

General Nonlinear 2-Fluid Hydrodynamics of Complex Fluids and Soft Matter

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Abstract. We discuss general 2-fluid hydrodynamic equations for complex fluids, where one kind is a simple Newtonian fluid, while the other is polymeric/elastomeric, thus being applicable to polymer solutions and swollen elastomers. The procedure can easily be generalized to other complex fluid solutions. Special emphasis is laid on such nonlinearities that originate from the 2-fluid description, like the transport part of the total time derivatives. It is shown that the proper velocities, with which the hydrodynamic quantities are convected, cannot be chosen at will, since there are subtle relations among them. Within allowed combinations the convective velocities are generally material dependent. The so-called stress division problem, i.e. how the elastic stresses are distributed between the two fluids, is shown to depend partially on the choice of the convected velocities, but is otherwise also material dependent. A set of reasonably simplified equations is given as well as a linearized version of an effective concentration dynamics that may be used for comparison with experiments.

INTRODUCTION

The thermodynamic and hydrodynamic properties of multi-component complex fluids are determined by the microscopic degrees of freedom of their constituents and the coupling between these degrees of freedom. Such systems can exhibit rather rich phase behavior and dynamics, especially when one or more components is a structured or macromolecular fluid [1]. Due in part to the coupling of internal degrees of freedom, these systems can also exhibit novel flow-induced structural evolution phenomena, including shear-induced phase transformations and flow alignment of constituents on microscopic to mesoscopic length scales. Such structural evolution in turn leads to nonlinear rheological behavior, such as stress overshoots in response to imposed rates of strain, plasticity, and thixotropy.

Due to the overwhelming complexity of the microscopic description of these systems, such a detailed description is often not well suited for analysis of the macroscopic dynamical behavior. Instead, explicit macroscopic models have been developed for this purpose. Some such models have been obtained by a suitable coarse-graining procedure starting from a microscopic theory. Others are purely phenomenological models constrained only by conservation laws, symmetry considerations and thermodynamics. The so-called "two-fluid" models for binary systems of distinct components or phases are useful examples of such a macroscopic ap-

proach [2]. In the two-fluid description, each component or phase is treated as a continuum described by local thermodynamic variables (e.g. temperature, density, and relevant order parameters), and dynamical quantities (e.g. velocity or momentum). In general, these variables for the constituents are coupled. For instance, the effective friction between components in a binary fluid mixture leads to a drag force in the macroscopic description that is proportional to the local velocity difference.

Two-fluid models have been employed in many different physical contexts. The two-fluid approach is a key element of many traditional models for multi-phase flow of bubbly liquids, fluid suspensions of particulates, and binary mixtures of simple fluids [3]. Other examples in condensed matter physics include two-fluid models for superfluid helium [4], dynamics of plasmas [5], transport in superconductors [6], viscoelasticity of concentrated fluid emulsions [7], flow-induced ordering of wormlike micelle solutions [8], flow of colloidal suspensions [9]. Two-fluid models have been used extensively to model a wide range of dynamical phenomena in polymer solutions and binary blends, including the hydrodynamic modes of quiescent polymer solutions [10, 11], kinetics of polymer dissolution [12], hydrodynamics and rheology of polymer solutions and blends [13]-[19], and polymer migration and phase separation under flow [20]-[27].

These examples share certain general features. In each, two distinct species or coexisting phases (gas and liquid, normal fluid and superfluid, polymer and solvent, meso-

gens and solvent etc.) with mass densities ρ_1 and ρ_2 , which are conserved individually in the absence of chemical reactions, move with distinct velocities \vec{v}_1 and \vec{v}_2 , respectively. Due to (usually strong) internal friction, the momenta of the constituent species, $\rho_1\vec{v}_1$ and $\rho_2\vec{v}_2$, are not conserved individually. Of course, total momentum is conserved. In most cases of fluid mixtures the friction is so strong that the velocity difference $\vec{v}_1 - \vec{v}_2$ is nonzero for very short times only, i.e. it is a very rapidly relaxing quantity that is not included in the hydrodynamic description for binary mixtures. However, there are systems and situations, where the relaxation of the relative momenta is slow enough to have a significant influence even on the hydrodynamic time scale. Then a two-fluid description is appropriate and useful.

In this communication we focus on a general nonlinear two-fluid description of complex fluids, where one species is a viscous Newtonian fluid and the other a polymer or elastomer. Emphasis is placed on the rigorous derivation of the dynamic equations within the framework of hydrodynamics as contrasted to ad-hoc treatments. The resulting equations are rather general and complicated. They can and have to be simplified for special applications or systems by appropriate and well-defined approximations. One of the advantages of starting from the general theory is the possibility to identify and characterize the approximations made. The hydrodynamic method, described in some detail in [28]-[30], is quite general and rigorous, being based on symmetries, conservation laws, and thermodynamics.

The linear hydrodynamic description of a single component elastic media has been given in [31], generalized to the nonlinear domain in [32, 33], where in addition the necessary steps are described when the elastic degree of freedom is not constant (as in elastomers), but decaying in time (as in viscoelastic media). In the following sections, we provide a detailed analysis of two-fluid models for isotropic polymers and elastomers (e.g. entangled polymer solutions and gels) in a simple viscous solvent. We close with a discussion of our general results and their possible implications for experiments.

THERMODYNAMICS

The hydrodynamics of fluid mixtures as described above is governed by conservation laws (individual masses, total momentum and total energy), balance equations for the liquid crystalline degrees of freedom, for the transient elasticity of polymers and for the relaxation of relative momentum. There are different ways of writing the appropriate equations. One popular choice is to use equations for individual mass densities and individual momentum densities, another to use the mass density

and one concentration variable and the total momentum density and the relative velocity difference. Since they both have their advantages and disadvantages we will present both ways of description, and show how they are connected. We will use an isotropic elastomer (or viscoelastic media) as the second, complex fluid.

The starting point of any macroscopic description is the total energy E of the system as a function of all the relevant variables. Since the energy is a first order Eulerian form of the extensive quantities, we can write

$$\begin{aligned} E &= \varepsilon V = \int \varepsilon dV \\ &= E(M_1, M_2, V, \vec{G}_1, \vec{G}_2, S, M_2 U_{ij}) \end{aligned} \quad (1)$$

The masses, M_1, M_2 and momenta \vec{G}_1, \vec{G}_2 of species 1 and 2 are related to the appropriate (volume) densities by $\rho_1 = M_1/V$, $\rho_2 = M_2/V$, $\vec{g}_1 = \vec{G}_1/V = \rho_1\vec{v}_1$, $\vec{g}_2 = \vec{G}_2/V = \rho_2\vec{v}_2$, thus defining the two velocities $\vec{v}_{1,2}$ whose components we write as $v_i^{(1,2)}$. The entropy density is $\sigma = S/V$. The elastic degree of freedom of species 2 is described by the Eulerian strain tensor U_{ij} [32], which is symmetric and often related to a displacement vector \vec{u} by $2U_{ij} = \nabla_i u_j + \nabla_j u_i + (\nabla_k u_i)(\nabla_k u_j)$. Introducing thermodynamic derivatives (partial derivatives where all other variables are kept fixed) we define temperature T , thermodynamic pressure p , chemical potentials μ_1, μ_2 and velocities \vec{v}_1, \vec{v}_2 of the two fluids, as well as the elastic stress Φ_{ij} conjugate to the elastic strain

$$\begin{aligned} T &= \frac{\partial E}{\partial S} = \frac{\partial \varepsilon}{\partial \sigma}, & \mu_1 &= \frac{\partial E}{\partial M_1} = \frac{\partial \varepsilon}{\partial \rho_1}, \\ \mu_2 &= \frac{\partial E}{\partial M_2} = \frac{\partial \varepsilon}{\partial \rho_2}, & p &= -\frac{\partial E}{\partial V}, \\ \vec{v}_1 &= \frac{\partial E}{\partial \vec{G}_1} = \frac{\partial \varepsilon}{\partial \vec{g}_1}, & \vec{v}_2 &= \frac{\partial E}{\partial \vec{G}_2} = \frac{\partial \varepsilon}{\partial \vec{g}_2}, \\ \Phi'_{ij} &= \frac{\partial E}{\partial (M_2 U_{ij})} = \frac{\partial \varepsilon}{\partial (\rho_2 U_{ij})} \equiv \rho_2^{-1} \Phi_{ij} \end{aligned} \quad (2)$$

Expanding eq.(1) into first order differentials, the condition $dV = 0$ leads to an expression for the pressure

$$p = -\varepsilon + T\sigma + \rho_1\mu_1 + \rho_2\bar{\mu}_2 + \vec{v}_1 \cdot \vec{g}_1 + \vec{v}_2 \cdot \vec{g}_2 \quad (3)$$

where we have introduced the effective chemical potential of the elastomer $\bar{\mu}_2 = \mu_2 + \rho^{-1}\Phi_{ij}U_{ij}$. In addition, the differentials are related by the Gibbs relation

$$\begin{aligned} d\varepsilon &= Td\sigma + \mu_1 d\rho_1 + \bar{\mu}_2 d\rho_2 + \vec{v}_1 \cdot d\vec{g}_1 + \vec{v}_2 \cdot d\vec{g}_2 \\ &\quad + \Phi_{ij} dU_{ij} \end{aligned} \quad (4)$$

From eqs.(3, 4) the expression for the differential pressure results (Gibbs-Duhem relation) that is useful in

switching from pressure to chemical potentials or vice versa

$$dp = \sigma dT + \rho_1 d\mu_1 + \rho_2 d\bar{\mu}_2 + \bar{g}_1 \cdot d\bar{v}_1 + \bar{g}_2 \cdot d\bar{v}_2 - \Phi_{ij} dU_{ij} \quad (5)$$

A second set of equations is obtained by switching to the total density, $\rho = \rho_1 + \rho_2$, and the total momentum, $\bar{g} = \bar{g}_1 + \bar{g}_2 = \rho_1 \bar{v}_1 + \rho_2 \bar{v}_2$, which are the sums of the original quantities and which are both conserved quantities. The two-fluid nature has then to be represented by additional variables. A natural choice seems to be the use of the density and momentum differences. However the latter choice is problematic, since it necessarily implies the conjugate quantities also to be the (arithmetic) sums and differences of the original conjugate quantities. Thus, the conjugate to \bar{g} would be $\bar{v}_1 + \bar{v}_2$, which does not reflect correctly the possible one-fluid limits $\rho_1 \rightarrow 0$ or $\rho_2 \rightarrow 0$. The physically acceptable conjugate to the total momentum is the mean velocity \bar{v} defined by $\rho^{-1} \bar{g}$. Insisting on \bar{v} , the mean velocity, to be the conjugate of the total momentum \bar{g} , the choice of the remaining variable describing the different velocities is severely limited. Compatibility with (4) allows as variable only the velocity difference $\bar{w} \equiv \bar{v}_1 - \bar{v}_2$ (with $\bar{m} \equiv \rho^{-1} \rho_1 \rho_2 \bar{w}$ as conjugate quantity) or more generally $\alpha \bar{w}$ as variable with $\alpha^{-1} \rho_1 \rho_2 \rho^{-1} \bar{w}$ as conjugate, where α can be freely chosen. There is no a-priori advantage for any of the choices and we will stick to $\alpha = 1$.¹ From $\bar{w} = \bar{g}_1/\rho_1 - \bar{g}_2/\rho_2$ one gets

$$\bar{v}_1 = \rho^{-1} \bar{g} + (1 - \phi) \bar{w}, \quad \bar{v}_2 = \rho^{-1} \bar{g} - \phi \bar{w} \quad (6)$$

The representation of the two different densities is less problematic. A convenient choice for that variable is the concentration, $\phi = \rho_1/\rho$, with $\rho_2/\rho = 1 - \phi$. If the expansion coefficients of the two fluids are the same, ϕ can be interpreted as the volume fraction as well. Instead of ϕ one could have used, e.g. the density difference $\rho_1 - \rho_2$ (or any other linear combination of ρ_1 and ρ_2 different from ρ) as variable without much change.

After some trivial algebra eqs.(3-5) can be written in the new variables as

$$p = -\varepsilon + T\sigma + \rho\mu + \rho^{-1} \bar{g}^2 \quad (7)$$

$$d\varepsilon = T d\sigma + \Pi' d\phi + \mu d\rho + \bar{v} \cdot d\bar{g} + \bar{m} \cdot d\bar{w} + \Phi_{ij} dU_{ij} \quad (8)$$

$$dp = \sigma dT + \rho d\mu + \bar{g} \cdot d\bar{v} - \bar{m} \cdot d\bar{w} - \Pi' d\phi - \Phi_{ij} dU_{ij} \quad (9)$$

¹ The choice $\alpha = \rho_1 \rho_2 \rho^{-1}$ would just interchange the roles of \bar{w} and \bar{m} as variable and conjugate.

where we have introduced the relative pressure Π' and the total chemical potential μ

$$\begin{aligned} \Pi' &= \rho(\mu_1 - \bar{\mu}_2) + \bar{w} \cdot \bar{g} + \rho \bar{w}^2 (1 - 2\phi) \equiv \rho \Pi \\ \mu &= \mu_1 \phi + \bar{\mu}_2 (1 - \phi) + \bar{w}^2 \phi (1 - \phi) \end{aligned} \quad (10)$$

or vice versa

$$\begin{aligned} \mu_1 &= \mu + \rho^{-1} \rho_2 (\Pi - \bar{w} \cdot \bar{v}_1) \\ \mu_2 &= \mu - \rho^{-1} \rho_1 (\Pi + \bar{w} \cdot \bar{v}_2) \end{aligned} \quad (11)$$

with the mean velocity \bar{v} and the weighted relative momentum \bar{m} given by

$$\begin{aligned} \bar{v} &= \phi \bar{v}_1 + (1 - \phi) \bar{v}_2 = \rho^{-1} (\bar{g}_1 + \bar{g}_2) \\ \bar{m} &= \rho (1 - \phi) \phi \bar{w} = (\rho_2 \bar{g}_1 - \rho_1 \bar{g}_2) \rho^{-1} \end{aligned} \quad (12)$$

The Gibbs relations connects variables that show different rotational behavior. Energy, entropy, the densities and the concentration are scalar quantities that do not change under (rigid) rotations, i.e. $d\varepsilon = d\sigma = d\rho = d\rho_1 = d\rho_2 = d\phi = 0$. The vectors and tensors are transformed according to $dv_i^{(1,2)} = \Omega_{ij} v_j^{(1,2)}$, $dg_i = \Omega_{ij} g_j$, $dw_i = \Omega_{ij} w_j$, $dU_{ij} = \Omega_{jk} U_{ik} + \Omega_{ik} U_{kj}$, where $\Omega_{ij} = -\Omega_{ji}$ is any constant antisymmetric matrix. The rotational invariance of the Gibbs relation (4,8) then leads to the relation

$$U_{ik} \Phi_{kj} = U_{jk} \Phi_{ki} \quad (13)$$

which has to be fulfilled by the conjugate quantities. There are no contributions from the momenta and velocities, since $\bar{g} \parallel \bar{v}$, $\bar{w} \parallel \bar{m}$, and $\bar{g}_{1,2} \parallel \bar{v}_{1,2}$. Relation (13) is useful for reformulating the total stress tensor (see below), in particular to symmetrize it explicitly.

Having set up the thermodynamics of the relevant variables we are now in a position to establish the structure of the dynamic equations.

SIMPLIFIED ELASTOMERIC TWO-FLUID EQUATIONS

Recently we have derived the most general and complete set of 2-fluid equations [34]. These equations are for most purposes unnecessarily complicated and can be simplified using reasonable assumptions. Starting from the correct general equations such assumptions, clearly spelled out, lead to controlled approximations and to a set of 2-fluid equations, whose limitations and implicit assumptions are clear and well defined in contrast to most ad-hoc approaches.

Here we want to display explicitly 2-fluid hydrodynamics under the following assumptions:

a) convection with natural velocities (for U_{ij} , \bar{g}_2 , ρ_2 and

\vec{g}_1 , ρ_1 this is \vec{v}_2 and \vec{v}_1 , respectively);
b) the linearized elastic force acts on the elastomeric fluid (index 2) only;
c) global incompressibility, $\delta\rho = 0$ (i.e. $\delta\rho_1 = -\delta\rho_2$); and
d) linearizing the phenomenological dissipative currents, but keeping quadratic nonlinearities otherwise. Then the following set of equations is obtained:
The incompressibility condition (in 3 equivalent versions),

$$0 = \operatorname{div} \vec{v} \quad (14)$$

$$0 = \vec{w} \cdot \vec{\nabla} \rho_1 + \rho_1 \operatorname{div} \vec{v}_1 + \rho_2 \operatorname{div} \vec{v}_2 \quad (15)$$

$$0 = \vec{w} \cdot \vec{\nabla} \phi + \phi \operatorname{div}(1 - \phi) \vec{w} - (1 - \phi) \operatorname{div} \phi \vec{w} \quad (16)$$

the concentration dynamics (in 3 equivalent versions),

$$\begin{aligned} \dot{\phi} + \nabla_i (\phi v_i + \phi(1 - \phi) w_i) - d_{ij} \nabla_i \nabla_j (\mu_1 - \bar{\mu}_2) \\ - \phi(1 - \phi) d_{ij}^{(T)} \nabla_i \nabla_j T = 0 \end{aligned} \quad (17)$$

$$\begin{aligned} \dot{\rho}_1 + \vec{v}_1 \cdot \vec{\nabla} \rho_1 + \rho_1 \operatorname{div} \vec{v}_1 - \rho d_{ij} \nabla_i \nabla_j (\mu_1 - \bar{\mu}_2) \\ - \frac{\rho_1 \rho_2}{\rho} d_{ij}^{(T)} \nabla_i \nabla_j T = 0 \end{aligned} \quad (18)$$

$$\begin{aligned} \dot{\rho}_2 + \vec{v}_2 \cdot \vec{\nabla} \rho_2 + \rho_2 \operatorname{div} \vec{v}_2 + \rho d_{ij} \nabla_i \nabla_j (\mu_1 - \bar{\mu}_2) \\ + \frac{\rho_1 \rho_2}{\rho} d_{ij}^{(T)} \nabla_i \nabla_j T = 0 \end{aligned} \quad (19)$$

the entropy dynamics (heat conduction equation),

$$\begin{aligned} \dot{\sigma} + v_i \nabla_i \sigma + \frac{\beta}{\rho} \nabla_i (\rho_1 \rho_2 w_i) - \kappa_{ij} \nabla_i \nabla_j T \\ - \frac{\rho_1 \rho_2}{\rho} d_{ij}^{(T)} \nabla_i \nabla_j (\mu_1 - \bar{\mu}_2) = 0 \end{aligned} \quad (20)$$

and the elasticity dynamics

$$\begin{aligned} \dot{U}_{ij} + v_k^{(2)} \nabla_k U_{ij} - \frac{1}{2} (\nabla_j v_i^{(2)} + \nabla_i v_j^{(2)}) \\ - \frac{\rho_1}{2} (w_i \nabla_j + w_j \nabla_i) \ln \frac{\rho_2}{\rho} + U_{ki} \nabla_j v_k^{(2)} + U_{kj} \nabla_i v_k^{(2)} \\ + \zeta_l \delta_{ij} \Phi_{kk} + \zeta_{lr} (\Phi_{ij} - \frac{1}{3} \delta_{ij} \Phi_{kk}) - \xi_1 \delta_{ij} \Delta \Phi_{kk} \\ - \xi_2 \Delta \Phi_{ij} - \xi_3 (\nabla_i \nabla_j \Phi_{kk} + \delta_{ij} \nabla_k \nabla_l \Phi_{kl}) \\ - \xi_4 (\nabla_i \nabla_k \Phi_{jk} + \nabla_j \nabla_k \Phi_{ik}) = 0 \end{aligned} \quad (21)$$

Since elasticity is assumed to be related to fluid 2, the linear and quadratic couplings to flow are related to velocity \vec{v}_2 only. The quadratic couplings are of the "lower convected" type, well-known for the single fluid description [32] - [35] and are of great importance in the viscoelastic case. In addition, there are nonlinear couplings to the concentration variable (the cubic one has been suppressed), which are not possible in a 1-fluid description. The ζ -terms describe relaxation of elastic strains and are

absent in a permanent network, where only the strain diffusion terms ($\sim \xi_{1,2,3,4}$) are present. Despite the global incompressibility assumption $\operatorname{div} \vec{v} = 0$, the trace of the elastic tensor, U_{kk} , does not vanish even in linear order, since neither $\operatorname{div} \vec{v}_1 = 0$, nor $\operatorname{div} \vec{v}_2 = 0$, nor $\operatorname{div} \vec{w} = 0$, generally.

For the momentum balance of the two different species we get

$$\begin{aligned} \rho_1 \dot{v}_i^{(1)} + \rho_1 v_j^{(1)} \nabla_j v_i^{(1)} + \frac{\rho_1}{\rho} \nabla_i (p + \frac{1}{2} \rho_2 w_j (v_j^1 + v_j^2)) \\ + \frac{\rho_1 \rho_2}{\rho} \nabla_i (\mu_1 - \bar{\mu}_2) + \frac{\rho_1}{\rho} \Phi_{kj} \nabla_i U_{kj} - \rho_1 \Phi_{ij} \nabla_j \ln \frac{\rho_2}{\rho} \\ + \frac{\rho_1 \rho_2}{\rho} \beta \nabla_i T + \xi \rho_1 \rho_2 w_i - v_{ijkl}^{(1)} \nabla_j \nabla_l v_k^{(1)} \\ - v_{ijkl}^{(2)} \nabla_j \nabla_l v_k^{(2)} = 0 \end{aligned} \quad (22)$$

$$\begin{aligned} \rho_2 \dot{v}_i^{(2)} + \rho_2 v_j^{(2)} \nabla_j v_i^{(2)} + \frac{\rho_2}{\rho} \nabla_i (p - \frac{1}{2} \rho_1 w_j (v_j^1 + v_j^2)) \\ - \frac{\rho_1 \rho_2}{\rho} \nabla_i (\mu_1 - \bar{\mu}_2) - \frac{\rho_1}{\rho} \Phi_{kj} \nabla_i U_{kj} + \rho_1 \Phi_{ij} \nabla_j \ln \frac{\rho_2}{\rho} \\ - \frac{\rho_1 \rho_2}{\rho} \beta \nabla_i T - \nabla_j \Phi_{ij} + \nabla_j (\Phi_{jk} U_{ik} + \Phi_{ik} U_{jk}) \\ - \xi \rho_1 \rho_2 w_i - v_{ijkl}^{(2)} \nabla_j \nabla_l v_k^{(2)} - v_{ijkl}^{(1)} \nabla_j \nabla_l v_k^{(1)} = 0 \end{aligned} \quad (23)$$

The ξ -terms describe the coupling of the the two momenta due to the difference of the two velocities (friction). Note that although we made the approximation that the linear elastic stress does only act on fluid 2, there are inevitably nonlinear contributions to fluid 1, too. There is also a (nonlinear) coupling of fluid 1 to the concentration, if elastic distortions are present.

In order to facilitate actual calculations we also give eqs.(22,23) as dynamic equations for the total momentum and for the relative velocity

$$\begin{aligned} \rho \dot{v}_i + \nabla_i p + \nabla_j (\rho v_i v_j + \frac{\rho_1 \rho_2}{\rho} w_i w_j) + 2 \nabla_j (\Phi_{jk} U_{ik}) \\ - \nabla_j \Phi_{ij} - v_{ijkl} \nabla_j \nabla_l v_k - \frac{\rho_1 \rho_2}{\rho} v_{ijkl}^{(c)} \nabla_j \nabla_l w_k = 0 \end{aligned} \quad (24)$$

$$\begin{aligned} \dot{w}_i + \left(v_j + \frac{\rho_2 - \rho_1}{\rho} w_j \right) \nabla_j w_i + \rho \xi w_i \\ + \nabla_i \left(\mu_1 - \bar{\mu}_2 + \vec{v} \cdot \vec{w} + \frac{\rho_2 - \rho_1}{2\rho} \vec{w}^2 \right) + \nabla_j \frac{1}{\rho_2} \Phi_{ij} \\ + \frac{1}{\rho_2} \Phi_{kj} \nabla_i U_{kj} - \frac{2}{\rho_2} \nabla_j (\Phi_{kj} U_{ik}) \\ - \frac{\rho_1 \rho_2}{\rho} v_{ijkl}^{(m)} \nabla_l \nabla_j w_k - v_{ijkl}^{(c)} \nabla_j \nabla_l v_k = 0 \end{aligned} \quad (25)$$

From (24) angular momentum conservation is obvious, since the total stress tensor (defined by $\rho v_i + \nabla_j \sigma_{ij} = 0$ in the incompressible limit) is symmetric due to the relation (13), if the viscosities are of the usual form $v_{ijkl} =$

$v_{\perp}(\delta_{ik}\delta_{jl} + \delta_{jk}\delta_{il} - \frac{2}{3}\delta_{ij}\delta_{kl}) + v_b\delta_{ij}\delta_{kl}$. Note that the viscosities introduced in (22,23), $v_{ijkl}^{(1,2,12)}$, are different from v_{ijkl} and $v_{ijkl}^{(c,m)}$ used in (24,25).

In order to conserve the global incompressibility condition for all times, i.e. $\text{div}\vec{v} = 0$, the pressure has to fulfill the relation

$$\begin{aligned} \Delta p = & -\nabla_i\nabla_j(\rho_1 v_i^{(1)} v_j^{(1)} + \rho_2 v_i^{(2)} v_j^{(2)}) + \nabla_i\nabla_j\Phi_{ij} \\ & -\nabla_i\nabla_j(\Phi_{kj}U_{ik} + \Phi_{ik}U_{jk}) + v_{ijkl}\nabla_i\nabla_j\nabla_l v_k \\ & + \rho_1\rho_2\rho^{-1}v_{ijkl}^{(c)}\nabla_i\nabla_j\nabla_l w_k \end{aligned} \quad (26)$$

In contrast to 1-fluid descriptions for simple fluids, where the incompressibility condition leads to a considerable mathematical simplification, this is no longer the case for a 2-fluid description due the complicated form of (26), even if incompressibility is a very good approximation in physical terms. In particular, Δp is not only connected to elastic compressions (U_{kk}), but also to shear deformations, even in linear order.

The static relations between the conjugate quantities to the variables close the system of equations

$$\delta T = TC_V^{-1}\delta\sigma + \alpha_{\phi}^{-1}\delta\phi + \alpha_3^{-1}U_{kk} \quad (27)$$

$$\begin{aligned} \Phi_{ij} = & c_{lr}(U_{ij} - \frac{1}{3}\delta_{ij}U_{kk}) + c_l U_{kk} \\ & + \alpha_3^{-1}\delta\sigma + \rho^{-1}\kappa_u^{-1}\delta\phi \end{aligned} \quad (28)$$

$$\begin{aligned} \delta(\mu_1 - \bar{\mu}_2) = & \rho^{-1}\kappa_{\phi}^{-1}\delta\phi + \rho^{-1}\alpha_{\phi}^{-1}\delta\sigma \\ & + \kappa_u^{-1}U_{kk} \end{aligned} \quad (29)$$

with $\delta\phi = \rho^{-1}\delta\rho_1 = -\rho^{-1}\delta\rho_2$. Note that $\delta\mu$ is not needed, but follows from δp via eq. (5) or (9).

DISCUSSION

Within the general framework of hydrodynamics and thermodynamics we have set up a consistent nonlinear 2-fluid description of complex fluids, in particular for polymer solutions or swollen elastomers. Such a general theory [34] determines the frame for any ad-hoc model, which has to be a special case of the general one. The comparison with the general theory also reveals implicit and explicit assumptions, approximations and possible generalizations of a given model. A ‘‘simple’’ or ‘‘natural’’ choice in a given model may not be mandatory, but rather imply a presumption.

Quite generally we find that neither the velocity, with which a certain variable is convected, nor the stress division between the different fluids can be determined by general principles, but is rather system or material dependent. On the other hand, there are certain restrictions and interrelations among the convective velocities

and other physical effects that limit the possible choices. For the two densities ρ_1, ρ_2 e.g., the ‘‘natural’’ choice for the convection velocities (taken in the preceding section) seems to be their native velocities \vec{v}_1 and \vec{v}_2 , respectively. This implies that the total density is convected with the mean velocity \vec{v} (as required by mass transport, but manifest only, if the incompressibility assumption is lifted), while the concentration ϕ is convected with $(1/\rho)(\rho_2\vec{v}_1 + \rho_1\vec{v}_2)$. Another obvious (‘‘simple’’) choice would be the mean velocity as convection velocity for both, the total density as well as the concentration implying that also ρ_1 and ρ_2 are convected with \vec{v} . However, the actual convection velocity depends on the value of a material dependent (reactive) flow parameter (call it γ) and is not restricted to the two choices mentioned above ($\gamma = 0$ and -1 for the ‘‘natural’’ and the ‘‘simple’’ one).

In the case of visco-elastic and elastic media, which are described by a dynamic equation for the (Eulerian) strain tensor U_{ij} , there are two velocities involved. One is the usual convection velocity ($v_k\nabla_k U_{ij}$) and the other one occurs in the ‘‘lower convected’’ part ($U_{kj}\nabla_i v_k + U_{ki}\nabla_j v_k$). There is no fundamental reason for the two to be equal, nor to be one of the obvious choices (\vec{v} or \vec{v}_2).

For the evolution equations of the momenta special care has to be taken to get a description, which is compatible with general laws. The currents and quasi-currents that enter the description in terms of either the total momentum and the velocity difference or in terms of the two individual momenta are not the same comparing eqs.(22, 23) with (24, 25). In particular, the momenta $\vec{g}_{1,2}$ are convected with $\vec{v}_{1,2}$ implying that the total momentum \vec{g} and the relative velocity \vec{w} are convected with \vec{v} and $(1/\rho)(\rho_2\vec{v}_1 + \rho_1\vec{v}_2)$, respectively. Instead of this ‘‘natural’’ choice there are other possibilities governed by some phenomenological parameters (independent from γ), e.g. the ‘‘simple’’ choice that all 4 quantities are convected with \vec{v} . Even the convection of the entropy can be tuned by choosing a parameter $\beta \equiv \beta_0 + \beta_{00}\sigma$ where $\beta_{00} = 1/\rho_1, = 0, = -1/\rho_2$ leads to the convective velocity to be $\vec{v}_1, \vec{v}, \vec{v}_2$, respectively.

In the preceding section it was assumed that (linear) elastic stress is carried only by fluid 2. Generally, however, the distribution of the elastic stress among the two fluids is governed by a phenomenological coefficient (call it $\lambda^{(U)}$). For, respectively, $2\lambda^{(U)} = 1/\rho_2, = 0, = -1/\rho_1$, the elastic stress is carried only by fluid 2, is equally distributed between 1 and 2, or carried only by fluid 1; but $\lambda^{(U)}$ can have any value in between.

A prominent feature of the 2-fluid description is the coupling of the concentration dynamics to the velocity difference. Linearizing and Fourier transforming the dynamic equations, thus eliminating \vec{w} from e.g. the concentration dynamics and neglecting fourth order gradient

terms we get

$$i\omega\phi - d^{eff}\Delta\Pi - \frac{\rho_1\rho_2}{\rho^2}d^{(T)eff}\Delta T - 2\lambda^{(\phi)}\nabla_i\nabla_j\Phi_{ij} = 0 \quad (30)$$

with frequency dependent effective diffusion and thermo-diffusion coefficients

$$d^{eff} = d + \frac{\rho_1\rho_2}{\rho^2} \frac{(\gamma+1)^2}{\rho\xi + i\omega} \quad (31)$$

$$d^{(T)eff} = d^{(T)} + \frac{\beta(\gamma+1)}{\rho\xi + i\omega} \quad (32)$$

and the dynamic coupling to the elastic degree of freedom by

$$\lambda^{(\phi)} = \frac{\rho_1\rho_2}{\rho} \lambda^{(U)} \frac{1+\gamma}{\rho\xi + i\omega} \quad (33)$$

where the dispersion step around $\omega \approx \rho\xi$ is due to the friction ($\sim \xi$) between the two fluid momenta. Again these possible additions to the concentration dynamics, however, depend on the choices for the convection velocities (i.e. on γ and β) as well as on the way how the elastic stress has been divided among the two fluids (on $\lambda^{(U)}$). E.g. if the stress is equally distributed among the 2 fluids and if both densities are convected with \vec{v} , there are no frequency dependent additions to the concentration dynamics at all.

Recently, 2-fluid descriptions of diffusion in polymeric systems have been given [36, 37] based on the GENERIC approach making use of Poisson brackets. A detailed comparison with these formulations is beyond the scope of this manuscript and will be discussed elsewhere.

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