\sigma}(\tau_{xy}, \cdot)$ is strictly increasing on (ϕ_0, λ_m) for each admissible value of τ_{xy}.
- If $\chi > \chi_c(\tau_{xy})$, $\tilde{\sigma}(\tau_{xy}, \cdot)$ is strictly increasing on $(\phi_0, \lambda_L(\tau_{xy}))$, strictly decreasing on $(\lambda_L(\tau_{xy}), \lambda_H(\tau_{xy}))$, and again strictly increasing on $(\lambda_H(\tau_{xy}), \lambda_m)$, where $\lambda_L(\tau_{xy}) = \phi_0/\phi_2(\tau_{xy})$ and $\lambda_H(\tau_{xy}) = \phi_0/\phi_1(\tau_{xy})$.
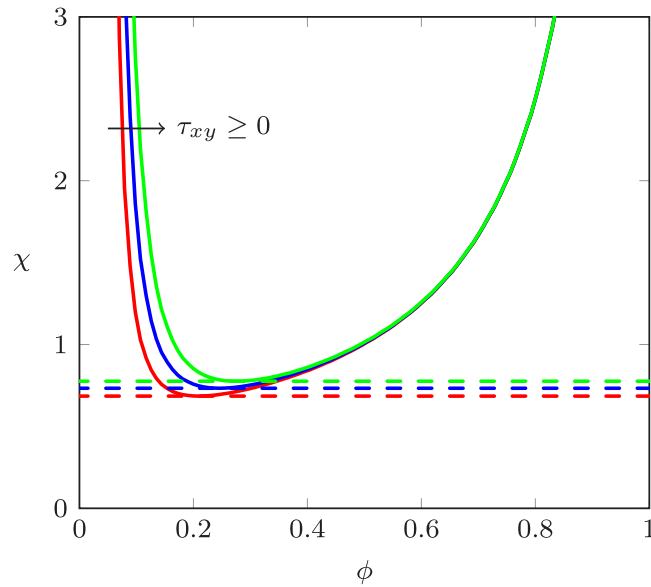
Fig. 2. (χ, ϕ)-diagram for several values of τ_{xy}, $\phi_0 = 0.1$, $J_m = 10$, and $k_B T / v G = 100$. For a given choice of τ_{xy}, the corresponding curve separates the diagram into two complementary regions, below and above the curve, within which $\partial \tilde{\sigma}(\tau_{xy}, \phi_0/\phi)/\partial \phi < 0$ and $\partial \tilde{\sigma}(\tau_{xy}, \phi_0/\phi)/\partial \phi > 0$, respectively. For each τ_{xy}, a corresponding dashed curve indicates the existence of a critical value of χ below which $\partial \tilde{\sigma}(\tau_{xy}, \phi_0/\phi)/\partial \phi < 0$ for all admissible ϕ. Notice, in particular, that, as indicated by the dashed red line, there is a critical value χ_c of χ below which $\partial \tilde{\sigma}(\tau_{xy}, \phi_0/\phi)/\partial \phi < 0$ for any value of τ_{xy} and, hence, for which $\tilde{\sigma}(\tau_{xy}, \cdot)$ is a monotonically decreasing function of ϕ independent of the value of τ_{xy}. Since the curves coalesce for $\phi \gtrsim 0.7$, the monotonicity of $\tilde{\sigma}(\tau_{xy}, \cdot)$ is independent of τ_{xy} for sufficiently dense polymer networks.
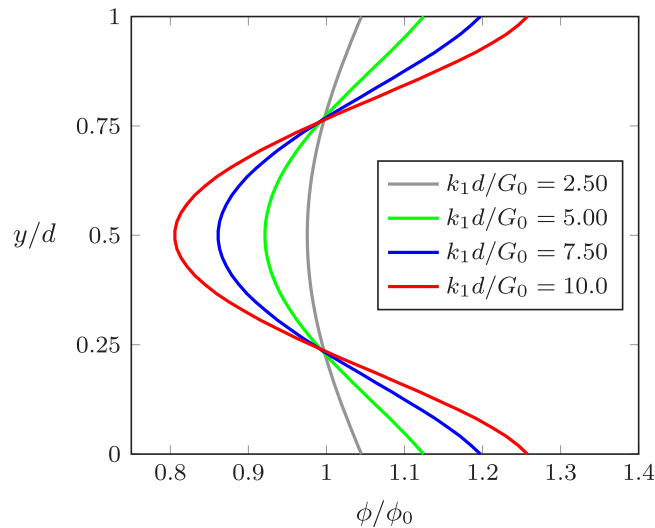

Fig. 3. Polymer volume fraction ϕ/ϕ_0 versus the dimensionless vertical coordinate y/d for $\phi_0 = 0.1$, $J_m = 10$, $k_B T / v G_0 = 100$, $\chi = 0.2$, and various choices of $k_1 d / G_0$.

7.4.3. Polymer volume fraction profile

We next consider the problem of determining the spanwise distribution of the polymer volume fraction ϕ for given input values of pressure drop k_1. In so doing, we consider two complementary cases according to whether $\chi < \chi_c(\tau_{xy})$ and $\chi > \chi_c(\tau_{xy})$). Henceforth, we take $\phi_0 = 0.1$, $J_m = 10$, and $k_B T / v G_0 = 100$, whereby $\chi_c \approx 0.6851$. The selected parameter values are physically viable but are chosen only for illustrative purposes.

In our first example, we take $\chi = 0.2 < \chi_c(\tau_{xy}) \approx 0.6851$ and solve (190) for several values of $k_1 d / G_0$. The results obtained are depicted in Fig. 3, which shows the polymer fraction distributions $\phi/\phi_0 = 1/\lambda$ versus the dimensionless vertical coordinate y/d between the channel walls for increasing values of $k_1 d / G_0$. Notice that the polymer content in the center of the channel diminishes (increases) as $k_1 d / G_0$ increases (diminishes).
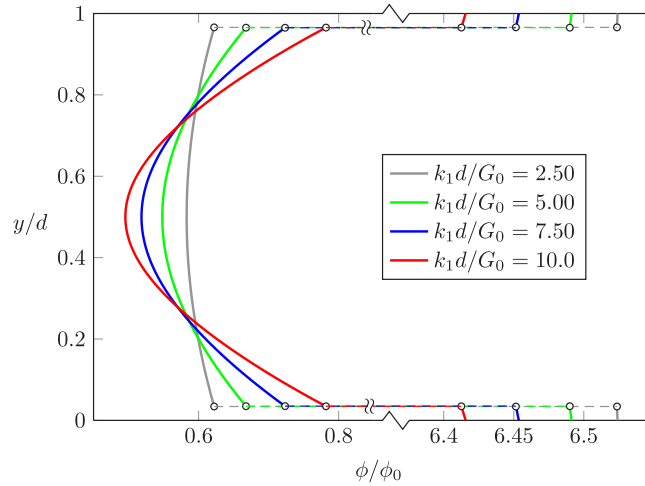
Fig. 4. Polymer volume fraction ϕ/ϕ_0 versus the dimensionless vertical coordinate y/d for $\phi_0 = 0.1$, $J_m = 10$, $k_B T/\upsilon G_0 = 100$, $\chi = 1.0$, and various choices of $k_1 d/G_0$.

For our second and final example, we take $\chi = 1.0 > \chi_c(\tau_{xy}) \approx 0.6851$ and seek the profiles for the polymer volume fraction ϕ corresponding to several values of $k_1 d/G_0$. To achieve this goal, we must solve (173)–(175) and (185) for λ σ_*, w. We construct a solution (185) by using the scheme described in Sub Section 7.3.2.2.

In Fig. 4, we display the polymer fraction distributions for increasing values of $k_1 d/G_0$, finding that the channel is divided into three regions, a central region with a lower polymer volume and two thin regions near the channel walls with a much higher polymer concentration. The figure also shows that the polymer content in the center of the channel decreases as $k_1 d/G_0$ increases and, concomitantly, that the solvent content in the center of the channel increases as $k_1 d/G_0$ decreases.

Finally, we discuss the profile for the ratio j_x^∞/j_x^0 between the steady and instantaneous components j_x^∞ and j_x^0 of the liquid flux in the streamwise direction. By $(147)_1$, $j_x^\infty/j_x^0 = \hat{m}(\lambda)/\hat{m}(1)$ and hence our discussion requires the specification of the constitutive response $\hat{m}$ for m. Here, we assume the power-law relation $\hat{m}(\lambda) = C\lambda^{3/2}$ for $\hat{m}$, where C is a positive constant,[7] which in turn implies that $j_x^\infty/j_x^0 = \lambda^{3/2} = (\phi_0/\phi)^{3/2}$. In Fig. 5, we plot j_x^∞/j_x^0 for $\phi_0 = 0.1$, $J_m = 10$, $k_B T/\upsilon G_0 = 100$, $k_1 d/G_0 = 10$ and two distinct values of χ, namely $\chi = 0.1$ (blue line) and $\chi = 1.0$ (brown line). For comparison, the reference profile $j_x/j_x^0 = 1$ (green line) achieved during the short-time response since there $\phi \approx \phi_0$ is also included in the figure. From Fig. 5, we see that the streamwise liquid flux j_x evolves from a flat profile j_x^0 to a curved profile j_x^∞ which can exhibit discontinuities for sufficiently large values of χ. In this case, the flow in the central part of channel is much larger than the flow close to the channel plates. This result is consistent with one obtained by Cogan and Keener (2005) with a gel model different from ours.

In some works on liquid flow through gels (see, for instance, Levick, 1987 and Loessberg-Zahl et al., 2019), the steady-state streamwise velocity profile is taken to be uniform (i.e., plug flow). Our findings show that this can occur only for specific elastic models for the polymer network, such as the neo-Hookean and Mooney models, in which case the polymer concentration remains unchanged regardless of the applied pressure drop, namely $\phi = \phi_0$ for all times. Otherwise, the applied pressure drop is sustained by an inhomogeneous network normal stress distribution which in turn drives changes in the polymer concentration leading to a nonuniform streamwise flow profile. The relevance and implications of this result remain yet to be explored. It is noteworthy that a nonuniform flow profile can be obtained by any other mechanism leading to changes in the polymer concentration. This point is exemplified by the work of Guy et al. (2011), who considered changes in polymer concentration caused by reformation/breakage of the network induced by fluid flow.

8. A cursory discussion of anisotropic gel networks

We have emphasized general results for isotropic gel networks described under the Flory–Rehner hypothesis of additivity and separability of the elastic and mixing contributions to the free-energy density of such a material. Those results can be generalized to account for network anisotropy by allowing the elastic former contribution to be dependent on additional anisotropic invariants. Such dependence gives rise to concomitant contributions to the stress. For transversely isotropic networks,[8] with preferred direction

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[7] Bearing in mind that $\lambda = \phi_0/\phi$, this relation can be justified by considering that: (i) the permeability of gels is often discussed in terms of a measured quantity, say ξ, called friction coefficient, which according to Tokita and Tanaka (1991) can be described by $\xi = \xi_0 \phi^{3/2}$, with $\xi_0 > 0$; (ii) the mobility m and the friction coefficient ξ are related by $\upsilon^2 m = 1/\xi$. See Drozdov et al. (2016) for pertinent discussion.

[8] Transverse isotropy is commonly used to model the behavior of compliant matrices reinforced with stiff polymeric filaments, a pursuit relevant to the study of biological tissues. See, for instance, the recent review by Wineman and Pence (2021).
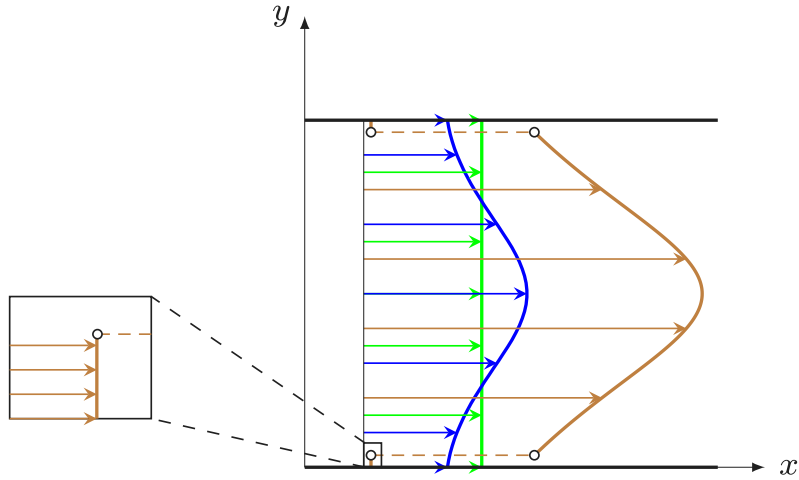
Fig. 5. Distribution of ratio j_x^∞ / j_x^0 between j_x^∞ and j_x^0, the steady and instantaneous components of the liquid flux in the main direction, across the channel for $\phi_0 = 0.1$, $J_m = 10$, $k_B T / v G_0 = 100$, $k_1 d / G_0 = 10$, and two values of χ, namely $\chi = 0.2$ (blue line) and $\chi = 1.0$ (brown line). For reference, the distribution corresponding to the instantaneous profile ($j_x / j_x^0 = 1$) is also represented (green).

in the reference configuration defined by the unit vector e_0, the additional invariants are given by $I_4 := e \cdot e$ and $I_5 := Be \cdot e$, with $e = Fe_0$. The corresponding network stress T_n is then given by

$$T_n = \frac{2}{\det F}\left(\alpha_1 B + \alpha_2(I_1 I - B)B + \alpha_4 e \otimes e + \alpha_5(e \otimes Be + Be \otimes e)\right) - \pi I, \tag{214}$$

where α_4 and α_5 denote the derivatives of the elastic free energy density with respect to the additional invariants I_4 and I_5, respectively. For generalization of our channel flow problem in which e_0 is aligned with the direction of the applied pressure drop, it can be shown that $I_4 = 1$, $I_5 = 1 + \gamma^2$, and that the matrix of T_n retains the form

$$[T_n] = \begin{bmatrix} \sigma_x & \tau_{xy} & 0 \\ \tau_{xy} & \sigma_y & 0 \\ 0 & 0 & \sigma_z \end{bmatrix}, \tag{215}$$

but with σ_x, σ_y, σ_z, and τ_{xy} now given by

$$\left.\begin{aligned}
\sigma_x &= \frac{2}{\lambda}(\alpha_1(1 + \gamma^2) + \alpha_2(1 + \gamma^2 + \lambda^2) + \alpha_4 + 2\alpha_5(1 + \gamma^2)) - \hat{\pi}(\lambda), \\
\sigma_y &= 2\lambda(\alpha_1 + 2\alpha_2) - \hat{\pi}(\lambda), \\
\sigma_z &= \frac{2}{\lambda}(\alpha_1 + \alpha_2(1 + \lambda^2 + \gamma^2)) - \hat{\pi}(\lambda), \\
\tau_{xy} &= 2\gamma(\alpha_1 + \alpha_2 + \alpha_5).
\end{aligned}\right\} \tag{216}$$

Of the network stress components σ_y and τ_{xy} that explicitly enter the analysis of the channel flow problem, it is evident from (129) that only τ_{xy} is affected directly by the transverse symmetry. We therefore suspect that the channel flow analysis can be conducted for the transversely isotropic case with very minor modifications. It would of course be of interest to further generalize the problem to allow for a deviation between the referential alignment e_0 and the channel walls.

We envisage that an immediate application of the present theory regards the study of general flow problems, including the pressure-induced flow through an infinite gel-filled pipe with circular cross-section. In this case, we would use the same procedure adopted in this paper based in the Ansatz

$$X = x + f(r, t), \qquad R = r + g(r, t), \qquad \Theta = \theta, \tag{217}$$

where (R, Θ, X) and (r, θ, x) are the cylindrical coordinates of a gel particle in the reference and actual configurations, respectively.

9. Summary and discussion

A theory for the interplay between large deformation, fluid permeation, and propagation of coherent interfaces in polymer gels was developed and then applied to the problem pressure-driven fluid permeation through a gel slab between two fixed impermeable infinite parallel walls. The theory was formulated by combining fundamental balances with thermodynamical consistent constitutive relations. In this regard, the salient features of the theory are:

1. Introduction of a configurational force system to account for the power expenditure associated with interface motion.
2. Consideration of an aggregate incompressibility condition as an internal constraint maintained by reactions whose associated dissipation rate is null.
3. Incorporation of a constitutive theory suitable for elastic polymer networks and interfaces that propagate without dissipation.

Our treatment of the internal constraint allowed us to conclude that the stress is constitutively assigned to within a pressure-like contribution determined by the chemical potential of the interstitial liquid. That chemical potential is, in turn, the sum of reactive and active parts. Whereas the reactive part of the chemical potential is problem dependent, the active part is prescribed by a constitutive relation. Two such prescriptions were discussed. For one of these, the active part of the chemical potential vanishes identically. For the other, it is prescribed in manner similar to the unconstrained case, namely as if the deformation gradient and fluid content density were independent variables. Using the latter prescription, we introduced a decomposition of the stress into two components: a network component consisting of elastic and osmotic contributions; an interstitial component described in terms of the pressure the interstitial liquid would have in the absence of the network. The aforementioned the decomposition of the stress resembles Terzaghi's (Terzaghi, 1943) principle in poromechanics, which encompasses the notion that the total stress in a liquid infused porous material is the sum of two parts, one associated with porous scaffold and the with the liquid. Notice, however, that here the notion of interstitial liquid pressure, rather than being primitive, was derived from the notion of chemical potential and is, in this sense, akin to the chemical-thermodynamic concepts of activity and fugacity introduced by Lewis and Randall (1923). The interfacial constitutive theory was built on the premise that the motion and chemical potential of the liquid are continuous across an interface. This led to a inequality involving the Eshelby stress tensor which, like the total stress, was decomposed into components associated with the network and the interstitial liquid. Instead of discussing the most general thermodynamically consistent constitutive theory for the interface, we considered a theory appropriate for situations in which interface motion does not generate dissipation. In combination with the configurational force balance, this specialization yields the well-known Maxwell coexistence condition.

The final governing equations of the theory were obtained by combining the fundamental balances and the thermodynamically compatible constitutive relations. These equations can be formulated in different but equivalent forms depending on the choice of primary unknown quantities. We placed emphasis on a formulation suitable for isotropic gels and where the basic unknowns are the network motion, the pressure of the interstitial liquid, and the interface motion, with the liquid pressure providing a two-way coupling between mechanics and permeation. The governing equations were presented in both referential and spatial forms, with the latter applied to investigate fluid permeation through a gel-filled channel. Specifically, we investigated liquid flow through a gel slab of uniform composition placed between two fixed infinite parallel walls, to which the gel adheres, and forced by a pressure drop. This problem was addressed by adopting an approach similar in spirit to that employed by Coleman et al. (1966) to investigate channel flow, or plane Poiseuille flow, of incompressible fluids. Accordingly, we stipulated that the motion experienced by the gel network has the form of an inhomogeneous shearing in the direction of the applied pressure drop followed by an inhomogeneous stretching in the direction perpendicular to the walls of the channel. We also allowed for the possibility that the state of the gel at a given time may involve the presence of planar interfaces symmetrically located with respect to the central plane between the walls of the channel. The governing equations of the theory consequently simplified to a system of equations involving time-dependent functions of the spanwise coordinate, except that the pressure that also varies in the streamwise direction and the interface location depends on time only.

The following picture emerged from our analysis: When a gel fixed between two impermeable walls at rest is forced by a suddenly applied pressure drop, mechanical equilibrium is attained instantaneously since inertia is neglected. The corresponding state of stress is such that the inhomogeneity of the normal network stress acting on material planes parallel to the channel walls drives motion of the interstitial liquid in the direction normal to the walls. That motion is in turn must accompanied by motion of the network in the opposite direction. This process continues until the network normal stress becomes homogeneous and a steady state is reached. For very short times, the gel behaves like an incompressible elastic solid since there is insufficient time for the liquid to flow. Hence, the short-time, or instantaneous, response of the gel is determined by its elastic properties and involves simple shear only. On the other hand, the long-time, or steady state, response of the gel is influenced by the elastic and osmotic properties of the gel.

Through the analysis of the instantaneous response, we exposed the tendency of the liquid to flow in the spanwise direction. We concluded that the interstitial liquid will flow from the channel walls towards the center of the channel whenever the normal network stress increases with the amount of shear, as is the case of strain-stiffening generalized neo-Hookean elastic models. The opposite occurs when the normal network stress decreases with the amount of shear, as in the case of strain-slackening generalized neo-Hookean elastic models. If the shear stress is unaffected by the amount of shear, as in the case of neo-Hookean and Mooney elastic models, there will be no tendency toward spanwise flow. Our investigation of the steady-state response was aimed at describing the profile reached by the only non-null component of the liquid flux, namely the component in the direction of and proportional to the applied pressure drop. Bearing in mind that the proportionality factor is determined by the liquid mobility, which increases with the stretch in the spanwise direction, we sought to obtaining the profile for that stretch. Assuming that the constitutive relation given the shear stress as a function of the stretch and the amount of shear can be inverted for the amount of shear, we arrived at set of equations to be solved for the stretch distribution. As a consequence of this assumption, the configurational force balance is equivalent to the Maxwell equal area rule written for the partial of Legendre transform of free-energy density with respect to the shear stress. We established a necessary condition for presence of an interface across which the stretch displays a jump discontinuity. This condition hinges on the non-monotonicity of the constitutive equation for normal network stress. This led us to address two separate cases, with and without interfaces, according to which the profile for the stretch is continuous or not.

We also provided illustrative examples by using the Gent and Flory–Huggins models for elastic and mixing contributions to the free-energy of the gel. The analysis of the instantaneous response showed that the interstitial liquid tends to flow inward

from the channel walls towards the center of the channel. This was corroborated by the steady-response which displays polymer accumulation and depletion, and, hence, lower and higher flow intensity, in the outermost and innermost regions of the channel. We also showed that there is a critical value for the Flory interaction parameter below of which the profile for the stretch in the direction perpendicular to the channel walls is smooth.

CRediT authorship contribution statement

Fernando P. Duda: Conceptualization, Methodology, Software, Formal analysis, Writing – original draft. **Angela C. Souza:** Conceptualization, Methodology, Visualization, Writing – review & editing. **Eliot Fried:** Conceptualization, Methodology, Formal analysis, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

Appendix A. The elastic and osmotic responses

A.1. General relations

It is evident from (129)–(130) that the components of the network stress $\boldsymbol{T}_{\mathrm{n}}$ are determined by the elastic response functions $\hat{\alpha}_1$ and $\hat{\alpha}_2$ and the osmotic response function $\hat{\pi}$. Whereas the shear stress τ_{xy} involves elastic contributions only, the normal stresses σ_x, σ_y, and σ_z incorporate elastic and osmotic contributions. Granted that $\gamma \neq 0$, the expressions $(129)_{1-3}$ for σ_x, σ_y, and σ_z can be used to express $\hat{\alpha}_1$, $\hat{\alpha}_2$, and $\hat{\pi}$ as

$$
\left.
\begin{aligned}
\hat{\alpha}_1(\gamma, \lambda) &= \frac{\lambda(\sigma_x - \sigma_z)}{2\gamma^2}, \\
\hat{\alpha}_2(\gamma, \lambda) &= \frac{((\lambda^2 - 1)(\sigma_x - \sigma_z) - \gamma^2(\sigma_y - \sigma_z))\lambda}{2\gamma^2(1 + \gamma^2 - \lambda^2)}, \\
\hat{\pi}(\lambda) &= \frac{((\gamma^2 + \lambda^2 - 1)(\sigma_x - \sigma_z) - 2\gamma^2(\sigma_y - \sigma_z))\lambda^2}{\gamma^2(1 + \gamma^2 - \lambda^2)} - \sigma_y.
\end{aligned}
\right\}
\tag{A.1}
$$

From (A.1), we see that, while $\hat{\alpha}_1$, $\hat{\alpha}_2$, and $\hat{\pi}$ depend on the normal stress differences $\sigma_x - \sigma_z$ and $\sigma_y - \sigma_z$, $\hat{\pi}$ depends in addition on the normal stress σ_y. Furthermore, from $(129)_{1,2,4}$ we obtain a relation,

$$
\sigma_x - \sigma_y = \left(\frac{1 + \gamma^2 - \lambda^2}{\lambda\gamma} \right) \tau_{xy},
\tag{A.2}
$$

that hold for every isotropic gel and hence is universal for that class of materials. A similar relation

$$
\lambda^2 \sigma_x - \sigma_y = \tau_{xy}\gamma + 2\frac{\partial \tilde{\psi}_e(I_1, I_2)}{\partial I_2}\lambda(\lambda^2 - 1)
\tag{A.3}
$$

also holds, but only for the class of gels whose elastic energies do not depend on the second invariant I_2. For $\lambda = 1$, (A.1)–(A.2) reduce to the classical relations obtained for simple shear deformation of homogeneous, isotropic elastic bodies. See, for instance, Gurtin et al. (2010). A relation similar to (A.2) was obtained by Wineman and Gandhi (1984).

A.2. The generalized shear modulus

We now make some observations regarding the positivity of the response for generalized shear modulus (132):

- For $(\gamma, \lambda) \neq (1, 0)$, this assumption follows from the classical Baker–Ericksen inequality. A calculation involving $(118)_1$, (128), (129), and $(131)_2$, leads to the conclusion $\hat{G}$ satisfies the equation

$$\hat{G}(\gamma, \lambda) = \lambda \left(\frac{\sigma_1 - \sigma_2}{\lambda_1^2 - \lambda_2^2} \right), \tag{A.4}$$

where σ_1 and σ_2 are the in-plane principal stresses associated with the principal stretches λ_1 and λ_2 given by

$$\lambda_{1,2}^2 = \frac{1 + \gamma^2 + \lambda^2}{2} \pm \sqrt{\left(\frac{1 + \gamma^2 - \lambda^2}{2} \right)^2 + \lambda^2 \gamma^2}. \tag{A.5}$$

From (A.5), we see that $\lambda_1 = \lambda_2$ if and only if $\lambda = 1$ and $\gamma = 0$. For elastic energies obeying the classical inequalities postulated by Baker & Ericksen in Baker and Ericksen (1954), the right-hand side of (A.4) must be positive whenever $\lambda_1 \neq \lambda_2$, from which it follows that $\hat{G}(\gamma, \lambda) > 0$ for $(\gamma, \lambda) \neq (1, 0)$.

- For each fixed choice of λ, the condition $\hat{G}(\lambda, 0) > 0$ implies that $\partial \tilde{\tau}(\gamma, \lambda) / \partial \gamma > 0$ at $\gamma = 0$. Granted that $\partial \hat{\tau}(\gamma, \lambda) / \partial \gamma$ is continuous, $\hat{\tau}(\gamma, \lambda)$ must therefore be a strictly increasing function of γ in some neighborhood of $\gamma = 0$, and, hence, can be inverted for γ in that neighborhood. However, it may happen that $\hat{\tau}(\cdot, \lambda)$ ceases to be monotonically increasing if γ exceeds a certain value. It is well known within the context of nonlinear elasticity that an isotropic and incompressible material cannot sustain equilibrated shear deformations involving coexistent phases unless its shear stress depends nonmonotonically on the amount of shear. See, for instance, Rajagopal and Wineman (1984), Fosdick et al. (1986), and Fosdick and MacSithigh (1986). However, such situations were not considered here.

We finalize the present discussion by providing two models for the elastic contribution $\hat{\psi}_e$ to the free energy response $\hat{\psi}$. These are the power-law and limiting-chain extensibility models proposed by Knowles (1977) and Gent (1996), wherein $\hat{\psi}_e$ are given, respectively, by

$$\hat{\psi}_e(I_1, I_2) = \frac{G_0}{2b} \left[\left(1 + \frac{b}{n}(I_1 - 3) \right)^n - 1 \right], \qquad G_0 > 0, \qquad n > 0, \qquad b > 0, \tag{A.6}$$

and

$$\hat{\psi}_e(I_1, I_2) = -\frac{G_0}{2} J_m \ln \left(1 - \frac{I_1 - 3}{J_m} \right), \qquad G_0 > 0, \qquad J_m > 0. \tag{A.7}$$

In (A.6) and (A.7), G_0 is the infinitesimal shear modulus. In (A.7), finite values of J_m models= limited chain extensibility by strongly penalizing values of $I_1 - 3$ that approach J_m. The generalized shear moduli corresponding to (A.6) and (A.7) are given by

$$\hat{G}(\gamma, \lambda) = G_0 \left(\frac{b}{n}(\gamma^2 + \lambda^2 - 1) + 1 \right)^{n-1} \tag{A.8}$$

and

$$\hat{G}(\gamma, \lambda) = \frac{G_0 J_m}{J_m - (\gamma^2 + \lambda^2 - 1)}, \tag{A.9}$$

respectively. Notice that the generalized shear modulus (A.9) for the Gent model (A.7) is always positive. On the other hand, the generalized shear modulus (A.8) corresponding to the power-law model (A.6) is positive if and only n and b are such that

$$b(\gamma^2 + \lambda^2) + n - b > 0 \tag{A.10}$$

for all admissible λ and γ. Thus, between the power-law model (A.6) and the Gent model (A.7), (A.6) allows for equilibria involving coexisting phases if (A.10) is violated.

Bearing in mind that λ can take values in the interval (ϕ_0, ∞) and that $\gamma \geq 0$, we infer that (A.10) is equivalent to

$$n \geq b(1 - \phi_0^2). \tag{A.11}$$

Thus, the generalized shear modulus is positive for all admissible λ, γ, and ϕ_0 if and only if $n > b$. This condition appears as a result of the compressibility since in the incompressible case $\hat{G}(\gamma, 1) > 0$ for any value of $n > 0$ and $b > 0$.

Appendix B. A necessary condition for the occurrence of interfaces

We now introduce a necessary condition for a solution of (155)–(157) to allow for the presence of an interface across which the amount of shear γ and stretch λ exhibit jump discontinuities. The negative of this condition suffices for the occurrence of smooth solutions of (155)–(157). The mentioned condition is written in terms of the elastic and mixing responses of a gel.

We therefore suppose that, at a given time t, there is an interface located at $y = w(t)$. The network stresses σ_y and τ_{xy} are continuous across the interface, with the corresponding values denoted by σ_* and τ_*. By $(156)_5$ and $(156)_6$, these stress components are related to the limiting values of γ and λ from each side of the interface by

$$\sigma_* = \hat{\sigma}\left(\overset{+}{\gamma}, \overset{+}{\lambda} \right) = \hat{\sigma}\left(\overset{-}{\gamma}, \overset{-}{\lambda} \right) \qquad \text{and} \qquad \tau_* = \hat{\tau}\left(\overset{+}{\gamma}, \overset{+}{\lambda} \right) = \hat{\tau}\left(\overset{-}{\gamma}, \overset{-}{\lambda} \right). \tag{B.1}$$

Thus, the presence of an interface requires that $(\bar{\gamma}, \bar{\lambda}) \neq (\overset{+}{\gamma}, \overset{+}{\lambda})$. Otherwise the deformation gradient would be continuous. Hence, the possibility of an interface across which the deformation gradient displays jump discontinuity is ruled out whenever the mapping $\hat{\sigma} := (\hat{\tau}, \hat{\sigma})$ is globally invertible on its domain of definition,[9] namely

$$\mathcal{D} := \{(\gamma, \lambda) \mid \ \gamma \geq 0 \quad \text{and} \quad \lambda > \phi_0\}. \tag{B.2}$$

We now introduce a sufficient condition for the globally invertibility of $\hat{\sigma}$. Towards this end, we begin by noticing that, by (134), the "gradient" of $\hat{\sigma}$ is equal to the Hessian matrix of $\hat{\varphi}$:

$$\begin{bmatrix} \dfrac{\partial \hat{\tau}}{\partial \gamma} & \dfrac{\partial \hat{\tau}}{\partial \lambda} \\ \dfrac{\partial \hat{\sigma}}{\partial \gamma} & \dfrac{\partial \hat{\sigma}}{\partial \lambda} \end{bmatrix} = \begin{bmatrix} \dfrac{\partial^2 \hat{\varphi}}{\partial \gamma^2} & \dfrac{\partial^2 \hat{\varphi}}{\partial \gamma \partial \lambda} \\ \dfrac{\partial^2 \hat{\varphi}}{\partial \gamma \partial \lambda} & \dfrac{\partial^2 \hat{\varphi}}{\partial \lambda^2} \end{bmatrix} =: \boldsymbol{H}. \tag{B.3}$$

On invoking results presented by Bernstein and Toupin (1962), it can be shown that if $\boldsymbol{H}$ is positive definite on $\mathcal{D}$, then $\hat{\varphi}$ is strictly convex there, which implies that the "gradient" of $\hat{\varphi}$ is globally invertible. From the above, we conclude that the positive-definiteness of $\boldsymbol{H}$ on $\mathcal{D}$ is sufficient for global invertibility of the mapping $\hat{\sigma}$ and hence rules out the possibility of interfaces. Equivalently, the non-positive definiteness of the Hessian matrix $\boldsymbol{H}$ on $\mathcal{D}$ is necessary for the existence of interfaces. At this juncture, the following remarks are salient:

- That the non-positive definiteness of the Hessian matrix of $\hat{\varphi}$ is necessary to the existence of interfaces may be seen as a specialization of general results established by Rosakis (1990) within the context of elastostatics.
- After a lengthy calculation, it can be shown that the Hessian matrix of $\hat{\varphi}$ is positive definite if and only if the acoustic tensor $\boldsymbol{Q}(\boldsymbol{F}, \boldsymbol{e}_2)$ in the direction of $\boldsymbol{e}_2$ parallel to the y-axis at $\boldsymbol{F}$ of the form (116), which can defined by

$$\boldsymbol{Q}(\boldsymbol{F}, \boldsymbol{e}_2)\boldsymbol{a} = (D\hat{\boldsymbol{S}}_{\mathrm{n}}(\boldsymbol{F})[\boldsymbol{a} \otimes \boldsymbol{e}_2])\boldsymbol{e}_2,$$

  for every vector $\boldsymbol{a}$, where $D\hat{\boldsymbol{S}}_{\mathrm{n}}(\boldsymbol{F})$ is the derivative of the network stress response at $\boldsymbol{F}$, is positive definite. Again, this result may be seen as a specialization of those established in Rosakis (1990). Notice that the definition of $\boldsymbol{Q}(\boldsymbol{F}, \boldsymbol{e}_2)$ was given intrinsically as in Šilhavỳ (1997).
- Within the context finite elasticity, the positive definiteness of the acoustic tensor $\boldsymbol{Q}(\boldsymbol{F}, \boldsymbol{n})$ for every $\boldsymbol{F}$ and direction $\boldsymbol{n}$ is equivalent to the requirement that the strain energy response of an elastic material satisfies the strong ellipticity condition. See, for instance, Rosakis (1990).

We now seek for conditions under which the Hessian matrix $\boldsymbol{H}$ fails to be positive definite on $\mathcal{D}$. Towards this end, we begin by observing that $\boldsymbol{H}$ is globally positive definite if and only if the conditions

$$\frac{\partial \hat{\tau}}{\partial \gamma} > 0 \qquad \text{and} \qquad \frac{\partial \hat{\tau}}{\partial \gamma}\frac{\partial \hat{\sigma}}{\partial \lambda} - \frac{\partial \hat{\tau}}{\partial \lambda}\frac{\partial \hat{\sigma}}{\partial \gamma} > 0 \tag{B.4}$$

hold simultaneously on $\mathcal{D}$. If at least one these conditions is violated, $\boldsymbol{H}$ fails to be positive definite on $\mathcal{D}$. Our attention in this paper is limited to situations in which the presence of interfaces stems from volume transition, as considered by Dušek and Dušková-Smrčková (2020) and Doi (2013), among others. This type of transition is driven by changes in the polymer-solvent affinity and hence by the mixing response of the gel. Since, by (131), only the normal stress response $\hat{\sigma}$ is affected by the mixing response, the condition $(B.4)_1$ is always satisfied on $\mathcal{D}$ and hence failure of the positive definiteness of $\boldsymbol{H}$ can occur only through the violation of $(B.4)_2$ on $\mathcal{D}$. We thus arrive at the sought necessary condition for the existence of interfaces:

$$\frac{\partial \hat{\tau}}{\partial \gamma}\frac{\partial \hat{\sigma}}{\partial \lambda} - \frac{\partial \hat{\tau}}{\partial \lambda}\frac{\partial \hat{\sigma}}{\partial \gamma} \leq 0 \tag{B.5}$$

at some points of $\mathcal{D}$. Notice that since $(B.4)_1$ is always satisfied on $\mathcal{D}$, the function $\hat{\tau}(\cdot, \lambda)$ can be inverted for any admissible λ and hence we can introduce the response function $\tilde{\sigma}$ defined by $\tilde{\sigma}(\tau, \lambda) = \hat{\sigma}(\tilde{\gamma}(\tau, \lambda), \lambda)$, where $\tau = \hat{\tau}(\gamma, \lambda)$ and $\gamma = \tilde{\gamma}(\tau, \lambda)$. Thus, after a simple calculation, we can conclude that $(B.4)_2$ is equivalent to the requirement that

$$\frac{\partial \tilde{\sigma}}{\partial \lambda} > 0 \tag{B.6}$$

hold for all admissible pairs (τ, λ) and that (B.5) is satisfied at $(\bar{\gamma}, \bar{\lambda})$ if and only if

$$\frac{\partial \tilde{\sigma}}{\partial \lambda} \leq 0 \tag{B.7}$$

holds at $(\bar{\tau}, \bar{\lambda})$, where $\bar{\tau} = \hat{\tau}(\bar{\gamma}, \bar{\lambda})$. Additional discussion pertaining to the response function $\tilde{\sigma}$ is presented next.

---

[9] The definition of $\mathcal{D}$ given above follows from $(131)_2$, $(155)_2$, and $(156)_4$ together with the fact that $\tau_{xy} \geq 0$ in the lower-half of the channel and that $\phi \in (0, 1)$.

Appendix C. The response function $\tilde{\sigma}$

The properties of the response function $\tilde{\sigma}$ have crucial effects on the problem under consideration in this paper. Some of the most relevant properties will be discussed below. Without further notice, we recall that τ_{xy} must satisfy $0 \leq \tau_{xy} \leq k_1 d/2$.

We begin by considering the domain and range of $\tilde{\sigma}(\tau_{xy}, \cdot)$. First, we observe that, according to (156)$_4$, λ can take values in the interval (ϕ_0, ∞), with $\lambda \to \phi_0$ as $\phi \to 1$ and $\lambda \to \infty$ as $\phi \to 0$. In the presence of limiting chain extensibility effects, λ takes values in the interval (ϕ_0, λ_m) instead, where $\lambda_m < \infty$, the limiting value for the stretch λ, may depend on γ, and hence on τ_{xy}. Moreover, we consider that $\tilde{\sigma}(\tau_{xy}, \cdot)$ obeys the conditions

$$\lim_{\lambda \to \phi_0} \tilde{\sigma}(\tau_{xy}, \lambda) = -\infty \qquad \text{and} \qquad \lim_{\lambda \to \lambda_m} \tilde{\sigma}(\tau_{xy}, \lambda) = \infty, \tag{C.1}$$

which may be justified by recalling that, in view of (131)$_1$ and (181)$_2$, $\tilde{\sigma}(\tau_{xy}, \cdot)$ is the sum of two contributions, one elastic and other osmotic, that is,

$$\tilde{\sigma}(\tau_{xy}, \lambda) = \underbrace{2\lambda(\tilde{\alpha}_1(\tau_{xy}, \lambda) + 2\tilde{\alpha}_2(\tau_{xy}, \lambda))}_{\tilde{\sigma}_e(\tau_{xy}, \lambda)} - \hat{\pi}(\lambda), \tag{C.2}$$

and assuming that the following conditions hold:

- The osmotic pressure $\hat{\pi}(\lambda)$ tends to ∞ as λ tends to ϕ_0 and remains bounded as λ tends to λ_m.
- The elastic contribution $\tilde{\sigma}_e(\tau_{xy}, \lambda)$ tends to ∞ as λ tends to λ_m and remains bounded as λ tends to ϕ_0.

Consider a given $\tau_{xy} \geq 0$. As for the monotonicity properties of $\tilde{\sigma}(\tau_{xy}, \lambda)$, we consider the following possibilities:

(i) $\tilde{\sigma}(\tau_{xy}, \cdot)$ is a monotonically increasing function of λ, that is,

$$\frac{\partial \tilde{\sigma}(\tau_{xy}, \lambda)}{\partial \lambda} > 0. \tag{C.3}$$

(ii) $\tilde{\sigma}(\tau_{xy}, \cdot)$ is non-monotonic, with

$$\frac{\partial \tilde{\sigma}(\tau_{xy}, \lambda)}{\partial \lambda} > 0 \quad \text{on} \quad (\phi_0, \lambda_m) \setminus [\lambda_L, \lambda_H] \qquad \text{and} \qquad \frac{\partial \tilde{\sigma}(\tau_{xy}, \lambda)}{\partial \lambda} < 0 \quad \text{on} \quad (\lambda_L, \lambda_H), \tag{C.4}$$

where λ_L and λ_H, with $\phi_0 < \lambda_L < \lambda_H < \lambda_m$, are solutions of the equation

$$\frac{\partial \tilde{\sigma}(\tau_{xy}, \lambda)}{\partial \lambda} = 0. \tag{C.5}$$

Notice that, like λ_m, λ_L and λ_H may depend on τ_{xy}, but this dependency is omitted here for the sake of simplicity.

For the purposes of the present work, we stipulate that the elastic part $\tilde{\sigma}_e$ of $\tilde{\sigma}$ is such that, for each τ_{xy},

$$\frac{\partial \tilde{\sigma}_e(\tau_{xy}, \lambda)}{\partial \lambda} > 0 \tag{C.6}$$

for all $\lambda \in (\phi_0, \lambda_m)$. In this way, the loss of monotonicity of $\hat{\sigma}(\tau_{xy}, \cdot)$ is caused by the osmotic pressure response $\hat{\pi}$ since, by (C.2), (C.3) is equivalent to

$$\frac{\partial \tilde{\sigma}_e(\tau_{xy}, \lambda)}{\partial \lambda} > \frac{\partial \hat{\pi}(\lambda)}{\partial \lambda}. \tag{C.7}$$

From this we can conclude the following:

- According to the classical theory of polymer solutions as presented, for instance, by Doi (2013), the properties of the osmotic pressure reflect the effective interaction between polymer molecules comprising the network. When the effective interaction is repulsive, the osmotic pressure increases monotonically with ϕ and hence, since $\lambda = \phi_0/\phi$, $\partial \hat{\pi}/\partial \lambda < 0$ for all λ. Otherwise, when the effective interaction is attractive, the osmotic pressure first decreases with ϕ, hence $\partial \hat{\pi}/\partial \lambda > 0$, and then increases with ϕ and hence $\partial \hat{\pi}/\partial \lambda < 0$. Thus:

  - If the effective interaction between polymer molecules is repulsive, then $\tilde{\sigma}(\tau_{xy}, \cdot)$ is a monotonic function of λ for any τ_{xy}.
  - If $\tilde{\sigma}(\tau_{xy}, \cdot)$ is non-monotonic, then the effective interaction between polymer molecules is attractive.

- It can be shown that (C.6) holds for elastic materials of the power-law type (A.6) if $n > 1/2$ as well for any elastic material of Gent type (A.7).

Appendix D. Solving $\tilde{\sigma}(\tau_{xy}, \lambda) = \sigma_y$ for λ

For the ensuing discussion, we drop the subscripts from σ_y and τ_{xy} for simplicity and consider a gel cube of unit dimensions, with faces parallel to Cartesian axes, undergoing the deformation (cf. (112))

$$x = X + \gamma Y, \qquad y = \lambda X, \qquad z = Z, \tag{D.1}$$

where (X, Y, Z) and (x, y, z) are the coordinates of a gel particle in the undeformed and deformed configurations, respectively. This deformation corresponds to stretch $\lambda > 0$ superimposed to a simple shear of amount γ. The cube is immersed in incompressible and pure fluid bath and subject to the normal stress $\sigma - p_a$ and τ in the face $Y = 1$, where p_a is fluid pressure in environment and τ the applied shear stress, which henceforth is supposed to be given as a fixed value. Alternatively, we may think of (D.1) as the deformation achieved by a gel slab placed between two parallel walls, one fixed and impermeable at $Y = 0$ and other movable and permeable to the fluid at $Y = 1$.

As Duda et al. (2010) show, to characterize the equilibrium states we must find critical points of the net potential energy which in the present case is given, modulo a inconsequential constant, by

$$\mathcal{F}(\gamma, \lambda) = \hat{\varphi}(\gamma, \lambda) - \tau\gamma - \sigma(\lambda - 1). \tag{D.2}$$

Thus, equilibrium states are characterized by

$$\left. \begin{aligned} \sigma = \hat{\sigma}(\gamma, \lambda) &:= \frac{\partial\hat{\varphi}(\gamma, \lambda)}{\partial\lambda}, \\ \tau = \hat{\tau}(\gamma, \lambda) &:= \frac{\partial\hat{\varphi}(\gamma, \lambda)}{\partial\gamma}. \end{aligned} \right\} \tag{D.3}$$

If, given λ, we suppose that $\hat{\tau}(\cdot, \lambda)$ is invertible with inverse $\tilde{\gamma}(\cdot, \lambda)$, we can express the net potential energy as a function of λ and τ through

$$\tilde{\mathcal{F}}(\tau, \lambda) = \tilde{\Phi}(\tau, \lambda) - \sigma(\lambda - 1), \tag{D.4}$$

where $\tilde{\Phi}(\tau, \lambda) = \hat{\varphi}(\tilde{\gamma}(\tau, \lambda), \lambda) - \tau\tilde{\gamma}(\tau, \lambda)$ is the negative of the partial Legendre transform of $\hat{\varphi}$ with respect to τ. The critical points of $\tilde{\mathcal{F}}$ are obtained by solving

$$\left. \begin{aligned} \sigma = \tilde{\sigma}(\tau, \lambda) &:= \frac{\partial\tilde{\Phi}(\tau, \lambda)}{\partial\lambda}, \\ \gamma = \tilde{\gamma}(\tau, \lambda) &:= \frac{\partial\tilde{\Phi}(\tau, \lambda)}{\partial\tau}. \end{aligned} \right\} \tag{D.5}$$

We now consider the following problem: Given σ, solve

$$\sigma = \tilde{\sigma}(\tau, \lambda) \tag{D.6}$$

for λ, with the shear stress $\tau \geq 0$ treated as a fixed parameter. Observe that the stated problem is equivalent to the problem of finding equilibrium configurations of elastic bars subjected to dead loads (see, for instance, the monographs by Ericksen, 1998 and Abeyaratne and Knowles, 2006).

As in the one-dimensional case, two possibilities are considered here: (i) $\hat{\sigma}$ is a monotonically increasing function of λ; (ii) $\hat{\sigma}$ is monotonically increasing in (ϕ_0, λ_L), monotonically decreasing on (λ_L, λ_H), and monotonically increasing on (λ_H, λ_m), with λ_L and λ_H obtained by solving

$$\frac{\partial\tilde{\sigma}(\tau, \lambda)}{\partial\lambda} = 0. \tag{D.7}$$

In view of (188), there always exists a solution (D.6) for a given σ. This solution is unique provided that $\tilde{\sigma}$ depends monotonically on λ. When $\tilde{\sigma}$ is non-monotonic, we have the following alternatives:

- one solution if $\sigma < \tilde{\sigma}(\tau, \lambda_L)$ or $\sigma > \tilde{\sigma}(\tau, \lambda_H)$;
- two solutions if $\sigma = \tilde{\sigma}(\tau, \lambda_L)$ or $\sigma = \tilde{\sigma}(\tau, \lambda_H)$;
- three solutions if $\tilde{\sigma}(\tau, \lambda_L) < \sigma < \tilde{\sigma}(\tau, \lambda_H)$.

In this case, the solution λ^* is said to correspond to low- and high-stretch phases if $\lambda < \lambda_L$ and $\lambda > \lambda_H$, respectively. If it is stipulated that a solution λ^* must be an absolute minimizer of $\tilde{\mathcal{F}}$, which is equivalent to requirement that the inequality

$$\tilde{\Phi}(\lambda^*, \tau) - \sigma\lambda^* \leq \tilde{\Phi}(\tau, \lambda) - \sigma\lambda \tag{D.8}$$

holds for all λ, such solution must be such that:

- $\lambda^* < \lambda_1^M$ when $\sigma < \sigma_M$,
- $\lambda^* > \lambda_2^c$ when $\sigma > \sigma_M$,
- $\lambda^* \in \{\lambda_1^M, \lambda_2^M\}$ when $\sigma = \sigma_M$,

where λ_1^M, λ_2^M, and σ_M, are such that

$$\left.\begin{aligned}
\sigma_M = \hat{\sigma}(\tau, \lambda_1^M) = \tilde{\sigma}(\tau, \lambda_2^M), \\
\bar{\Phi}(\tau, \lambda_1^M) - \sigma_M \lambda_1^M = \bar{\Phi}(\tau, \lambda_2^M) - \sigma_M \lambda_2^M.
\end{aligned}\right\} \tag{D.9}$$

The latter conditions are the Maxwell equal area rule and σ_M is the Maxwell stress.

Appendix E. Reformulation of the governing equations

We recall that (155)–(157) furnish a system of equations to be solved for f, g, λ, γ, ϕ, σ_y, w, and p, with the latter quantity being determinate to within a time-dependent part. The quantities given are the applied pressure drop per unit length k_1, the polymer volume fraction ϕ_0 in the reference state, the volume v occupied by a fluid molecule, and the constitutive responses $\hat{\psi}$ — from which $\hat{\tau}$ and $\hat{\sigma}$ are derived — and $\hat{m}$. The resulting system must be supplemented with appropriate boundary and initial conditions (158) and (159).

We now reformulate the aforementioned system of equations for situations where the shear stress τ_{xy} can be treated as an independent constitutive variable. In this case, the appropriate energy potential is given $\tilde{\Phi}$, from which $\tilde{\gamma}$ and $\tilde{\sigma}$ are derived by

$$\tilde{\gamma} = \frac{\partial \tilde{\Phi}}{\partial \tau_{xy}} \quad \text{and} \quad \tilde{\sigma} = \frac{\partial \tilde{\Phi}}{\partial \lambda}.$$

Thus, bearing in mind that

$$\tau_{xy} = k_1 \left(\frac{d}{2} - y \right) \tag{E.1}$$

for all $0 \leq y \leq d/2$ and that k_1, ϕ_0, v, $\tilde{\Phi}$, and $\hat{m}$ are given, the problem of obtaining f, g, γ, λ, v_y, ϕ, σ_y, and w can be solved in steps as follows:

Step 1: Obtain g, λ, v_y, ϕ, σ_y, and w by solving

$$\left.\begin{aligned}
\lambda &= \left(1 + \frac{\partial g}{\partial y} \right)^{-1}, \\
v_y &= -\lambda \frac{\partial g}{\partial t}, \\
\lambda \phi &= \phi_0, \\
\sigma_y &= \tilde{\sigma}(\tau_{xy}, \lambda), \\
v_y &= v^2 \hat{m}(\lambda) \frac{\partial \sigma_y}{\partial y},
\end{aligned}\right\} \tag{E.2}$$

for all $0 < y < w$ and $w < y < d/2$, and

$$\left.\begin{aligned}
[\![\phi]\!]\dot{w} + [\![\phi v_y]\!] &= 0, \\
\tilde{\sigma}(\tau_*, \overset{-}{\lambda}) &= \sigma_*, \\
\tilde{\sigma}(\tau_*, \overset{+}{\lambda}) &= \sigma_*, \\
[\![\tilde{\Phi}(\tau_{xy}, \lambda)]\!] - \sigma_*[\![\lambda]\!] &= 0,
\end{aligned}\right\} \tag{E.3}$$

for $y = w$, where $\sigma_* = \sigma_y(w)$ and $\tau_* = \tau_{xy}(w)$. These equations must be supplemented by the boundary and initial conditions $g(0, t) = g(d/2, t) = 0$ and $g(y, 0) = 0$ for g as well as by the initial condition $w(0) = 0$ for w under the proviso that an interface initiates at the channel walls.

Step 2: Obtain f and γ by solving

$$\left.\begin{aligned}
\gamma &= \lambda \frac{\partial f}{\partial y}, \\
\gamma &= \tilde{\gamma}(\tau_{xy}, \lambda),
\end{aligned}\right\} \tag{E.4}$$

for all $0 < y < w$ and $w < y < d/2$, and

$$\left.\begin{aligned}
\overset{-}{\gamma} &= \tilde{\gamma}(\tau_*, \overset{-}{\lambda}), \\
\overset{+}{\gamma} &= \tilde{\gamma}(\tau_*, \overset{+}{\lambda}),
\end{aligned}\right\} \tag{E.5}$$

for $y = w$, where $\sigma_* = \sigma_y(w)$ and $\tau_* = \tau_{xy}(w)$. These equations must be supplemented by the boundary conditions for f stating that its value and y-derivative vanish at $y = 0$ and $y = d/2$ for any time.

Notice that, by using (E.2), one can conclude that g obeys a reaction–diffusion-like equation of the form

$$\frac{\partial g}{\partial t} = \tilde{D}(\tau_{xy}, \lambda)\frac{\partial^2 g}{\partial y^2} + \tilde{r}(\tau_{xy}, \lambda), \tag{E.6}$$

where $\hat{D}(\tau_{xy}, \lambda)$ and $\tilde{r}(\tau_{xy}, \lambda)$ are given by

$$\tilde{D}(\tau_{xy}, \lambda) := v^2\hat{m}(\lambda)\lambda\frac{\partial\tilde{\sigma}(\tau_{xy}, \lambda)}{\partial\lambda} \qquad \text{and} \qquad \tilde{r}(\tau_{xy}, \lambda) := k_1 v^2\hat{m}(\lambda)\frac{1}{\lambda}\frac{\partial\tilde{\sigma}(\tau_{xy}, \lambda)}{\partial\tau_{xy}}. \tag{E.7}$$

When $\tilde{\sigma}$ is independent of τ_{xy}, it is easy to see that $g_0 \equiv 0$ satisfies (E.6) as well as the boundary and initial conditions $g_0(0, t) = g_0(d/2, t) = 0$ and $g_0(y, 0) = 0$ for all $0 < y < d/2$ and $0 < t < \infty$. This corresponds to a trivial solution of (E.2) and (E.3). To assess the stability of this solution with respect to small perturbations, we linearize (E.6) around g_0 and get that a small perturbation $\tilde{g}$ has to satisfy the linear diffusion equation

$$\frac{\partial\tilde{g}}{\partial t} = D\frac{\partial^2\tilde{g}}{\partial y^2}, \tag{E.8}$$

where D is given by

$$D = v^2\hat{m}(1)\left.\frac{\partial\tilde{\sigma}(\lambda)}{\partial\lambda}\right|_{\lambda=1}. \tag{E.9}$$

When D is negative, (E.8) is a *backward-diffusion equation* (see, for instance, Novick-Cohen, 2008). In this case, a non-trivial solution of (E.8) grows without bound over time, showing that the solution g_0 is unstable whenever $\partial\tilde{\sigma}/\partial\lambda < 1$ at $\lambda = 1$.

Appendix F. Time scale associated with liquid transport and the responses for very short and long times

The purpose of this section is two-fold: i) identify an intrinsic time scale associated with liquid transport in the direction perpendicular to the channel plates; ii) use this time scale to characterize the very short- and long-time responses of the gel. For the sake of simplicity and without loss of generality, we suppose that no interfaces are present and base our analysis on the system of equations

$$\left.\begin{aligned}\lambda &= \left(1 + \frac{\partial g}{\partial y}\right)^{-1}, \\ a &= -k_1\tilde{\zeta}(\tau_{xy}, \lambda) + \tilde{\eta}(\tau_{xy}, \lambda)\frac{\partial\lambda}{\partial y}, \\ \lambda\frac{\partial g}{\partial t} &= -v^2\hat{m}(\lambda)a, \\ \tau_{xy} &= k_1\left(\frac{d}{2} - y\right)\end{aligned}\right\} \tag{F.1}$$

for all $0 < y < d/2$ and $t > 0$, where $\tilde{\zeta}$ and $\tilde{\eta}$ are given by

$$\tilde{\zeta} := \frac{\partial\tilde{\sigma}}{\partial\tau_{xy}} \qquad \text{and} \qquad \tilde{\eta} := \frac{\partial\tilde{\sigma}}{\partial\lambda}. \tag{F.2}$$

We have considered the pressure gradient in the y-direction $a = \partial p/\partial y = \partial\sigma_y/\partial y$ as a primary variable and introduced $\tilde{\zeta}$ and $\tilde{\eta}$ to simplify notation. The conditions in (F.1), which follow directly from (E.1) and (E.2), comprises a system of equations for g, λ, and a. This system has a trivial solution given by $g \equiv 0$, $\lambda \equiv 1$, and $a \equiv 0$ if and only if $\tilde{\zeta} \equiv 0$, which is tantamount to say that the normal network stress σ_y is unaffected by the shear stress τ_{xy} and hence by the applied pressure drop k_1. In the sequel, we rule out the possibility of a trivial solution.

To reach the goals set at the beginning of this section, we use the standard scaling procedure (see, for instance, Lin and Segel, 1988) as applied to (F.1). Accordingly, we begin by introducing the following dimensionless quantities:

- Independent variables:

$$\bar{y} = \frac{y}{y_c}, \qquad \bar{t} = \frac{y}{t_c}, \tag{F.3}$$

  where y_c and t_c are characteristic length and time;
- Dependent variables:

$$\bar{g}(\bar{y}, \bar{t}) = \frac{g(y_c\bar{y}, t_c\bar{t})}{g_c}, \qquad \bar{\lambda}(\bar{y}, \bar{t}) = \frac{\lambda(y_c\bar{y}, t_c\bar{t})}{\lambda_c}, \qquad \text{and} \qquad \bar{a}(\bar{y}, \bar{t}) = \frac{a(y_c\bar{y}, t_c\bar{t})}{a_c}, \tag{F.4}$$

  where g_c, λ_c, and a_c are characteristic values for g, λ, and a;
- Shear stress:

$$\bar{\tau}_{xy} = \frac{\tau_{xy}}{\tau_c}, \tag{F.5}$$

  where τ_c is a characteristic value for the shear stress τ_{xy};

- Constitutive responses:

$$\bar{\zeta}(\bar{\tau}_{xy}, \bar{\lambda}) = \frac{\tilde{\zeta}(\tau_c \bar{\tau}_{xy}, \lambda_c \bar{\lambda})}{\zeta_c}, \qquad \bar{\eta}(\bar{\tau}_{xy}, \bar{\lambda}) = \frac{\tilde{\eta}(\tau_c \bar{\tau}_{xy}, \lambda_c \bar{\lambda})}{\eta_c}, \qquad \text{and} \qquad \bar{m}(\bar{\lambda}) = \frac{\hat{m}(\lambda_c \bar{\lambda})}{m_c}, \tag{F.6}$$

where ζ_c, η_c, and m_c are characteristic values for response functions $\tilde{\zeta}$, $\tilde{\eta}$, and $\hat{m}$.

With the quantities introduced above, we can write the dimensionless version of (F.1) as

$$\left. \begin{aligned} \bar{\lambda} &= \lambda_c^{-1} \left(1 + \frac{g_c}{y_c} \frac{\partial \bar{g}}{\partial \bar{y}} \right)^{-1}, \\ \bar{a} &= -\frac{k_1 \zeta_c}{a_c} \bar{\zeta}(\bar{\tau}_{xy}, \bar{\lambda}) + \frac{\eta_c \lambda_c}{a_c y_c} \bar{\eta}(\bar{\tau}_{xy}, \bar{\lambda}) \frac{\partial \bar{\lambda}}{\partial \bar{y}}, \\ \bar{\lambda} \frac{\partial \bar{g}}{\partial \bar{t}} &= -\frac{t_c v^2 m_c a_c}{\lambda_c g_c} \bar{m}(\bar{\lambda}) \bar{a}, \\ \bar{\tau}_{xy} &= \frac{k_1 d}{2\tau_c} \left(1 - \frac{2 y_c \bar{y}}{d} \right) \end{aligned} \right\} \tag{F.7}$$

for all $0 < \bar{y} < d/(2y_c)$ and $0 < \bar{t} < \infty$. In the sequel we address the question on how the characteristic scales appearing in (F.7) are to be selected.

We begin by stating some conditions to be satisfied by the characteristic scales corresponding to the unknown variables g, λ, and a, and constitutive functions $\tilde{\zeta}$, $\tilde{\eta}$, and $\hat{m}$. Accordingly, we consider that g_c, λ_c, and a_c must be consistent with the conditions

$$g_c \geq |g|_{\max}, \qquad \lambda_c \geq \lambda_{\max}, \qquad \text{and} \qquad a_c \geq |a|_{\max}, \tag{F.8}$$

where $h_{\max}$ denotes the maximum pointwise value of h attained during the gel transition from its initial to the final and steady state.[10] We also consider that ζ_c, η_c, and m_c must comply with the conditions

$$\zeta_c \geq \max_{(\tau_{xy}, \lambda) \in C} |\tilde{\zeta}(\tau_{xy}, \lambda)|, \qquad \eta_c \geq \max_{(\tau_{xy}, \lambda) \in C} |\tilde{\eta}(\tau_{xy}, \lambda)|, \qquad \text{and} \qquad m_c \geq \hat{m}(\lambda_{\max}) \tag{F.10}$$

where $C = \{(\tau_{xy}, \lambda) : 0 \leq \tau_{xy} \leq k_1 d/2, \lambda_{\min} \leq \lambda \leq \lambda_{\max}\}$ and $\lambda_{\min}$ is the minimum pointwise value of λ achieved during the gel evolution from its initial to the final state. To write (F.10)$_3$, we have used the reasonable assumption that fluid mobility decreases with the polymer content and hence increases with λ.

We now turn to the characteristic scales selection per se. First, we observe that natural choices for the length and shear-stress scales are $y_c = d/2$ and $\tau_c = k_1 d/2$, in which case $\bar{y} \in [0, 1]$ and $\bar{\tau}_{xy} = 1 - \bar{y}$. Further, in view of the fact that $g(y) \leq d/2 - y$, $g_c = d/2$ is a legitimate choice for g_c. We now consider estimates for λ_c, a_c, ζ_c, η_c, and m_c. We expect that $\lambda_{\max}$ and $\lambda_{\min}$, the greatest and lowest values of the stretch λ experienced by the gel, are achieved under steady-state conditions at, for definiteness, $y = d/2$ and $y = 0$, respectively. Hence, $\lambda_{\max}$ and $\lambda_{\min}$ can be obtained by solving $\tilde{\sigma}(0, \lambda_{\max}) = \sigma_\infty$ and $\tilde{\sigma}(\tau_c, \lambda_{\min}) = \sigma_\infty$, where σ_∞ is the steady-state constant value reached by the normal network stress σ_y. Since fluid flow in the direction perpendicular to the plates takes place in response to $\partial \sigma_y / \partial y$, it is also natural to expect that $\sigma_{\min}$ and $\sigma_{\max}$, the minimum and maximum values of σ_y attained during the evolution of the gel up to steady state, are attained as soon as k_1 is applied, that is,

$$\sigma_{\min} = \min_{0 \leq \tau_{xy} \leq \tau_c} \tilde{\sigma}(\tau_{xy}, 1) \qquad \text{and} \qquad \sigma_{\max} = \max_{0 \leq \tau_{xy} \leq \tau_c} \tilde{\sigma}(\tau_{xy}, 1).$$

Bearing in mind that $\sigma_{\min} < \sigma_\infty < \sigma_{\max}$ and that $\partial \tilde{\sigma}/\partial \lambda > 0$ on its domain of definition, it follows that λ_l and λ_u, obtained by solving

$$\tilde{\sigma}(\tau_c, \lambda_l) = \sigma_{\min} \qquad \text{and} \qquad \tilde{\sigma}(0, \lambda_u) = \sigma_{\max},$$

are such that $\lambda_l < \lambda_{\min}$ and $\lambda_u > \lambda_{\max}$. Thus, the choices

$$\lambda_c = \lambda_u, \quad \zeta_c = \max_{(\tau_{xy}, \lambda) \in C} |\tilde{\zeta}(\tau_{xy}, \lambda)|, \qquad \eta_c = \max_{(\tau_{xy}, \lambda) \in C} |\tilde{\eta}(\tau_{xy}, \lambda)|, \qquad \text{and} \qquad m_c = \hat{m}(\lambda_u),$$

for λ_c, ζ_c, η_c, and m_c, where now $C = \{(\tau_{xy}, \lambda) : 0 \leq \tau_{xy} \leq \tau_c, \lambda_l \leq \lambda \leq \lambda_u\}$, are consistent with (F.10). Finally, on considering the reasonable assumption that the maximum value of $|a|$ is achieved as soon as k_1 is applied, we come up with the following choice for a_c:

$$a_c = k_1 \max_{0 \leq \tau_{xy} \leq \tau_c} |\tilde{\zeta}(\tau_{xy}, 1)|.$$

Notice $a_c > 0$ only if the condition $\tilde{\zeta} \equiv 0$ does not hold. Otherwise, we would have $a \equiv 0$ as already discussed.

---

[10] Notice that $h_{\max}$ is the supremum of the space–time dependent function h on $\mathcal{R} := \{(y, t) : 0 \leq y \leq d/2, 0 < t < \infty\}$, that is,

$$h_{\max} = \sup_{(y,t) \in \mathcal{R}} h(y, t). \tag{F.9}$$

With the aforementioned choices, we can rewrite (F.7) as

$$
\left.\begin{aligned}
\bar{\lambda} &= \lambda_c^{-1}\left(1 + \frac{\partial \bar{g}}{\partial \bar{y}}\right)^{-1}, \\
\bar{a} &= -\frac{k_1 \zeta_c}{a_c}\bar{\zeta}(\bar{\tau}_{xy}, \bar{\lambda}) + \frac{\eta_c \lambda_c}{a_c y_c}\bar{\eta}(\bar{\tau}_{xy}, \bar{\lambda})\frac{\partial \bar{\lambda}}{\partial \bar{y}}, \\
\bar{\lambda}\frac{\partial \bar{g}}{\partial \bar{t}} &= -\frac{t_c}{t_*}\bar{m}(\bar{\lambda})\bar{a}, \\
\bar{\tau}_{xy} &= 1 - \bar{y},
\end{aligned}\right\}
\tag{F.11}
$$

for all $0 < \bar{y} < 1$ and $0 < \bar{t} < \infty$, where

$$
t_* = \frac{\lambda_c d}{2v^2 m_c a_c}
\tag{F.12}
$$

is the intrinsic time scale we were looking for. We are now in position to discuss the gel behavior for very short and long times according to which the conditions $t \in (0, t_{st})$ with $t_{st} \ll t_*$ and $t \in (t_{lt}, \infty)$ with $t_{lt} \gg t_*$ hold. For this, it suffices to consider (F.11)$_3$, the only equation in which time appears explicitly.

In the first case, after selecting $t_c = t_{st}$ it follows from (F.11)$_3$ that

$$
\bar{\lambda}\frac{\partial \bar{g}}{\partial \bar{t}} = -\varepsilon \bar{m}(\bar{\lambda})\bar{a}
\tag{F.13}
$$

for $\bar{t} \in (0, 1)$, where $\varepsilon := t_c/t_* \ll 1$. Bearing in mind that $|\bar{m}(\bar{\lambda})| \leq 1$, $|\bar{a}| \leq 1$, and that $\lambda_1/\lambda_c \leq \bar{\lambda} \leq 1$ on the corresponding domains of definition, the term preceded by the small parameter ε in (F.13) can neglected and hence

$$
\frac{\partial \bar{g}}{\partial \bar{t}} \approx 0
\tag{F.14}
$$

for $\bar{t} \in (0, 1)$. On the other hand, for very long times we select $t_c = t_{lt}$ and get from (F.11)$_3$ that

$$
\varepsilon \bar{\lambda}\frac{\partial \bar{g}}{\partial \bar{t}} = -\bar{m}(\bar{\lambda})\bar{a}
\tag{F.15}
$$

for $\bar{t} \in (1, \infty)$, where now $\varepsilon := t_*/t_c \ll 1$. Bearing in mind that $\bar{\lambda} \leq 1$ and that $\partial \bar{g}/\partial \bar{t}$ goes to zero as $\bar{t}$ goes to infinity, one may conclude that the term preceded by the small parameter ε in (F.15) can neglected and thus

$$
\bar{a} \approx 0
\tag{F.16}
$$

for $\bar{t} \in (1, \infty)$. Hence, to a first approximation, we may consider that these regimes are described by the instantaneous and steady-state responses exhibited by the gel which are, respectively, characterized by the conditions $\partial g/\partial t \equiv 0$ and $\partial p/\partial y \equiv 0$.

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