Benchmark Calculations in Eccentric Taylor-Couette Systems

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3.1. Parameters and their assigned values for preconditioned inexact Newton method 45
Circular flow in the annular gap between eccentric cylinders is commonly used as a benchmark problem for testing numerical methods and fluid models. The eccentric cylinder system is also of relevance to the industrial processing of complex fluids because of its highly distributive and dispersive mixing performance. This dissertation focuses on a steady eccentric Taylor-Couette flow of polymeric solutions, melts, and blends with droplet morphology. To describe the rheology and microstructural dynamics of these polymeric fluids, we adopted differential-type constitutive equations developed by other researchers in the past using the generalized bracket approach of nonequilibrium thermodynamics. This dissertation discusses the development of a novel numerical approach for solving the benchmark problem. The proposed approach is unique in that the nonlinear system of discretized algebraic flow equations is solved iteratively using a Newton-Krylov method along with an inverse-based incomplete lower-upper preconditioner. The iterative solution technique is validated by solving the benchmark problem for the upper-convected Maxwell model at the largest Deborah number for which a boundary-layer solution is available in literature at a moderate eccentricity ratio of the cylinder system. The excellent agreement of the results with the numerical data from the literature encouraged further application of the preconditioned Newton-Krylov method in benchmark simulations. In addition, we solved the benchmark problem for other polymeric fluid models at different model parameter values. A large eccentricity ratio was chosen for the cylinder system in order to allow flow recirculation to occur. We detected several interesting phenomena caused by the highly eccentric configuration of the cylinder system and the viscoelastic nature of the polymeric fluids. The model predictions provide a more fundamental understanding of the interrelationship between the rheological behavior and the microstructural dynamics of polymeric fluids under industrially relevant flow conditions. However, experimental data would be needed to verify the model predictions. The results of this dissertation should encourage further investigation of viscoelastic fluids having a complex microstructure in an eccentric annular flow field.

**Keywords:** Newton-Krylov methods; ILU preconditioning; benchmark calculations; eccentric cylinders; nonequilibrium thermodynamics; complex fluids
Für das Testen von numerischen Verfahren und Flüssigkeitsmodellen wird häufig eine kreisför-
nige Strömung in einem Ringspalt zwischen exzentrischen Zylindern als Benchmark-Problem
verwendet. Aufgrund ihres stark distributiv- und dispersiv-mischenden Betriebsverhaltens
spielt die exzentrische Zylinderanordnung auch bei der industriellen Bearbeitung komple-
xer Flüssigkeiten eine bedeutende Rolle. Die vorliegende Dissertation beschäftigt sich mit
einer stationären exzentrischen Taylor-Couette-Strömung für Polymerlösungen, -schmelzen
und -gemische mit Tropfenmorphologie. Um die Rheologie und Mikrostrukturdynamik dieser
polymeren Flüssigkeiten zu beschreiben, haben wir konstitutive Gleichungen differenzieller
Art übernommen, die von anderen Wissenschaftlern unter Verwendung des verallgemeinerten
Klammeransatzes der Nichtgleichgewichtsthermodynamik entwickelt wurden. Die vorliegen-
de Dissertation diskutiert die Entwicklung eines neuartigen numerischen Ansatzes zur Lösung
der Benchmark-Probleme. Der vorgeschlagene Ansatz ist einzigartig in der Hinsicht, dass das
nichtlineare System diskreter algebraischer Strömungsgleichungen iterativ unter Verwendung
eines Newton-Krylov-Verfahrens und eines Vorkonditioniers in Form einer invers-basierten
unvollständigen Dreiecksszerlegung gelöst wird. Die Validierung der iterativen Lösungstechn-
nik erfolgte, indem das Benchmark-Problem für das obere konvektive Maxwell-Modell bei
der größten Deborah-Zahl gelöst wurde, für welche in der Literatur eine Grenzschichtlö-
sung bei einem moderaten Exzentrizitätsverhältnis der Zylinderanordnung existiert. Die her-
vorragende Übereinstimmung der Ergebnisse mit den numerischen Literaturdaten ermutigte
die weitere Verwendung des konditionierten Newton-Krylov-Verfahrens in Benchmark-
Simulationen. Außerdem haben wir das Benchmark-Problem für andere polymerische Flüssig-
keitsmodelle bei unterschiedlichen Modellparametern gelöst. Für die Zylinderanordnung
wurde ein großes Exzentrizitätsverhältnis gewählt, um das Auftreten einer Rezirkulationsströ-
mung zu ermöglichen. Wir haben zahlreiche interessante Phänomene entdeckt, die von der
stark exzentrischen Anordnung des Zylindersystems und den viskoelastischen Eigenschaften
der polymeren Flüssigkeiten verursacht werden. Die Modellvorhersagen liefern ein grundlie-
gendes Verständnis der wechselseitigen Beziehung zwischen dem rheologischen Verhalten
und der Mikrostrukturdynamik polymerer Flüssigkeiten unter industriell bedeutsamen Ström-
ungsbedingungen. Jedoch sind experimentelle Daten für die Bestätigung der Modellvorher-
sagen unerlässlich. Die Ergebnisse dieser Dissertation sollen zu weiteren Untersuchungen von
viskoelastischen Flüssigkeiten mit komplexer Mikrostruktur in exzentrischen Ringströmungs-
feldern motivieren.
1. Introduction

Most types of foods, cosmetics, and pharmaceutics belong to the class of complex fluids. Furthermore, all fluids having a microstructure also belong to this class of fluids. When complex fluids are subjected to flow, the dynamics of their microstructure typically produces an unusual response. A majority of complex fluids exhibit viscoelastic behavior. Viscoelastic materials have properties that are intermediate between those of elastic solids and viscous fluids. Their most characteristic property is that of limited memory; that is, they partially resume their initial state of deformation upon removal of an externally applied force. Other characteristic properties include a variable viscosity during deformation and the occurrence of nontrivial normal stresses in simple shear. To successfully develop and optimize industrial processes and products involving the use of complex fluids, detailed knowledge about the rheology-microstructure relationships of complex fluids under industrially relevant flow conditions is crucial.

Nonequilibrium thermodynamics offers a suitable framework for modeling complex fluids. The starting point of any thermodynamic modeling is the selection of appropriate state variables of the system under investigation. The selection of state variables that describe the microstructural dynamics of complex fluids determines the level of description. Complex fluids can be modeled using either coarse-grained or fine-grained approaches. In coarse-grained approaches, the microstructural dynamics of complex fluids is described in terms of mesoscopic variables that span a three-dimensional geometrical space and time. In contrast, in fine-grained approaches, the microstructural dynamics of complex fluids is described in terms of microscopic variables of higher dimensionality. Coarse-grained models are preferred in benchmark simulations, because the computational time and memory required to solve microscopic models for benchmark flows are significantly higher. Examples of popular theories include the matrix model of thermodynamically driven systems [51, 52, 53], the general equation for the nonequilibrium reversible-irreversible coupling (GENERIC) framework [47, 65, 64, 63], and the generalized bracket approach [10, 11, 33, 34, 9].

Circular flow in the annular gap between eccentric cylinders is commonly used as a benchmark problem for testing numerical methods and fluid models. Unlike other benchmark problems such as the flow through a contraction or the flow around a sphere in a tube, this problem has a simple flow geometry whose boundaries are smooth and contain no singularities. However, this flow is still complex in that it involves simple shear, planar extension, and rigid body
rotation. Another advantage of using the eccentric cylinder system as a benchmark flow geometry is that exact analytical solutions exist for many fluid models in the limit of a vanishing eccentricity ratio, and thus, they can be used to verify the consistency of numerical codes. In addition, the eccentric cylinder system has a capability for highly distributive and dispersive mixing, and therefore, it is of interest in the industrial processing of complex fluids.

Over the last three decades, many studies have focused on simulating the Taylor-Couette flow of complex fluids between eccentric cylinders. Beris et al. [13] were the first to report successful calculations of the upper-convected Maxwell (UCM) model at large Deborah numbers. The Deborah number is a dimensionless number that is defined as the ratio of the characteristic time scale of the fluid material to that of the flow process. A spectral/finite difference method was used for the spatial discretization of the benchmark problem. The resulting nonlinear system of spatially discretized algebraic flow equations was solved using a specialized Newton code that employs an out-of-core direct frontal method as a linear solver. Thus far, their data remains the most accurate and extensive one reported, and therefore, it is frequently used to validate newly developed numerical codes. Huang et al. [50] solved the UCM model using an unstructured control volume (UCV) method for spatial discretization and the well-known semi-implicit method for the pressure-linked equations revised (SIMPLER) algorithm as a time-stepper. To attain convergence at large Deborah numbers, the UCV method was combined with a self-consistent false diffusion technique. Their solution could be obtained by performing a simulation on currently available computers, and additionally, it further extends the range of achievable Deborah numbers. Unlike Beris et al. [13], they could not, however, resolve the stress boundary layers that are predicted by the UCM model at Deborah numbers larger than unity.

In addition to the UCM model, various other viscoelastic fluid models have been solved. For instance, Baloch et al. [3] solved the linear and exponential versions of the Phan-Thien-Tanner (PTT) model. They studied the impact of the two versions of the model on the flow kinematics, stress fields, local rate of work, and power consumption. Davies and Li [26] solved a White-Metzner model to investigate the effects of temperature and pressure. Singh and Leal [74] solved the Chilcott-Rallison version of the finitely extensible nonlinear elastic (FENE-CR) dumbbell model to examine the influence of polymer concentration on polymer conformation and flow kinematics.

During the last few decades, several studies have focused on the transient dynamics of a single droplet moving in a fully developed eccentric annular flow field. In all these studies, the immiscible fluid components exhibited Newtonian behaviors within the range of the investigated deformation rates. Boonen and coworkers [61, 18, 20] visualized the deformation and orientation of the droplet using an optical microscope with a charge-coupled device (CCD) camera. Their experimental data agreed well with the numerical predictions of the MM model under subcritical flow conditions, i.e., under conditions in which droplet breakup cannot occur. Higher-viscosity droplets deformed less and oriented themselves slightly more toward the flow direction. The selection of a corotating cylinder configuration enhanced the relative importance of the extensional flow to such an extent that it significantly affected the droplet characteristics. In a purely experimental work, the same authors [19] found that the global dynamics of a droplet breakup were governed by a simple shear even when elongational contributions to the mixed flow field were enhanced. Feigl et al. [41] conducted similar experiments at larger length scales. To perform numerical simulations, they developed a new three-step numerical procedure: they employed finite-element and particle-tracking techniques to determine the history of the shear and elongation rates along a particle path, and from this history, they employed boundary integral techniques to calculate the deformation that a droplet undergoes along this path. These authors observed that the local residence time of the droplet plays an important role in determining the location of its breakup. However, to our knowledge, there have been no studies thus far on the dynamics of multiple droplets in a viscoelastic medium in an eccentric Taylor-Couette flow field.

1.1. Organization of Dissertation

The objective of this dissertation was to gain a more fundamental understanding of the rheology-microstructure relationships of polymeric fluids in a steady circular flow in the annular gap between eccentric cylinders. To achieve this objective, we solved the benchmark problem for coarse-grained polymeric fluid models developed by other researchers in the past using the generalized bracket approach.

The rest of this dissertation is organized as follows. In Chap. 2, we first provide a general background of how complex fluids having a microstructure whose dynamics is physically restricted or otherwise can be modeled using the generalized bracket approach. As an example of a model that describes the dynamics of a complex fluid having a constrained microstructure, we consider the extended Dressler-Edwards (EDE) model [30]. This model describes the dynamics of an immiscible blend of a polymeric matrix phase and a Newtonian droplet phase. The droplets are assumed to have a narrow size distribution. Furthermore, they are allowed to break up and coalesce during flow. Note that the power-law extended White-Metzner (EWM) model [75] is a limiting case of the EDE model. It is obtained by considering only the dynamics of the matrix fluid and assuming a vanishing total volume fraction of the droplet phase. This model describes the unconstrained dynamics of polymeric solutions and melts.

Chapter 3 is dedicated to a steady eccentric Taylor-Couette flow of polymeric solutions and melts. First, we formulate the benchmark problem for the power-law EWM model. Then, we introduce a numerical approach developed for the benchmark simulations. This numerical approach is unique in that the nonlinear system of discretized algebraic flow equations is solved iteratively using a Newton-Krylov method along with an inverse-based incomplete
lower-upper (ILU) preconditioner. We show that the iterative solution technique is capable of solving the benchmark problem for the UCM model at the largest Deborah number that has been reported by Beris et al. [13] for a moderate eccentricity ratio of the cylinder system. Then, we report sample calculations and discuss interesting phenomena that are caused by the highly eccentric cylinder system and the viscoelastic behavior of the polymeric liquids. Finally, we draw some conclusions.

Chapter 4 describes steady eccentric Taylor-Couette flow of immiscible polymeric blends with droplet morphology. First, we formulate the benchmark problem for the EDE model. Then, we present necessary adaptations of the numerical approach for performing the simulations. Next, we report sample calculations and discuss interesting phenomena that are related to the large eccentricity ratio of the cylinder system and the viscoelastic nature of the blend. Special attention is paid to determining how the droplet characteristics can be tailored by modifying the rheology of the matrix fluid. Finally, we make some conclusions.

Chapter 5 summarizes the main results of this dissertation and provides an overview of future research.

**2. Thermodynamic Modeling**

This chapter illustrates how the dynamics of complex fluids can be modeled using the generalized bracket approach. In Sec. 2.1, we provide a brief introduction to bracket formalism and show its close interconnection with GENERIC formalism. In Sec. 2.2, we describe the derivation of a general set of flow equations for complex fluids having an unconstrained microstructure. In Sec. 2.3, we explain how the general procedure must be adopted in the case that the microstructural dynamics is physically restricted. To illustrate the modeling of the dynamics of complex fluids having a constrained microstructure, we introduce the EDE model in Sec. 2.4.

**2.1. Theoretical Framework**

Let us consider a complex fluid being confined to an isolated system of volume $\Omega$. Allowing the flow to be nonisothermal and compressible, the state variables of the system are typically the scalar mass density $\rho$; the momentum density vector $M = \rho v$, with $v$ being the velocity vector; the internal energy density $u$; and the conformation density tensor $C = \rho \varepsilon$. The symmetric second-order tensor $\varepsilon$ is referred to as the conformation tensor; it describes the microstructure of the complex fluid. The eigenvalues of this tensor are a measure of its deformation and extension, and its eigenvectors are a measure of its orientation. Let the set of state variables of the system be collectively represented by the vector $x = [\rho, M, u, C]$. According to GENERIC formalism [47, 65, 64, 63], the time evolution equation for the set of state variables is written as follows:

$$
\frac{\partial x}{\partial t} = P(x) \cdot \frac{\delta E(x)}{\delta x} + Q(x) \cdot \frac{\delta S(x)}{\delta x},
$$

where $E$ and $S$ are generating functionals representing the global energy and entropy of the system, respectively. Furthermore, $P$ and $Q$ are operator matrices that dictate the conservative and dissipative contributions of the system dynamics, respectively. Each of these operator matrices must satisfy certain properties: the operator matrix $P$ must be antisymmetric and must satisfy the Jacobi identity. The operator matrix $Q$ must be symmetric and positive semi-definite. To obtain a thermodynamically consistent description, some degeneracy
2. Thermodynamic Modeling

requirements must be met, i.e.,

\[ P(x) \frac{\partial S(x)}{\partial x} = 0, \quad (2.2) \]
\[ Q(x) \frac{\delta E(x)}{\delta x} = 0. \quad (2.3) \]

Condition (2.2) expresses the fact that the functional form of the global entropy is such that it cannot be affected by the operator generating reversible dynamics. Condition (2.3) expresses the necessity that the total energy of the system must be conserved by the dissipative contributions. To obtain a specific model, the generating functionals \( E \) and \( S \) and the operator matrices \( P \) and \( Q \) must be specified.

Many complex fluids have a microstructure whose dynamics is physically restricted. An example of such a complex fluid is a liquid of rod-like polymers whose extension does not change by application of an external force. It should be mentioned that it is not straightforward to derive a general set of flow equations for complex fluids having a constrained microstructure using the GENERIC formalism. The bracket approach is much better suited for this purpose. According to the two-generator bracket formalism [47, 65, 64, 63, 32, 37, 17, 8], the time evolution equation for the state variables of the system can be equivalently expressed as

\[ \frac{dF}{dt} = [F, E] + [F, S], \quad (2.4) \]

where \( F \) is an arbitrary functional of the state variables of the system. The reversible and irreversible contributions to the system dynamics are represented by the Poisson bracket \([\cdot, \cdot]\), and the dissipation bracket \([\cdot, \cdot]\), respectively. Equations (2.1) and (2.4) are related to each other as

\[ \frac{dF}{dt} = \int_{\Omega} \frac{\delta F}{\delta x} \frac{\partial}{\partial t} \delta x \delta r = \int_{\Omega} \frac{\delta F}{\delta x} P \frac{\partial E}{\partial x} \delta r + \int_{\Omega} \frac{\delta F}{\delta x} Q \frac{\partial S}{\partial x} \delta r. \quad (2.5) \]

A comparison between (2.1) and (2.5) shows that the Poisson and dissipation brackets can be determined by defining the operator matrices \( P \) and \( Q \). In the presence of local thermal equilibrium, the total entropy of the system, \( S \), can be expressed in terms of the total energy of the system, \( E \). Then, the two-generator bracket formalism collapses to one-generator bracket formalism [10, 11, 33, 9]

\[ \frac{dF}{dt} = [F, H] = [F, H] + [F, H]. \quad (2.6) \]

Because \([\cdot, \cdot]\) is referred to as a generalized bracket, the one-generator bracket formalism is frequently referred to as generalized bracket formalism. In this formalism, the system dynamics is generated by the Hamiltonian of the system, \( H \). As a consequence, the complete set of state variables of the system is now represented by the vector \( x = [p, M_r, s, C_\alpha]^T \), where the scalar \( s \) denotes the entropy density. Like the operator matrices, each of these brackets must satisfy certain properties. For example, the Poisson bracket must be bilinear and anti-symmetric with respect to \( F \) and \( H \) and it must satisfy the Jacobi identity. By definition, we have \([H, H] = 0\). Moreover, the Poisson bracket should be constructed in a manner such that the global entropy of the isolated system does not change by reversible processes, i.e.,

\[ [S, H] = 0. \quad (2.7) \]

The dissipation bracket must be linear in \( F \), but it may be nonlinear in \( H \). To obtain a thermodynamically consistent description, it must meet the following requirement:

\[ [H, H] = 0, \quad (2.8) \]
\[ [S, H] \geq 0. \quad (2.9) \]

The condition (2.8) guarantees that the total energy of the system remains constant in time, whereas the condition (2.9) expresses the fact that any associated entropy production must be non-negative. The total mass of an isolated system must also be constant. To obtain a specific model, the master equation (2.6) together with the Hamiltonian of the system, \( H \), must be specified.

2.2. Complex Fluids with Unconstrained Microstructure

In this section, we describe the procedure for deriving a general set of flow equations for complex fluids having an unconstrained microstructure. Henceforth, we assume that the configuration density tensor deforms as a symmetric contravariant second-order tensor.

2.2.1. Functional Calculus

To understand the derivation of the general set of flow equations, it is necessary to remember some aspects of functional calculus.

Let us consider an arbitrary functional \( F \) that depends on the state variables of the system, \( a, b, \ldots \in \Omega \) (\( \Omega \) being the operating space of the problem under consideration), as

\[ F(a, b, \ldots) \equiv \int_{\Omega} f(a, \nabla a, b, \nabla b, \ldots) \delta x \delta r, \quad (2.10) \]

where \( \Omega \) is the domain of interest with a fixed boundary \( \partial \Omega \).
Since \( a, b, \ldots \in \mathcal{P} \) are functions of time \( t \) and space \( \mathbf{r} \), the total time derivative of \( F \) is written as
\[
\frac{\partial F}{\partial t} = \sum_i \left( \frac{\partial F}{\partial a_i} \frac{\partial a_i}{\partial t} + \frac{\partial F}{\partial \mathbf{b}} \frac{\partial \mathbf{b}}{\partial t} \right) + \ldots
\]
In order that the above inner products be proper ones, their subjects must belong to the same operating space. The Volterra derivative of \( F \) with respect to a state variable of the system of arbitrary tensorial order, \( a \), is properly defined as follows:
\[
\frac{\partial F}{\partial a} = \Pi^\varepsilon \left( \frac{\partial f}{\partial a} - \mathbf{\nabla} \cdot \frac{\partial f}{\partial (\mathbf{V} \mathbf{a})} \right) \in \mathcal{P},
\]
where \( \Pi^\varepsilon \) is a projection operator that ensures that \( \partial F/\partial a \) and \( \partial a_i/\partial t \) belong to \( \mathcal{P} \). For an unconstrained variable \( a \), the projection operator \( \Pi^\varepsilon \) is simply the identity operator. Note that the above functional derivative \( \partial F/\partial a \) is just a notation representing the Euler-Lagrange equation obtained by varying \( F \) with respect to \( a \). The definition of \( \partial F/\partial a \) can be easily extended to the case where \( F \) depends implicitly on higher-order spatial derivatives of \( a \).

### 2.2. Complex Fluids with Unconstrained Microstructure

The next step of the derivation procedure is to specify the master equation (2.6) together with the Hamiltonian of the system, \( H \). On the basis of the chain rule of differentiation, the total time derivative of the functional \( F \) appearing on the left-hand side of (2.6) can be expressed as
\[
\frac{\partial F}{\partial t} = \int_\Omega \left( \frac{\partial F}{\partial \Phi} \frac{\partial \Phi}{\partial t} + \frac{\partial F}{\partial \mathbf{M}_a} \frac{\partial \mathbf{M}_a}{\partial t} \right) d^3 r.
\]

The Poisson bracket appearing on the right-hand side of (2.6) are described in Sec. 2.2.3 and Sec. 2.2.4, respectively. A general expression for the Hamiltonian of the system, \( H \), is provided in Sec. 2.2.5.

#### 2.2.3. Poisson bracket

The Poisson bracket in the spatial description of fluid flow can be derived from the corresponding bracket in the material description of fluid flow. The latter Poisson bracket is a continuum analog of the Poisson bracket of the discrete particle system [43], i.e.,
\[
\{F, G\} = \int_\Omega \left( \frac{\partial F}{\partial \Phi} \frac{\partial G}{\partial \Phi} - \frac{\partial F}{\partial \mathbf{M}_a} \frac{\partial G}{\partial \mathbf{M}_a} \right) d^3 r,
\]
where \( \Phi \) and \( \Pi \) denote the material coordinate and the conjugate momentum vector field, respectively. To obtain the Poisson bracket in the spatial description of fluid flow, the Volterra derivatives of the arbitrary functions \( F \) and \( G \) with respect to the material variables \( \Phi \) and \( \Pi \) must be expressed in terms of those with respect to the spatial variables \( \mathbf{r} \). The antisymmetry property of the Poisson bracket into account, it can be expressed as follows in the material description of fluid flow [10, 11, 33, 34, 9]:
\[
\{F, G\} = -\int_\Omega \left[ \int_\Omega \left( \frac{\partial F}{\partial \Phi} \frac{\partial G}{\partial \Phi} - \frac{\partial F}{\partial \mathbf{M}_a} \frac{\partial G}{\partial \mathbf{M}_a} \right) d^3 r \right. - \left. \int_\Omega \left( \frac{\partial F}{\partial \Phi} \frac{\partial G}{\partial \Phi} - \frac{\partial F}{\partial \mathbf{M}_a} \frac{\partial G}{\partial \mathbf{M}_a} \right) d^3 r \right].
\]
The first three integrals of the Poisson bracket (2.16) represent the reversible contributions of the fluid medium to the system dynamics. They lead to hydrodynamic flow equations for a conservative ideal fluid. The remaining integrals represent the reversible contributions of the microstructure of the fluid to the system dynamics. It should be pointed out that these integrals are only valid for a contravariant second-order conformation density tensor. They lead to an upper-convected derivative in the time evolution equation of the conformation density tensor. The lower-convected and corotational derivatives can be obtained by assuming that the conformation density tensor deforms as a covariant or mixed second-order tensor, respectively.

### 2.2.4. Dissipation Bracket

The most general possible expression for the dissipation bracket that is consistent with the first and second laws of thermodynamics is given as [33, 34, 9]

\[
[F, G] = - \int \left\{ \left( \frac{\delta F}{\delta x} \cdot \nabla \frac{\delta G}{\delta x} \right) \frac{\delta G}{\delta W} \nabla \frac{\delta G}{\delta W} + \frac{1}{T} \frac{\delta F}{\delta S} \left( \frac{\delta G}{\delta x} \cdot \nabla \frac{\delta G}{\delta x} \right) \frac{\delta G}{\delta W} \nabla \frac{\delta G}{\delta W} \right\} d^3r ,
\]

(2.17)

where \( T \) denotes the absolute temperature and \( L (\cdot) \) indicates that the dependence of \( \Xi \) is linear with respect to \( \cdot \). Note that the linear dependence on \( \delta F/\delta x \) has been written separately in the above expression. The state variables of the system without entropy density are collectively represented by the vector \( \mathbf{w} = [\rho, M_\alpha, C_{\alpha}]^T \). The quantities \( \delta G/\delta x \) and \( \nabla \delta G/\delta x \) represent system affinities that are associated with volumetric (relaxational) and surface (transport) fluxes of the state variables of the system, respectively. Their determination requires the consideration of the conservation of mass, the principles of frame indifference and material invariance, and the microscopic time reversibility of the system.

By definition, \( \Xi \) must be linear with respect to \( F \). However, in general, it may be nonlinear with respect to \( G \). For systems close to thermal equilibrium, \( \Xi \) is linear with respect to both \( F \) and \( G \). Then, the dissipation bracket becomes [33, 9]

\[
[F, G] = - \int \left\{ \frac{\delta G}{\delta M_\alpha} \frac{\delta F}{\delta M_\alpha} \nabla_\alpha \frac{\delta G}{\delta M_\alpha} + \frac{1}{T} \frac{\delta F}{\delta C_{\alpha} \quad \delta C_{\alpha}} \nabla_\alpha \frac{\delta G}{\delta C_{\alpha} \quad \delta C_{\alpha}} \frac{\delta G}{\delta W} \nabla \frac{\delta G}{\delta W} \right\} d^3r ,
\]

(2.18)

The above integrals with the phenomenological tensors \( Q, B, R, A, P \) represent the effects of viscous dissipation, heat conduction, molecular diffusion, molecular relaxation, and non-affine motion, respectively. Note that these phenomenological tensors must satisfy symmetry conditions of the form \( Q_{\alpha \beta \gamma} = Q_{\beta \gamma \alpha} = Q_{\gamma \alpha \beta} \).

### 2.2.5. Hamiltonian

The Hamiltonian of a system quantifies the total system energy. It is obtained through a Legendre transformation of the corresponding Lagrangian and can be written as [43]

\[
H (\rho, M, x, C) = \int \left\{ \frac{M_\alpha M_\alpha}{2} p_{\alpha \beta} + q_\alpha + \alpha (\rho, x, C) \right\} d^3r ,
\]

(2.19)

where \( r_\alpha \) denotes an external field potential (for instance, gravitational, electric, or magnetic fields). Evaluating the Volterra derivative of \( H \) with respect to the state variables yields

\[
\frac{\delta H}{\delta p} = - \frac{M_\alpha M_\alpha}{2} p_{\alpha \beta} + q_\alpha + \frac{\partial \alpha}{\partial p} ,
\]

(2.20)

\[
\frac{\delta H}{\delta M_\alpha} = - \frac{M_\alpha}{p} v_\alpha ,
\]

(2.21)

\[
\frac{\delta H}{\delta x} = T ,
\]

(2.22)

\[
\frac{\delta H}{\delta C_{\alpha}} = \frac{\partial \alpha}{\partial C_{\alpha}} ,
\]

(2.23)

### 2.2.6. Flow Equations

We proceed to derive a general set of flow equations for complex fluids having an unconstrained microstructure. The master equation (2.6) has been fully specified: the total time
derivative of the functional $F$ appearing on its left-hand side has been expressed in terms of the state variables of the system in (2.11). The Poisson and dissipation brackets appearing on its right-hand side have been specified in (2.16) and (2.18), respectively. Comparison of the left- and right-hand sides gives a general set of time evolution equations for the state variables of the system. Using the general expression for the Hamiltonian of the system, $H$, provided in (2.19), we obtain

\[
\frac{\partial \rho}{\partial t} = - \nabla_s (\psi_s \rho), \quad (2.24)
\]

\[
\frac{\partial \psi_s}{\partial t} = - \rho \nabla_s \psi_s - \rho \nabla_s \psi - \nabla_s \rho + \nabla_s \rho^T, \quad (2.25)
\]

\[
\frac{\partial s}{\partial t} = - \nabla_s (\psi_s s) + \frac{1}{T} \nabla_s \left( B_{\psi_s} T V_s \psi_s \right) + \frac{1}{T} Q_{\psi_s} \nabla_s V_s V_s + \frac{1}{T} \Lambda_{\psi_s} \frac{\partial \psi_s}{\partial \psi_s}, \quad (2.26)
\]

\[
\frac{\partial \psi_{C1}}{\partial t} = - \nabla_s \left( \psi_s C_{\psi_s} \right) + C_{\psi_s} V_s V_s + C_{\psi_s} \psi_s V_s + P_{\psi_s} \psi_s V_s - \Lambda_{\psi_s} \frac{\partial \psi_s}{\partial \psi_s} + \nabla_s \left( R_{\psi_s} \psi_s \right). \quad (2.27)
\]

Equation (2.24) is the mass balance equation. Equation (2.25) is the Cauchy momentum balance equation. Equation (2.26) is the entropy balance equation. Equation (2.27) describes the dynamics of the microstructural constituents of the fluid. Together with the partial time derivative on the left-hand side of (2.27), the first three terms on its right-hand side constitute the upper-convected time derivative. These four terms describe how the microstructural components of the fluid evolve with velocity. To close the general set of flow equations, an explicit expression for the stress tensor can be determined through the Poisson and dissipation brackets: it is given as

\[
\sigma_{\alpha \beta} = 2 C_{\rho \psi_s} \frac{\partial \psi_s}{\partial \psi_s} + Q_{\psi_s} \nabla_s \psi_s + P_{\psi_s} \frac{\partial \psi_s}{\partial \psi_s}. \quad (2.28)
\]

A specific model can be obtained by specifying the phenomenological tensors $Q, B, R, A$, and $P$ and the internal energy density $u$. A complex fluid model to be thermodynamically admissible, condition (2.9) must be satisfied, i.e.,

\[
\nabla_s \left( B_{\psi_s} T V_s \psi_s \right) + Q_{\psi_s} \nabla_s V_s \psi_s + \frac{1}{T} \Lambda_{\psi_s} \frac{\partial \psi_s}{\partial \psi_s} + \frac{1}{T} \Lambda_{\psi_s} \frac{\partial \psi_s}{\partial \psi_s} \nabla \psi_s \geq 0. \quad (2.29)
\]

For polymeric fluids, it is standard practice to express the system Hamiltonian $H$ in terms of the Helmholtz free energy density $a$, rather than in terms of the internal energy density $u$. These two physical quantities are related to each other as follows:

\[
u = a + T \rho. \quad (2.30)
\]

To obtain a general set of flow equations for polymeric fluids, the partial derivative $\partial u/\partial C$ appearing in the general set of flow equations is commonly expressed in terms of the partial derivative $\partial a/\partial C$. Applying the chain rule of differentiation, the partial derivative $\partial u/\partial C$ can be expressed as

\[
\frac{\partial u}{\partial C_{\psi_s}} \bigg|_{\rho=M_T} = \frac{\partial a}{\partial C_{\psi_s}} \bigg|_{\rho=M_T} + \frac{\partial a}{\partial C_{\psi_s}} \bigg|_{\rho=M_T} \frac{\partial T}{\partial C_{\psi_s}}. \quad (2.31)
\]

However, differentiating the partial derivative $\partial u/\partial C$ by parts gives

\[
\frac{\partial u}{\partial C_{\psi_s}} \bigg|_{\rho=M_T} = \frac{\partial a}{\partial C_{\psi_s}} \bigg|_{\rho=M_T} + \frac{\partial a}{\partial C_{\psi_s}} \bigg|_{\rho=M_T} \frac{\partial T}{\partial C_{\psi_s}}. \quad (2.32)
\]

If we substitute (2.32) back in (2.31), we obtain

\[
\frac{\partial u}{\partial C_{\psi_s}} \bigg|_{\rho=M_T} = \frac{\partial a}{\partial C_{\psi_s}} \bigg|_{\rho=M_T}. \quad (2.33)
\]

Several polymeric fluid models can be expressed in the generalized bracket formalism [10, 33, 9]. An example of such a model is the UCM model. This simple phenomenological model originates by considering a Hookean spring, which is associated with the elasticity of the polymeric constituents, connected in series to a dashpot, which is associated with viscous damping. The corresponding set of nonisothermal and compressible flow equations can be obtained by using the following expression for the Helmholtz free energy density:

\[
\mathcal{A} = \frac{\nu K}{2} \ln \mathbf{C} - \frac{\nu K}{2} \ln \det (\mathbf{C}/p) + \mathcal{A} (p, s), \quad (2.34)
\]

where $\nu$ is the number of polymeric constituents per unit mass and $K$ is the elastic (Hookean) constant. The first term on the right-hand side of (2.34) is the potential energy density that is associated with the elasticity of the polymeric constituents of the fluid. The second term represents the entropic contribution associated with the distribution of the polymeric constituents of the fluid. Finally, the third term represents the Helmholtz free energy density in the absence of elastic effects. Furthermore, the components of the phenomenological tensor $\Lambda$ are given as

\[
\Lambda_{\psi_s} = \frac{1}{2 K \lambda} \left( C_{\psi_s \psi_s} + C_{\psi_s \psi_s} + C_{\psi_s \psi_s} + C_{\psi_s \psi_s} \right). \quad (2.35)
\]

where $\lambda$ is the characteristic relaxation time of the polymeric components of the fluid. The components of the other phenomenological tensors $Q, B, R$, and $P$ are all identically zero.
2. Thermodynamic Modeling

2.3. Complex Fluids with Constrained Microstructure

In the preceding section, we described the derivation of a general set of flow equations for complex fluids having an unconstrained microstructure. In this section, we extend this procedure to the derivation of a general set of flow equations for complex fluids having a constrained microstructure. The approach is discussed only for the case of a volume-preserving microstructure. This constraint is not difficult to understand. Therefore, the approach is considered only for the case of a volume-preserving microstructure. This will be considered again while presenting the theory of the EDE model in Sec. 2.4.

2.3.1. Operating Space

In an isolated system, the flow is confined to a fixed domain of volume \( \Omega \) and surface boundary \( \partial \Omega \). The volume of the microstructural constituents of the fluid is assumed to remain constant during flow. To describe their dynamics, we adopt a unit-free symmetric second-order contravariant tensor \( K \) satisfying the mathematical constraint \( f^i = 1 \), where \( f^i = \det K \) is its third invariant. The constrained tensor \( K \) is obtained from the unconstrained tensor \( C \) by applying the following mapping:

\[
C \rightarrow K = \frac{C}{\sqrt{f^i}}. \tag{2.36}
\]

The operating space is now defined as follows:

\[
p = \left\{ \begin{array}{ll}
\rho(r, t) \in \mathbb{R}^3; & \rho(r, 0) = \rho_0(r) \text{ in } \Omega, \\
M(r, t) \in \mathbb{R}^3; & M(r, 0) = M_0(r) \text{ in } \Omega, \quad n \cdot M = 0 \text{ on } \partial \Omega, \\
x(r, t) \in \mathbb{R}^3; & x(r, 0) = x_0(r) \text{ in } \Omega, \\
\mathbf{K}(r, t) \in \mathbb{R}^{3\times3}; & \mathbf{K}(r, 0) = \mathbf{K}_0 \text{ in } \Omega.
\end{array} \right. \tag{2.37}
\]

Henceforth, we restrict the arbitrary functionals \( F \) and \( G \) to those that depend on \( \mathbf{K} \) (and \( \nabla \mathbf{K} \)) only through their dependence on \( C \) (and \( \nabla C \)): \( F[\rho, M, x, \mathbf{K}] \) and \( G[\rho, M, x, \mathbf{K}] \). Applying the chain rule of differentiation, the total time derivative of \( F \) can be written as

\[
\frac{dF}{dt} = \int_{\Omega} \left( \frac{\partial F}{\partial \rho} \frac{\partial \rho}{\partial t} + \frac{\partial F}{\partial M} \frac{\partial M}{\partial t} + \frac{\partial F}{\partial x} \frac{\partial x}{\partial t} + \frac{\partial F}{\partial \mathbf{K}} \frac{\partial \mathbf{K}}{\partial t} \right) \, d^3r.
\]

Next, we transform the Poisson bracket (2.16) to an equivalent one that can describe the reversible dynamics of a complex fluid having a volume-preserving microstructure. If we consider the mathematical structure of the Poisson bracket (2.16), we observe that it is linear in \( \delta F / \delta C \) and \( \delta G / \delta C \). Therefore, it is necessary to transform both these Volterra derivatives according to the transformation rule (2.36). Applying the chain rule of differentiation, they become

\[
\delta \cdot \frac{\partial F}{\partial C} = \nabla_r \cdot \left( \frac{\partial F}{\partial C} \frac{\partial C}{\partial K} \mathbf{K} \right) = \nabla_r \cdot \left( \frac{\partial F}{\partial C} \frac{\partial C}{\partial \mathbf{K}} \mathbf{K} \right).
\]

Accordingly, the Poisson bracket (2.16) becomes

\[
\{ F, G \} = \int_{\Omega} \left( \delta F \left( \frac{\partial G}{\partial M} - \frac{\partial G}{\partial M} \frac{\partial M}{\partial \mathbf{K}} \mathbf{K} \right) \delta G \left( \frac{\partial F}{\partial M} - \frac{\partial F}{\partial M} \frac{\partial M}{\partial \mathbf{K}} \mathbf{K} \right) \right) \, d^3r.
\]
2. Thermodynamic Modeling

2.3. Complex Fluids with Constrained Microstructure

2.3.3. Dissipation Bracket

Next, we transform the dissipation bracket (2.18) to an equivalent one that can describe the reversible dynamics of a complex fluid having a volume-preserving microstructure. To transform it properly, we must consider its mathematical structure. The integrals with the phenomenological tensors \( Q \) and \( B \) remain unchanged since they do not contain any terms involving the state variable \( C \). The integrals with the phenomenological tensors \( R \) and \( \Lambda \) are bilinear in \( \delta F / \delta C \) and \( \delta G / \delta C \). To obtain a proper expression for the dissipation bracket that takes into account the volume preservation constraint, it is sufficient to transform only one of these two Volterra derivatives according to the transformation rule (2.36). Since the integral with the phenomenological tensor \( P \) is linear in \( \delta F / \delta C \) and \( \delta G / \delta C \), it is necessary to transform both the Volterra derivatives according to the transformation rule (2.36). In doing so, the dissipation bracket (2.18) becomes

\[
[F,G] = - \int Q_{xy} \left( \frac{\delta F}{\delta M} - \frac{\delta G}{\delta M} \right) \frac{\delta F}{\delta M_b} \frac{\delta M}{\delta M_b} \, d^3r
- \int B_{xy} \left( \frac{\delta F}{\delta S} - \frac{\delta G}{\delta S} \right) \frac{\delta F}{\delta S_b} \frac{\delta S}{\delta S_b} \, d^3r
- \int R_{x} \left( \frac{\delta F}{\delta K_{\alpha\beta\gamma}} - \frac{\delta G}{\delta K_{\alpha\beta\gamma}} \right) \frac{\delta F}{\delta K_{\alpha\beta\gamma}} \frac{\delta K_{\alpha\beta\gamma}}{\partial K_{\alpha\beta\gamma}} \, d^3r
+ \left( \frac{1}{3} \right) \int R_{xy} \left( \frac{\delta F}{\delta K_{\alpha\beta\gamma}} - \frac{\delta G}{\delta K_{\alpha\beta\gamma}} \right) \frac{\delta F}{\delta K_{\alpha\beta\gamma}} \frac{\delta K_{\alpha\beta\gamma}}{\partial K_{\alpha\beta\gamma}} \, d^3r
- \int \Lambda_{xy} \left( \frac{\delta F}{\delta K_{\alpha\beta\gamma}} - \frac{\delta G}{\delta K_{\alpha\beta\gamma}} \right) \frac{\delta F}{\delta K_{\alpha\beta\gamma}} \frac{\delta K_{\alpha\beta\gamma}}{\partial K_{\alpha\beta\gamma}} \, d^3r
+ \left( \frac{1}{3} \right) \int \Lambda_{x} \left( \frac{\delta F}{\delta K_{\alpha\beta\gamma}} - \frac{\delta G}{\delta K_{\alpha\beta\gamma}} \right) \frac{\delta F}{\delta K_{\alpha\beta\gamma}} \frac{\delta K_{\alpha\beta\gamma}}{\partial K_{\alpha\beta\gamma}} \, d^3r.
\]

(2.43)

2.3.4. Hamiltonian

Application of the mapping (2.36) to the internal energy density in the general expression of the Hamiltonian given in (2.19) yields

\[
H(p,M,s,K) = \int Q \left( \frac{M}{2p} + p \mathbf{v} + \alpha(p,s,K) \right) \, d^3r.
\]

(2.45)

2.3.5. Flow Equations

We proceed to derive a general set of flow equations for complex fluids having a volume-constrained microstructure. The master equation (2.6) has been fully specified: the total time derivative of the functional \( F \) appearing on the left-hand side has been expressed in terms of the state variables in (2.38). The Poisson and dissipation brackets appearing on its right-hand side have been specified in (2.43) and (2.44), respectively. Comparison of the two sides of (2.6) gives a general set of time evolution equations for the state variables of the system. Using the general expression for the system Hamiltonian \( H \) provided in (2.45), we obtain

\[
\frac{\partial \phi}{\partial t} = - \nabla_{\mathbf{v}} \left( \phi \mathbf{v} \right),
\]

(2.46)

\[
\frac{\partial \mathbf{v}}{\partial t} = - \mathbf{v} \nabla \cdot \mathbf{v} - \rho \nabla \mathbf{p} + \nabla \mathbf{P} - \nabla \mathbf{F}.
\]

(2.47)
2. Thermodynamic Modeling

\[
\frac{\partial K_{ij}}{\partial t} = -\nabla_i \left( v_i K_{ij} \right) + K_{ij} \nabla_i v_j + K_{ij} \nabla_i v_j - \frac{2}{3} K_{ii} \nabla_j v_j + P_{ij} \nabla_i v_j - \frac{1}{3} P_{iklj} K_{kl} \nabla_i v_j - \frac{1}{3} \nabla_j K_{ij} \nabla_i v_j + \frac{1}{3} \nabla_i \left( K_{ij} \nabla_j \frac{\partial u}{\partial K_{ij}} \right) + \frac{1}{3} \nabla_i \left( K_{ij} \nabla_j \frac{\partial u}{\partial K_{ij}} \right) - \frac{1}{3} \nabla_i \left( K_{ij} \nabla_j \frac{\partial u}{\partial K_{ij}} \right) K_{ij} K_{ii}^{-1}. \tag{2.48}
\]

Equation (2.46) is the mass balance equation. Equation (2.47) is the Cauchy momentum balance equation. Equation (2.48) is the entropy balance equation. Equation (2.49) describes the dynamics of a volume-preserving polymeric microstructure. To close the general set of flow equations, an explicit expression for the extra stress tensor can be determined through the Poisson and dissipation brackets, as

\[
\sigma_{ij} = 2K_{ij} \frac{\partial u}{\partial K_{ij}} - \frac{1}{3} K_{ii} \frac{\partial u}{\partial K_{ij}} - \frac{1}{3} \frac{\partial u}{\partial K_{ij}} + Q_{ij} \nabla_i v_j + P_{ij} \nabla_i v_j - \frac{1}{3} P_{iklj} K_{kl} \nabla_i v_j - \frac{1}{3} \nabla_j K_{ij} \nabla_i v_j + \frac{1}{3} \nabla_i \left( K_{ij} \nabla_j \frac{\partial u}{\partial K_{ij}} \right) + \frac{1}{3} \nabla_i \left( K_{ij} \nabla_j \frac{\partial u}{\partial K_{ij}} \right) - \frac{1}{3} \nabla_i \left( K_{ij} \nabla_j \frac{\partial u}{\partial K_{ij}} \right) K_{ij} K_{ii}^{-1}. \tag{2.50}
\]

We observe that the mass balance equation is the same for a complex fluid with or without an unconstrained microstructure; the same is also true for the Cauchy momentum balance equation. Additional terms arise in the entropy balance equation, the time evolution equation (2.49) describes the deformation of the droplet shape. The Maffettone-Minale (MM) model was also based on the assumption that the two immiscible Newtonian fluids. Since the droplet was restricted to have an ellipsoidal shape at all times, it could be characterized in terms of a symmetric positive-definite second-order tensor with eigenvalues representing its square semiaxes. In order to maintain a constant droplet volume, the third invariant (i.e., the determinant) of the tensor was constrained to some constant. In a subsequent study, Gmünter et al. [48] provided a thermodynamically consistent expression for the extra stress generated by the deformation of the droplet shape. Subsequently, the MM model was extended to the case of non-Newtonian fluid components as well [57, 60].

A specific model can be obtained by specifying the phenomenological tensors \(Q, B, R, A\), and \(P\) and the internal energy density \(u\). For a complex fluid model to be thermodynamically admissible, the condition (2.9) must be met, i.e.,

\[
\nabla_i \left( R_{ij} T \nabla_j v_i + Q_{ij} \nabla_j v_i + R_{ij} \nabla_j v_i + Q_{ij} \nabla_j v_i \right) + \nabla_i \left( R_{ij} \nabla_j v_i \right) + \frac{1}{3} \frac{\partial u}{\partial K_{ij}} \nabla_i \frac{\partial u}{\partial K_{ij}} + \frac{1}{3} \frac{\partial u}{\partial K_{ij}} \nabla_i \frac{\partial u}{\partial K_{ij}} + \frac{1}{3} \frac{\partial u}{\partial K_{ij}} \nabla_i \frac{\partial u}{\partial K_{ij}} \geq 0. \tag{2.51}
\]

2.4. Example

In the first part of this chapter, we provided relevant background that is necessary for understanding how complex fluids can be modeled using the generalized bracket approach. We therefore hope that the theory of the EDE model presented in this section can now be understood more easily. We first review the most important modeling efforts that were made in the past to obtain a continuum description of immiscible blends with droplet morphology.

In the last 80 years, considerable research has been carried out to develop models of immiscible blends with droplet morphology. The first models were purely analytical ones that described the deformation of a single Newtonian droplet in a Newtonian matrix [77, 78, 25, 42, 4, 5, 69]. Choi and Schowalter [24] extended Cox and Acrinos’ work in order to explain the effects of neighboring droplets. Subsequently, Maffettone and Minale [58, 59] developed a phenomenological model that could describe the time evolution of the droplet shape. The Maffettone-Minale (MM) model was also based on the assumption that the two immiscible Newtonian fluids. Since the droplet was restricted to have an ellipsoidal shape at all times, it could be characterized in terms of a symmetric positive-definite second-order tensor with eigenvalues representing its square semiaxes. In order to maintain a constant droplet volume, the third invariant (i.e., the determinant) of the tensor was constrained to some constant. In a subsequent study, Gmünter et al. [48] provided a thermodynamically consistent expression for the extra stress generated by the deformation of the droplet shape. Subsequently, the MM model was extended to the case of non-Newtonian fluid components as well [57, 60].

An important step toward the development of a continuum description of multiple droplets immersed in a matrix fluid was taken by Doi and Ohta [28]. The Doi-Ohta (DO) model considered a blend of two immiscible Newtonian fluids of equal density and concentration. The dynamics of the interface formed between the blend components was characterized in terms of a specific interfacial area and a specific anisotropy tensor that had been previously developed by Batchelor [6]. In addition to the droplet deformation, this model also considered droplet breakups and coalescences. However, because the model does not have any intrinsic time scales, it exhibits certain unusual scaling behaviors; for example, the model predicts that the first normal stress difference depends linearly (and not quadratically) on the shear rate in the case of a steady simple shear flow. Several modifications have been made to improve the model predictions both rigorously [46, 54, 79] and in a somewhat ad hoc manner [55]. Inspired by the two abovementioned different phenomenological approaches for describing the droplet morphology, i.e., the DO and MM theories, Gmünter et al. [48] developed flow equations to describe immiscible blends of two viscoelastic fluids. These researchers adopted the DO theory to describe the dynamics of the interface formed between the blend components and the MM theory to describe the dynamics of their polymeric constituents.
Unlike the MM model, the DO model does not require the average droplet volume to be conserved. Almusallam et al. [1] proposed a phenomenological model for the blends of two immiscible Newtonian fluids having equal density. In this model, a new droplet relaxation mechanism that approximately conserves the average droplet volume is combined with the affine deformation of interfaces developed by Doi and Ohta [28] and improved by Wetzel and Tucker [80]. However, it is quite difficult to constrain the average droplet volume such that it remains constant under flow, and such a constraining was fully achieved only two years later [35]. By using the generalized bracket approach, Dressler and Edwards [29] derived flow equations for immiscible polymer blends with droplet morphology. Further, in order to simplify the model equations as much as possible, the droplet phase was restricted to be Newtonian. Dressler and Edwards [30] made another important contribution by extending their model to the case wherein the droplets are allowed to breakup and coalesce during flow. The flow is supposed to be isothermal and incompressible. The size distribution of the droplets is assumed to be narrow. Moreover, they are allowed to break up and coalesce during flow. Using the same thermodynamic framework, other researchers also considered the case of a viscoelastic droplet phase [81, 40]. However, their models are unable to predict the breakup and coalescence phenomena, which occur naturally at larger deformations.

### 2.4.1. Operating Space

Let us start with the theoretical derivation of the flow equations. The EDE model describes the dynamics of immiscible blends of a polymeric matrix phase and a Newtonian droplet phase. The size distribution of the droplets is assumed to be narrow. Moreover, they are allowed to break up and coalesce during flow. The flow is supposed to be isothermal and incompressible. Under these flow conditions, the scalar mass density \( \rho \) and the entropy density \( s \) are no longer state variables of the system. Then, it is natural to describe the microstructural dynamics of the blend in terms of mass density-free quantities. For consistency with the notation that is commonly used to denote the structural variables appearing in this model, we denote these mass density-free quantities in uppercase letters. The shape and number density of the dispersed Newtonian droplets were described in terms of a symmetric contravariant second-order droplet shape tensor \( S \) and a droplet number density \( n \), respectively. Consequently, the set of state variables of the system comprises the momentum density vector \( \mathbf{M} = \mathbf{r} \mathbf{v} \), where \( \mathbf{r} \) is the scalar mass density and \( \mathbf{v} \) is the velocity vector; the deformation tensor \( \mathbf{C} \); the droplet shape tensor \( S \); and the droplet number density \( n \). The operating space of the problem is thus defined as follows:

\[
\mathcal{P} = \{ \mathbf{M}(r,t) \in \mathbb{R}^3 \mid \mathbf{M}(r,0) = \mathbf{M}_0(r) \text{ in } \Omega, \quad \mathbf{v}(r,t) \in \mathbb{R}^{3 \times 3}, \quad \mathbf{C}(r,t) \in \mathbb{R}^{3 \times 3}, \quad \mathbf{S}(r,t) \in \mathbb{R}^{3 \times 3}, \quad n(r,t) \in \mathbb{R}^+ \}.
\]

### 2.4.2. Poisson Bracket

To derive a set of time evolution equations for the state variables of the system, we focused only on arbitrary functionals that depend only implicitly on the partial derivatives of the state variables. Their Volterra derivatives with respect to the state variables are defined as in (2.12). For all unconstrained state variables, the projection operator \( \Pi \) in (2.12) reduces to the identity operator. From the set of state variables, the structural variables \( \mathbf{C} \) and \( n \) are unconstrained. For incompressible flows, the hydrodynamic variable \( \mathbf{M} \) must be divergence-free. The treatment of the divergence-free condition is intimately connected to how a flow problem is formulated and then numerically solved. Therefore, it is standard practice to consider the hydrodynamic variable \( \mathbf{M} \) for the derivation of the flow equations as unconstrained and to impose the divergence-free condition later when a specific flow problem is formulated. Since the blend is assumed to be incompressible, it is physically reasonable to suppose that the droplet phase is also incompressible. Consequently, the droplet shape tensor \( S \) was required to satisfy the volume preservation constraint \( \int_\Omega n d\mathbf{r} = 1 \). A proper definition of the Volterra derivatives with respect to the structural variable \( S \) is given in (2.41).

The next step of the derivation procedure is to specify the master equation (2.6) together with the system Hamiltonian \( H \). On the basis of the chain rule of differentiation, the total time derivative of the functional \( F \) appearing on the left-hand side of (2.6) can be expressed as

\[
\frac{dF}{dt} = \int_\Omega \left[ \frac{\partial F}{\partial \mathbf{M}_0} \frac{\partial \mathbf{M}_0}{\partial t} + \frac{\partial F}{\partial \mathbf{C}_0} \frac{\partial \mathbf{C}_0}{\partial t} + \frac{\partial F}{\partial \mathbf{S}_0} \frac{\partial \mathbf{S}_0}{\partial t} + \frac{\partial F}{\partial n} \frac{\partial n}{\partial t} \right] d^3r. \quad (2.53)
\]

The Poisson and dissipation brackets appearing on the right-hand side of (2.6) have been described in Sec. 2.4.2 and Sec. 2.4.3, respectively. An explicit expression for \( H \) has been provided in Sec. 2.4.4.

### 2.4.3. Example

The standard procedure presented in the previous two sections was adopted to build the Poisson and dissipation brackets for a medium described in terms of \( \mathbf{M}, \mathbf{C}, \mathbf{S}, \) and \( n \). The resulting Poisson bracket is given as follows:

\[
\{F,G\} = \int_\Omega \left[ \frac{\partial F}{\partial \mathbf{M}_0} \frac{\partial G}{\partial \mathbf{C}_0} \mathbf{C}_0 + \frac{\partial F}{\partial \mathbf{C}_0} \frac{\partial G}{\partial \mathbf{M}_0} \mathbf{M}_0 + \frac{\partial F}{\partial \mathbf{S}_0} \frac{\partial G}{\partial \mathbf{S}_0} \mathbf{S}_0 \right] d^3r
\]

The standard procedure presented in the previous two sections was adopted to build the Poisson and dissipation brackets for a medium described in terms of \( \mathbf{M}, \mathbf{C}, \mathbf{S}, \) and \( n \). The resulting Poisson bracket is given as follows:
2.4. Example

The above dissipation bracket is a generalization of that for a two-coupled-mode Maxwell model described in terms of an unconstrained second-order contravariant conformation tensor $C$ and a contravariant second-order droplet shape tensor $S$, which is subject to the mathematical constraint $I_2 = 1$. The first integral with the phenomenological tensor $A^k$ accounts for the relaxation of the polymeric constituents of the matrix phase. The second and third integrals with the phenomenological tensor $A^k$ account for the relaxation of the dispersed Newtonian droplets. The third and fourth integrals with the phenomenological tensor $A^k$ account for the relaxation of the droplet number density. To ensure simplicity of the resulting time evolution equations for the state variables, an irreversible coupling of the droplet number density with the droplet shape tensor has not been considered.

2.4.4. Hamiltonian

The Hamiltonian of the system was modeled as a volume integral of the kinetic and Helmholtz free energy densities as follows:

$$H_n(M, C, S, n) = \int_{\Omega} \left[ \frac{1}{2} \rho \dot{r}^2 + \frac{1}{2} \left( 1 - \phi \right) G \frac{K}{k_b T} I_2 + \frac{1}{2} \phi \ln \left( \frac{K}{k_b T} \right) I_1 + \frac{1}{6} \phi \frac{K}{k_b T} \ln \left( \frac{n}{n_0} \right) f_1 \left( I_1, I_2 \right) \right] \, d^3 r,$$

(2.56)

where $f_1 \equiv \nabla X$ and $f_2 \equiv \left( (\nabla X)^2 - K X \right) / 2$ are the first and second invariants of the structural variable $X \in \{C, S\}$, respectively. Furthermore, $\phi$ is the total blend concentration of the droplet phase; $G$, the elastic modulus of the polymeric matrix phase, defined as $G = n_c k_b T$, with $n_c$ being the number density of the polymeric constituents of the matrix phase; $\Gamma$, the elastic modulus of the interface formed between the two immiscible fluids; $n_0$, the droplet number density at rest; and $\epsilon_1, \epsilon_2$ a measure of the asphericity of the dispersed droplets. Note that the subscript “$n$” in the notation $H_n$ indicates that only the mechanical contribution to the Hamiltonian of the system is considered. Under isothermal flow conditions, it is not necessary to consider the transfer of mechanical energy into internal degrees of freedom.

The first term in the volume integral on the right-hand side of (2.56) represents the kinetic energy density of the polymer blend. The second and third terms represent the contributions of the polymeric matrix phase to the Helmholtz free energy density. These contributions have been described in the same manner as those in the UCM model. The remaining terms within the abovementioned volume integral represent the contributions of the Newtonian droplet...
2. Thermodynamic Modeling

phase to the Helmholtz free energy density. More precisely, the fourth term represents the energy density of the elastic interface. It was taken to be proportional to the relative number density of the droplets, \( n/n_0 \), and the average surface area of a single droplet, \( p_1 \). This assumption is physically justified as long as the average deviations of the droplet shape from spherical remain small. The fourth term accounts for the effects that determine the droplet number density away from equilibrium. This term is similar in spirit to the Flory-Huggins mixing term; a logarithmic dependence on the relative number density of the dispersed droplets, \( n/n_0 \), was selected. Since the deformation of the polymeric constituents of the matrix fluid in the linear viscoelastic limit. (ii) If the droplet deformation into prolate or oblate configurations depends on the viscosity and the linear viscoelastic response of the two phases [49, 56]. Therefore, Dressler and Edwards [29] suggested two different possibilities to relate the phenomenological parameter \( p^* \) to the material properties of the polymer blend. (i) If the deformation behavior into prolate or oblate droplet configurations is assumed to depend on the viscous properties of the two immiscible phases, then \( p^* \) should be taken as their negative viscosity ratio, i.e.,

\[
p^* = -\eta_p/\eta_d,
\]

where \( \eta_d \) is the Newtonian viscosity of the droplets. Furthermore, the parameter \( \eta_p \) represents the Maxwellian viscosity of the matrix fluid, \( \eta_p = G \eta_d \). It corresponds to the viscosity of the matrix fluid in the linear viscoelastic limit. (ii) If the droplet deformation into prolate or oblate droplet configurations is assumed to depend on the viscoelastic properties of the interface formed between the two immiscible phases, then \( p^* \) should be taken as follows:

\[
p^* = -\eta_{\text{ff}}/\eta_d,
\]

where \( \eta_{\text{ff}} \) is the interfacial viscosity, \( \eta_{\text{ff}} = \Gamma \eta_d \). For the remaining phenomenological tensor \( \Lambda^\alpha \), the following simple expression was introduced:

\[
\Lambda^\alpha = 2 \frac{K_c}{\Lambda_c} \eta_d \mu_0.
\]

2.4. Phenomenological Tensors

Let us move on to the specifications of the phenomenological tensors \( \Lambda^C \), \( \Lambda^S \), \( \Lambda \), and \( \Lambda^\theta \) appearing in the dissipation bracket (2.55). The relaxation mechanism of the polymeric matrix phase was also described as in the UCM model, i.e.,

\[
\Lambda^C_{\alpha \beta \gamma} = \frac{1}{2} \frac{K_c}{\Lambda_c} \left( C_{\alpha \beta \gamma} + C_{\alpha \gamma \beta} + C_{\beta \alpha \gamma} + C_{\beta \gamma \alpha} + C_{\gamma \alpha \beta} + C_{\gamma \beta \alpha} \right),
\]

where \( \Lambda_c \) is the characteristic relaxation time of the polymeric constituents of the matrix fluid. Although the UCM model is capable of predicting a fading memory, it cannot predict a varying viscosity in a simple shear flow. A simple way of accounting for the experimentally and widely observed shear-thinning behavior of viscoelastic fluids is to allow their relaxation time to be a scalar function of the conformation tensor. As proposed by the developers of the EWM model [75], a simple power-law dependence of \( \Lambda_c \) on the first invariant of the conformation tensor, \( p_1 \),

\[
\Lambda_c = \Lambda_{c0} \left( \frac{K}{K_{c0}} p_1 \right)^{\delta},
\]

was adopted, where \( \Lambda_{c0} \) and \( K \) are a pair of phenomenological model constants. Their meaning will be discussed later in Sec. 3.1.1.

The coupling tensor \( \Lambda \) was adopted in analogy to the coupling tensor of the two-mode Maxwell model [36, 9] as follows:

\[
\Lambda^\theta_{\alpha \beta \gamma} = \frac{1}{2} \frac{(1 + p^*)^2}{\sqrt{\eta_c \theta G \sqrt{\Lambda}}} \left( C_{\alpha \beta \gamma} S_{\alpha \beta} + C_{\alpha \gamma \beta} S_{\alpha \gamma} + C_{\beta \alpha \gamma} S_{\beta \alpha} + C_{\beta \gamma \alpha} S_{\beta \gamma} + C_{\gamma \alpha \beta} S_{\gamma \alpha} + C_{\gamma \beta \alpha} S_{\gamma \beta} \right),
\]

where \( \theta \) is a coupling parameter. For \( \theta > 0 \), the phenomenological tensor \( \Lambda \) accounts for oblate droplet configurations. To ensure simplicity of the resulting flow equations, the coupling parameter \( \theta \) was restricted to some constant. However, it may also be considered as a scalar function of the structural variables \( C \) and \( S \), if so desired.

The relaxation of the interface formed between the polymeric matrix fluid and the dispersed Newtonian droplets was described as

\[
\Lambda^\theta_{\alpha \beta \gamma} = \frac{1}{\Lambda_c} \left( \frac{(1 + p^*)^2}{2} \left( S_{\alpha \beta} \delta_{\gamma \gamma} + S_{\alpha \gamma} \delta_{\beta \beta} + S_{\beta \alpha} \delta_{\gamma \gamma} + S_{\beta \gamma} \delta_{\alpha \alpha} \right) + \frac{3p^*}{\Lambda_c} \left( \delta_{\alpha \beta} \delta_{\gamma \gamma} + \delta_{\alpha \gamma} \delta_{\beta \beta} + \delta_{\beta \alpha} \delta_{\gamma \gamma} + \delta_{\beta \gamma} \delta_{\alpha \alpha} \right) \right),
\]

where \( \Lambda_c \) is the characteristic time scale of interfacial relaxation. Furthermore, \( p^* \) is a phenomenological parameter, as specified below. The first and second terms within the outer brackets on the right-hand side of (2.60) account for oblate or prolate droplet configurations at the start-up of a steady simple shear flow, respectively.

The physical mechanisms governing the deformation of Newtonian droplets dispersed in a matrix fluid are not well understood. Experimental measurements reveal that the deformation behavior into prolate or oblate configurations depends on the viscosity and the linear viscoelastic response of the two phases [49, 56]. Therefore, Dressler and Edwards [29] suggested two different possibilities to relate the phenomenological parameter \( p^* \) to the material properties of the polymer blend. (i) If the deformation behavior into prolate or oblate droplet configurations is assumed to depend on the viscous properties of the two immiscible phases, then \( p^* \) should be taken as their negative viscosity ratio, i.e.,

\[
p^* = -\eta_p/\eta_d,
\]

where \( \eta_d \) is the Newtonian viscosity of the droplets. Furthermore, the parameter \( \eta_p \) represents the Maxwellian viscosity of the matrix fluid, \( \eta_p = G \eta_d \). It corresponds to the viscosity of the matrix fluid in the linear viscoelastic limit. (ii) If the droplet deformation into prolate or oblate droplet configurations is assumed to depend on the viscoelastic properties of the interface formed between the two immiscible phases, then \( p^* \) should be taken as follows:

\[
p^* = -\eta_{\text{ff}}/\eta_d,
\]

where \( \eta_{\text{ff}} \) is the interfacial viscosity, \( \eta_{\text{ff}} = \Gamma \eta_d \).
where \( \lambda_n \) and \( f_n \) are the characteristic time scale and the probability of the breakup and coalescence processes. The parameter \( f_n \) may be considered as a scalar function of the phenomenological parameter \( \rho' \) and the structural variables \( C \) and \( S \), if so desired.

### 2.4. Example

To close the general set of flow equations, an explicit expression for the extra stress tensor can be determined through the Poisson and dissipation brackets, i.e.,

\[
\sigma = (1 - \phi) G \left( \frac{K}{\eta_0} C - \mathbf{1} \right) + \phi \left\{ \frac{n}{\eta_0} \left( \mathbf{f} S - S S - \frac{2}{3} \mathbf{f} \mathbf{f} \right) \right\} + \frac{1}{3} \frac{K}{\eta_0} \ln \left( \frac{n}{\eta_0} \right) \left( \mathbf{f} S + \mathbf{f} \mathbf{f} C - \frac{1}{3} \mathbf{f} \mathbf{f} \mathbf{f} \right). \tag{2.68}
\]

Equation (2.64) is the Cauchy momentum balance equation. It is important to realize that we have described the polymer blend as one single fluid having a heterogeneous microstructure. Consequently, the momentum density vector is an averaged state variable that determines the hydrodynamics of the polymer blend.

Equation (2.65) is the conformation tensor equation. The first three terms on its right-hand side are the reversible contributions to the dynamics of the polymeric constituents of the matrix fluid. Together with its left-hand side, these terms constitute the upper-convected derivative of the conformation tensor. The fourth term on the right-hand side accounts for the relaxation of the polymeric constituents of the matrix fluid. The fifth term on the right-hand side accounts for the influence of the dynamics of the droplets away from equilibrium. The sixth term on the right-hand side represents the influence of the coupling between the two immiscible phases.

Equation (2.66) is the droplet shape equation. The first four terms on its right-hand side represent the reversible contributions to the dynamics of the shape of the dispersed Newtonian droplets. Together with its left-hand side, these terms constitute the generalized upper-convected derivative of the droplet shape tensor. The fifth and sixth terms on the right-hand side account for interfacial relaxation. Finally, the seventh term represents the influence of the coupling between the two immiscible phases.

Equation (2.67) is the droplet number density equation. The first term on its right-hand side represents the reversible contribution to the dynamics of the breakup and coalescence processes of the droplets. Together with its left-hand side, this term constitutes the material derivative of the droplet number density. The first and second terms in the brackets on the right-hand side account for droplet coalescence and breakup, respectively.

Equation (2.68) provides an explicit expression for the extra stress tensor. For a low total volume fraction of the droplet phase (\( \phi \to 0 \)), the extra stress of the polymer blend is mainly determined by the dynamics of the polymeric matrix fluid and the coupling terms are dominant in the droplet shape equation. For a high total volume fraction of the droplet phase (\( \phi \to 1 \)), the coupling terms are dominant in the conformation tensor equation and the dynamics of the dispersed Newtonian droplets mainly determines the extra stress of the polymer blend.

The set of model equations represents a system of coupled nonlinear partial differential equations (PDEs). To solve this system of PDEs for a given flow, the divergence-free condition of...
2. Thermodynamic Modeling

the momentum density vector and some appropriate initial and boundary conditions must be
imposed. A simple analytical solution is obtained in the equilibrium state of rest. Then, we
have $L = 0$, which implies $C = 1$, $S = 1$, and $n = n_0$.

2.4.7. Thermodynamic Admissibility

For a model that describes nonisothermal and compressible flow as being thermodynamically
admissible, it has to satisfy the requirements (2.8) and (2.9). Under isothermal flow condi-
tions, only the mechanical part of the Hamiltonian of the system, $H_m$, is considered; thus,
the requirement (2.8) can be ignored. Furthermore, the entropy production then results only
from the dissipation of the mechanical energy, and the requirement (2.9) is replaced with the
following equivalent one [11, 9]:

$$\frac{dH_m}{dt} = \{H_m, H_m\} \leq 0. \tag{2.69}$$

In addition to some mathematical requirements that need to be taken into account, the ther-
modynamic requirement (2.69) places some physical restrictions on the permissible range of
model parameter values. It is important to realize that the requirement (2.69) must be fulfilled
for the model predictions to be physically reliable. Therefore, it is advisable to check the
requirement (2.69) before extracting any microstructural information from the model predic-
tions.

Unfortunately, the requirement (2.69) in case of the EDE model is too complex for the admis-
sible range of its parameters values to be determined analytically. According to the definition
of the parameters $\lambda_{c_0}$, $A_0$, and $A_\tau$ as characteristic time scales, they should be non-negative
quantities. In a numerical analysis performed for the start-up of a weak simple shear flow [30],
it has been shown that the parameter $p^*$ should be a negative quantity. In general, it is also
possible that the contribution of the dissipation of the mechanical energy due to the coupling
of the state variables is positive. Therefore, there is a priori no restriction imposed on the
choice of the sign of the parameter $\theta$.

3. Eccentric Taylor-Couette Flow of Polymeric Solutions and Melts

In the previous chapter, we presented the theory of the EDE model. Then, we developed a
novel numerical approach for solving this model for a steady eccentric Taylor-Couette flow.
This numerical approach is unique in that the nonlinear system of discretized algebraic flow
equations is solved iteratively using a Newton-Krylov method along with an inverse-based
ILU preconditioner. For code development, we considered the power-law EWM model. This
model is obtained from the EDE model if only the matrix phase is considered and a vanishing
total volume fraction of the droplet phase is assumed. It describes the flow of solutions and
melts of polymeric constituents whose dynamics is physically unconstrained. Using only one
single relaxation time, the model can accurately predict the shear viscosity and first normal
stress of these polymeric fluids in a simple shear flow. However, like many other viscoelastic
fluid models, it fails to predict a nontrivial second normal stress difference. Interestingly, it
is capable of predicting a maximum in the elongational viscosity that is followed by a de-
crease with increasing elongation rate. Because it can control only the slope of the extensional
viscosity and not its maximum value, multimode versions of other viscoelastic fluid models,
such as the exponential version of the PTT model, are better suited to describe the extensional
behavior of polymeric melts.

This chapter deals with numerical simulations of a steady eccentric Taylor-Couette flow of
polymeric solutions and melts. In Sec. 3.1.1, we introduce the flow equations according to the
power-law EWM model. In Sec. 3.1.2, we introduce the benchmark geometry. In Sec. 3.1.3,
we describe how the flow problem was formulated as a boundary value problem (BVP). In
Sec. 3.1.4, we present the pseudospectral collocation method that was used for its spatial dis-
cretization. In Sec. 3.2, we introduce the preconditioned Newton-Krylov method that was
adopted to solve the resulting nonlinear system of discretized algebraic equations. The vali-
dation of the numerical method is reported in Sec. 3.3. Sample calculations are presented in
Sec. 3.4. Conclusions are presented in Sec. 3.5.
3. Eccentric Taylor-Couette Flow of Polymeric Solutions and Melts

3.1. Problem Formulation

3.1.1. Flow Equations

The flow of a polymeric fluid is typically described using a set of conservation balance equations and some constitutive equation. In this work, we select the EWM model as the constitutive equation. Assuming isothermal and incompressible flow conditions, the physical variables of the flow problem are the momentum density vector \( \mathbf{M} = \rho \mathbf{v} \), with \( \rho \) being the scalar mass density and \( \mathbf{v} \) being the velocity vector, and the symmetric contravariant second-order conformation tensor \( \mathbf{C} \). The general set of flow equations is thus given as

\[
\begin{align*}
\frac{\partial \rho}{\partial t} &= -\nabla \cdot (\rho \mathbf{v}) , \\
\frac{\partial \mathbf{v}}{\partial t} &= -\mathbf{v} \cdot \mathbf{L}^T - \nabla p + \nabla \cdot \mathbf{\sigma} , \\
\frac{\partial \mathbf{C}}{\partial t} &= -\mathbf{v} \cdot \nabla \mathbf{C} + \mathbf{C} \cdot \mathbf{L}^T + \mathbf{L} \cdot \mathbf{C} - \frac{1}{\lambda} \left( \mathbf{C} - \frac{k_B T}{k} \right) ,
\end{align*}
\]

where \( t \) is the time and \( \mathbf{L} \) the transpose of the velocity gradient tensor; \( \mathbf{L} = (\nabla \mathbf{v})^T \). Equation (3.1) is the continuity equation for the mass density \( \rho \). Equation (3.2) is the Cauchy momentum balance equation for the velocity vector \( \mathbf{v} \), where the pressure and the extra stress tensor are denoted by \( p \) and \( \mathbf{\sigma} \), respectively. Equation (3.3) is the time evolution equation for the conformation tensor \( \mathbf{C} \), where \( \lambda \) is a variable relaxation time; \( k \), the (Hookean) elastic constant; \( k_B \), the Boltzmann factor; and \( T \), the absolute temperature. The relaxation time, \( \lambda \), of the fluid is allowed to be a scalar function of the conformation tensor. As proposed by Souvaliotis and Beris [75], we adopt a simple power-law dependence of \( \lambda \) on the first invariant of \( \mathbf{C} \), i.e.,

\[
\lambda = \lambda_0 \left( \frac{1}{3} \frac{K}{k_B T} \right)^{\frac{1}{2}} ,
\]

where \( I_1 \) is the first invariant of \( \mathbf{C} \). Furthermore, \( \lambda_0 \) and \( k \) are a pair of model constants. In the regime of linear viscoelasticity, \( \lambda \) reduces to the first model constant \( \lambda_0 \). In a double-logarithmic plot, the second model constant \( k \) controls the slope of both the varying shear and the extensional viscosity with respect to the shear and the extension rate, respectively. At larger shear rates, the slope of the shear viscosity with respect to the shear rate is approximately equal to \( 2k/(1 - 2k) \). The slope of the extensional viscosity with respect to the extension rate differs depending on the type of extensional flow. Because of the simplicity of the chosen power-law dependence on \( I_1 \), the model constants \( k \) and \( \lambda_0 \) can be easily determined from experimental data obtained in steady simple shear and/or extensional flows. Note that the UCM model is a limiting case of the EWM model. It is recovered by setting \( k = 0 \). To close the above system of time evolution equations, the extra stress tensor in (3.2) is related to the conformation tensor in (3.3) by the linear constitutive relationship

\[
\mathbf{\sigma} = G \left( \frac{K}{k_B T} \mathbf{C} - \mathbf{I} \right) ,
\]

where \( G \) is the modulus of elasticity.

3.1.2. Flow Geometry

The benchmark geometry consists of two infinitely long cylinders of radii \( R_1 \) and \( R_2 \) (\( R_1 < R_2 \)). Their axes are separated by a distance \( e \). The inner cylinder is rotated clockwise at a constant angular velocity \( V \). The outer cylinder is kept stationary. The configuration of the flow geometry is characterized by the eccentricity ratio \( e \) and the relative gap width \( \mu \). These parameters are defined as

\[
\begin{align*}
\epsilon &= \frac{e}{R_1 - R_2} , \\
\mu &= \frac{R_2 - R_1}{R_1} ,
\end{align*}
\]

The eccentric cylinder system is described in a bipolar cylindrical coordinate system \((\xi, \theta, z)\) [14]. This coordinate system is convenient because it allows an easy description of the cylindrical boundaries. It is related to the Cartesian coordinate system \((x, y, z)\) as

\[
\begin{align*}
x &= a \sin \xi \cosh \theta + \cos \theta , \\
y &= a \sin \theta \cosh \xi + \cos \theta , \\
z &= \pm \xi ,
\end{align*}
\]

where the constant \( a \) can be expressed in terms of the geometrical quantities introduced above as in [13]:

\[
a = R_1 \sqrt{\frac{1 - e^2}{e} \left( 1 + \mu + \frac{e^2}{4} \mu^2 \right)} .
\]

Consider that the axes of the cylinders are positioned on the positive x-axis and tangentially to the xy-plane of the Cartesian coordinate system. Figure 3.1 shows a tangential cross-section of the flow geometry in the bipolar cylindrical coordinate system. The coordinate curves of constant \( \xi \) and \( \theta \) values are circles that orthogonally intersect with each other. The constant \( a \) determines the location of the poles of the bipolar cylindrical coordinate system on the x-axis of the Cartesian coordinate system. The boundaries of the flow geometry exactly match with coordinate surfaces. Their locations are given by [14]

\[
\xi_i = \arcsin \left( \frac{a}{R_i} \right) (i = 1, 2) .
\]
3.1. Problem Formulation

3.1.3. Boundary Value Problem

In this work, we solved the flow equations introduced in Sec. 3.1.1 for the flow in the benchmark geometry introduced in Sec. 3.1.2. A modified stream function formulation [13, 23, 76] was adopted to reduce the size of the resulting BVP. In contrast to these researchers, we did not use a modified extra stress tensor as a primary variable of the flow problem. We preferred the use of a modified conformation tensor because the EWM model is a conformation-tensor-based viscoelastic fluid model.

Henceforth, we make use of dimensionless quantities. The quantities used in this study are rendered dimensionless using the scalings \( \bar{p} = p/R, \bar{v} = v/V, \bar{\sigma} = \sigma/G, \bar{C} = (K/k_{\lambda}) \bar{C} \), and \( \bar{\nabla} = R \nabla \). A Deborah number consistent with these scalings is defined as \( De \equiv \lambda V/R \).

In addition to the assumptions made in Sec. 3.1.1 and 3.1.2, we assumed the flow to be laminar. Furthermore, we assumed that the inertia is vanishingly small so that the convective term in the Cauchy momentum balance equation (3.2) could be neglected. Hence, the velocity field takes the form

\[
\bar{\nabla} = \left[ \bar{v}_r(\xi, \theta), \bar{v}_\theta(\xi, \theta), 0 \right].
\]  

(3.10)

Moreover, we assumed that the velocity field satisfies no-slip and no-penetration conditions at the boundaries, i.e.,

\[
\bar{v}_r|_{\xi = 0} = 0, \quad \bar{v}_r|_{\xi = 1} = 1, \quad \bar{v}_\theta|_{\xi = 0} = 0, \quad \bar{v}_\theta|_{\xi = 1} = 0.
\]  

(3.11)

(3.12)

In the following, we formulate the BVP. First, let us introduce the stream function \( \bar{\psi} \), which has been made dimensionless using the scaling \( \sigma V \). It is related to the velocity field as follows:

\[
\bar{\nabla} = \left[ \frac{\partial \bar{\psi}}{\partial \theta}, -\frac{\partial \bar{\psi}}{\partial \xi}, 0 \right].
\]  

(3.13)

where \( \chi = \cosh \xi + \cos \theta \). A modified stream function \( \tilde{\psi} \) and a modified conformation tensor \( \tilde{C} \) were selected as the primary variables. They are defined as

\[
\tilde{\psi} = \bar{\psi} / \chi, \quad \tilde{C} = \bar{C} / \chi.
\]  

(3.14)

Like the stream function, its modification satisfies the mass balance equation (3.1) identically. Consequently, only the Cauchy momentum balance equation (3.2) and the EWM model (3.3)–(3.5) must be considered from the flow equations introduced in Sec. 3.1.1. Utilizing the above definition of the primary variables, we obtain the following nontrivial components of the conformation tensor equation (3.4):

\[
\tilde{C}_{\tilde{\psi}} = \frac{1}{\chi} + De \left( \chi \frac{\partial \tilde{\psi}}{\partial \theta} + \tilde{\psi} \sin \theta \right) \frac{\partial^2 \tilde{C}_{\tilde{\psi}}}{\partial \xi^2} + \left( \chi \frac{\partial \tilde{\psi}}{\partial \xi} + \tilde{\psi} \sin \theta \right) \frac{\partial^2 \tilde{C}_{\tilde{\psi}}}{\partial \theta^2}.
\]
where $\tilde{Q}$ is some yet unknown constant. The constant $\tilde{Q}$ has the physical meaning of an azimuthal flux between the cylindrical boundaries that has been rendered dimensionless using the scaling $aV$. It was found simultaneously with the entire flow problem by requiring the pressure to be $2\pi$-periodic in the $\theta$-direction. The pressure periodicity condition is obtained by integrating the first component of the Cauchy momentum balance equation (3.2) with respect to $\theta$ from 0 to $2\pi$ at a fixed radial position $\xi = \xi_1$ and setting the resulting integral expression to zero. Substituting the constitutive relationship (3.5) into the obtained equation and reusing the definition for $C^e$ given in (3.14) yields the integral equation

$$\int_0^{2\pi} \left( \frac{\partial C^e_{11}}{\partial \xi} + \frac{\partial C^e_{12}}{\partial \theta} \right) \chi C_{ij} \sin \theta - C_{ij} \sinh \xi \frac{\partial C^e_{ij}}{\partial \theta} \right) d\theta = 0.$$  \hspace{1cm} (3.22)

To ensure that all other flow variables are $2\pi$-periodic in the $\theta$-direction, we impose this condition explicitly for the primary variables. The nonlinear system of partial differential equations (3.15)-(3.19) and (3.22), together with the boundary conditions (3.20) and (3.21) and the $2\pi$-periodicity condition of the primary variables in the $\theta$-direction, form a BVP.

### 3.1.4. Discretization

To convert the BVP into a system of algebraic equations, it was discretized using a pseudospectral collocation method. The radial dependence of its unknown variables had first to be transformed to the interval $[-1, 1]$. The transformation was achieved by the linear mapping $[70]

$$\xi = \frac{(\xi - \xi_1)(\xi' - 1)}{2} + \xi_1.$$  \hspace{1cm} (3.23)

The following global interpolation polynomial was used to approximate the unknown variables of the BVP:

$$s_{MN}(\xi', \theta) = \sum_{m=0}^{M} \sum_{n=0}^{N} s_{i,j} \xi^{m} \chi^{n} b_{i}(\theta),$$  \hspace{1cm} (3.24)

where $s_{i,j}$ are yet unknown coefficients; $\xi(\xi')$, Lagrange polynomials of degree $M$; and $b_{i}(\theta)$, trigonometric polynomials of degree $N$. Their explicit expressions can be found in App. A.

The above interpolation polynomial is based on the grid of collocation points

$$\xi_{ij} = \cos(i \pi M), \chi_{ij} = \frac{\pi j}{N} \hspace{1cm} i = 0, \ldots, M, \hspace{0.5cm} j = 0, \ldots, 2N - 1.$$  \hspace{1cm} (3.25)

where $\xi_{ij}$ and $\chi_{ij}$ are referred to as Chebyshev-Gauss-Lobatto and Fourier points, respectively.

By definition, the coefficients of the above interpolation polynomial are identical to its values at the collocation points, i.e., $s_{i,j} = s_{i,j}(\xi_{ij}, \chi_{ij})$. 

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3. Eccentric Taylor-Couette Flow of Polymeric Solutions and Melts

3.1. Problem Formulation
In pseudospectral collocation methods, the derivatives of the interpolation polynomials are computationally evaluated at the collocation points as matrices. Chebyshev and Fourier differentiation matrices of the first and second order reduce to simple analytical formulas and can be found in App. A. The Chebyshev differentiation matrix of the $p$-th order is determined by

$$
\left( D_p^{ij} \right)_{ij} = \frac{d^p g_i (\xi)}{d \xi^p} \bigg|_{\xi = \xi_j}, \quad i, j = 0, \ldots, M. \quad (3.26)
$$

Irrespective of whether the Chebyshev differentiation matrices are computed analytically or numerically, they suffer from round-off errors for a large $M$. These errors are most pronounced in the upper-left and lower-right corners of the Chebyshev differentiation matrices [22]. To minimize them, we adopted the negative sum trick proposed by Bayliss et al. [7]. This trick involves computing the diagonal elements from the non-diagonal ones by using the relation

$$
\sum_{j=0}^{M} D_p^{ij} = 0 \text{ for } i = 0, \ldots, M, \text{ i.e.,}
$$

$$
\left( D_p^{00} \right)_{00} = \sum_{j=1}^{M} D_p^{0j}, \quad i = 0, \ldots, M. \quad (3.27)
$$

After the BVP was spatially discretized, it was evaluated at the collocation points of the Chebyshev-Fourier grid. To be more precise, the discretized form of (3.15)–(3.17) together with (3.18) was evaluated at all the collocation points. Because the boundary conditions (3.20) and (3.21) uniquely define the flow field at the boundaries, it was sufficient to evaluate the discretized form of (3.19) at the inner radial points, i.e., at $\xi_i, i = 1, \ldots, M - 1$. The boundary conditions (3.20) and the discretized form of the integral equation (3.22) were evaluated at the corresponding radial boundary point, i.e., at $\xi_0$. To achieve numerical stability, the boundary conditions (3.21) were treated differently. They were satisfied by modifying the polynomial approximation of $\tilde{\psi}^*$ as [23]

$$
\tilde{\psi}_{ME} (\xi, \theta) = (\xi - \xi_1)^2 \sum_{j=0}^{M-1} \sum_{k=0}^{2N-1} \hat{g}_j (\xi^*) \tilde{h}_j (\theta). \quad (3.28)
$$

By approximating the unknown variables of the BVP in the azimuthal direction with $2\pi$-periodic polynomials, the $2\pi$-periodicity condition is automatically satisfied and need not be considered further. The integral in (3.22) was evaluated using a 10-point Gauss-Legendre quadrature [68]. Overall, the evaluation of the discretized BVP resulted in a nonlinear system of $4 \times (M + 1) \times 2N + 1$ algebraic equations. In the next section, we describe the numerical solution of the same.

3.2. Solution of Discretized Equations

Newton methods are frequently used to solve the nonlinear systems of equations arising from the discretization of viscoelastic fluid flow problems. These methods are attractive because they can completely address the coupling between the flow variables. Additionally, they enable rapid convergence, provided that the initial estimate is sufficiently close to a solution of the nonlinear system. Direct methods such as some variants of the lower-upper decomposition method are commonly employed to solve the linear Jacobian system within the Newton iterations. A viable alternative is to solve the linear Jacobian system iteratively using one of the various Krylov subspace methods. Krylov subspace methods are considered as alternatives because they can solve the linear system in some small-dimensional subspace. For Krylov subspace methods to converge within a moderate number of iterations, they must usually be used in combination with some preconditioner. However, Krylov subspace methods have not yet been widely employed in practical applications because of the lack of robust general-purpose preconditioners. Most general-purpose preconditioners are derived from ILU factorizations of the coefficient matrices. Bollhöfer and Saad [16] recently developed a new ILU preconditioner with increased robustness. The key feature of this ILU preconditioner is that the magnitude of the perturbations in the inverse triangular factors is controlled during its construction.

This section deals with the numerical solution of the discretized benchmark problem. We developed a new numerical approach to solve the nonlinear system of discretized algebraic flow equations. The proposed approach is unique in that this nonlinear system is solved iteratively using a Newton-Krylov method along with the abovementioned inverse-based ILU preconditioner. The generalized minimum residual (GMRES) method [73] was selected as the Krylov subspace method. In Sec. 3.2.1, we present the inexact Newton method. In Sec. 3.2.2, we describe the improvement in its radius of convergence. The GMRES method is introduced in Sec. 3.2.3. The novel preconditioner is presented in Sec. 3.2.4.

3.2.1. Newton Iterations

The nonlinear system of discretized algebraic flow equations can be represented as

$$
F (x) = 0, \quad (3.29)
$$

where $F : \mathbb{R}^n \rightarrow \mathbb{R}^n$ is a nonlinear vector function. Let $x_0 \in \mathbb{R}^n$ be an initial estimate for the solution of (3.29). At the $k$-th Newton iteration, a new approximate solution of (3.29), i.e., $x_{k+1} \in \mathbb{R}^n$, was calculated according to the recurrence

$$
x_{k+1} = x_k + \Delta_k, \quad (3.30)
$$
The vector \( \mathbf{s}_k \in \mathbb{R}^n \) in (3.30) is referred to as a Newton step and was determined by approximately solving the linear system
\[
\mathbf{J}(\mathbf{x}_k) \cdot \mathbf{s}_k = -\mathbf{F}(\mathbf{x}_k),
\]
where \( \mathbf{J}(\mathbf{x}_k) \) denotes the Jacobian matrix of \( \mathbf{F}(\mathbf{x}_k) \).

The Newton iterations were considered to have converged if the following condition [66] was satisfied:
\[
W \left( \frac{1}{N} \sum_{i=1}^{n} \left( \frac{\|\mathbf{x}(i)\|}{\|\mathbf{x}(0)\|} + e_i \right)^2 \right) < 1,
\]
where \( W \) is a given constant. The parameters \( e_i \) and \( e_s \) are some relative and absolute tolerances, respectively. The convergence was checked using the above criterion because it is effective irrespective of the problem size. Furthermore, the combined control over the absolute and relative errors permits adequate resolution of physical quantities that differ widely in magnitude.

### 3.2. Solution of Discretized Equations

To minimize the number of backtracking steps, the constant \( \theta \in (0,1) \) in (3.35) cannot be chosen arbitrarily. It was determined by minimizing a polynomial in \( \theta \) that interpolates \( f(\mathbf{x}_k + \theta \mathbf{r}_k) \). In the reduction of the first step, we minimized a quadratic \( g(\theta) \) that satisfies \( g(0) = f(\mathbf{x}_k) \), \( (d/d\theta)g(0) = (d/d\theta)f(\mathbf{x}_k + \theta \mathbf{r}_k) \|g\|, \) and \( g(1) = f(\mathbf{x}_k + \mathbf{r}_k) \). In the reduction of all subsequent steps, we minimized a cubic \( g(\theta) \) that satisfies the conditions of the first-step reduction and the additional condition \( g(\theta_{\text{max}}) = f(\mathbf{x}_k + \theta_{\text{max}} \mathbf{r}_k) \), where \( \theta_{\text{max}} \) is equal to the constant \( \theta \) from the reduction of the previous step. To prevent scaled Newton steps \( \delta \mathbf{r}_k \) from being too short or too long, backtracking was safeguarded by restricting \( \theta \in [\theta_{\text{min}}, \theta_{\text{max}}] \).

#### 3.2.3. Krylov Subspace Method

To describe how the linear Jacobian system (3.31) was solved within the Newton iterations, we rewrite it as follows:
\[
\mathbf{A} \cdot \mathbf{x} = \mathbf{b}.
\]

The GMRES method was selected as the iterative linear solver. This is a standard Krylov subspace method for solving general nonsymmetric linear systems of equations. Let \( \mathbf{x}_0 \) be some initial estimate for the solution of (3.36) and let \( \mathbf{r}_0 = \mathbf{b} - \mathbf{A} \cdot \mathbf{x}_0 \) be its corresponding residual. At the \( m \)-th GMRES iteration, an approximate solution of (3.36), i.e.,
\[
\mathbf{x}_m = \mathbf{x}_0 + \mathbf{V}_m \cdot \mathbf{y}_m,
\]
was sought, where \( \mathbf{V}_m \) is a matrix whose columns form a basis of the \( m \)-th Krylov subspace that is spanned by the power vectors \( \mathbf{r}_0, \mathbf{A} \cdot \mathbf{r}_0, \ldots, \mathbf{A}^{m-1} \cdot \mathbf{r}_0 \), i.e.,
\[
\mathcal{K}_m = \text{span} \{ \mathbf{r}_0, \mathbf{A} \cdot \mathbf{r}_0, \ldots, \mathbf{A}^{m-1} \cdot \mathbf{r}_0 \}.
\]

The vector \( \mathbf{y}_m \) in (3.37) was determined by minimizing the Euclidean norm of the residual \( \mathbf{r}_m = \mathbf{b} - \mathbf{A} \cdot \mathbf{x}_m \) over \( \mathcal{K}_m \), i.e.,
\[
\|\mathbf{r}_m\| = \min_{\mathbf{z} \in \mathcal{K}_m} \|\mathbf{b} - \mathbf{A} \cdot \mathbf{z}\|.
\]

An obvious choice for a basis of \( \mathcal{K}_m \) is the power basis \( \{ \mathbf{r}_0, \mathbf{A} \cdot \mathbf{r}_0, \ldots, \mathbf{A}^{m-1} \cdot \mathbf{r}_0 \} \). However, this basis is not a good choice from a numerical point of view since it is likely to be ill-conditioned. Therefore, another basis, termed the Arnoldi basis, was used instead. This basis is constructed via the well-known Arnoldi process, which is an orthonormalization process applied to the power basis \( \{ \mathbf{r}_0, \mathbf{A} \cdot \mathbf{r}_0, \ldots, \mathbf{A}^{m-1} \cdot \mathbf{r}_0 \} \). In the usual algorithmic implementation of the GMRES method, the orthogonalization within the Arnoldi process is performed using the classical Gram-Schmidt algorithm, the modified Gram-Schmidt algorithm, or the Householder algorithm. In this work, we adopted the modified Gram-Schmidt algorithm, which is
a rearrangement of the classical Gram-Schmidt algorithm. It has better numerical stability than the classical Gram-Schmidt algorithm; furthermore, it is less expensive than the Householder algorithm in terms of arithmetic operation counts. If a Gram-Schmidt algorithm is used within the Arnoldi process, the orthonormal basis \( \{v_1, \ldots, v_m\} \) of \( K_m \) is constructed as follows. Starting with the normalized vector \( v_1 = v_1 / \| v_1 \| \) as a basis for \( K_1 \), the orthonormal basis for \( K_{m+1} \) is constructed recursively from \( K_1 \) by orthonormalizing the vector \( A \cdot v_1 \) against \( K_1 \). As a by-product of the Arnoldi process, an upper Hessenberg matrix \( H_n \in \mathbb{R}^{(m+1) \times m} \) is formed:

\[
A \cdot V_n = V_{n+1} \cdot H_n. \tag{3.40}
\]

Using \( y_0 = A \cdot x_0 + b \), the explicit expression for \( x_0 \) given in (3.37), and the above Arnoldi relation, the least-squares problem (3.39) can be reformulated as

\[
\| r_0 \|_2 = \min_{y_0 \in \mathbb{R}^m} \| y_0 - A \cdot V_n \cdot y \|_2.
\]

(3.41)

To solve the above \((m+1) \times m\) least-squares problem, the upper Hessenberg matrix \( H_n \) is factored into the product \( Q_n \cdot R_n \), where \( Q_n \in \mathbb{R}^{(m+1) \times (m+1)} \) is an orthogonal matrix and \( R_n \in \mathbb{R}^{(m+1) \times m} \) is an upper triangular matrix whose \((m+1)\)-th row has zero elements. This is commonly accomplished by means of Givens rotations, since they allow annihilation of the nonzero subdiagonal elements of the matrix \( R_n \) specifically. Using the QR factorization and considering \( Q_n \) as unitary leads to the following reformulation of the least-squares problem (3.42):

\[
\| r_{n+1} \|_2 = \min_{y_{n+1} \in \mathbb{R}^m} \| | y_{n+1} - \overline{Q}_n \cdot \overline{R}_n \cdot y \|_2.
\]

(3.43)

where \( \overline{Q} = \frac{1}{\| v_1 \|} e_1 \). Since \( \overline{Q}_n \) is an \((m+1) \times m\) upper triangular matrix whose \((m+1)\)-th row has zero elements, the solution of (3.43) is the same as that of the upper triangular system

\[
R_n \cdot y_n = \overline{r}_n.
\]

(3.44)

where \( \overline{r}_n \) and \( \overline{e}_n \) are obtained by removing the \((m+1)\)-th row from \( \overline{R}_n \) and the \((m+1)\)-th component from \( \overline{y}_n \), respectively.

The GMRES iterations were considered to have converged if the condition

\[
\| r_n \|_2 \leq \eta \| b \|_2
\]

was satisfied, where \( \eta \in [0, 1) \) is a given tolerance. From the solution of the triangular system (3.44), it is clear that the residual norm \( \| r_n \|_2 \) is nothing but the absolute value of the \((m+1)\)-th component of the vector \( \overline{e}_n \) [71]. Therefore, convergence can be checked without explicitly evaluating the residual norm \( \| r_n \|_2 \). Consequently, the triangular system (3.44) must be solved only if the solution \( x_0 \) is of interest. However, in order to check convergence, it is still necessary to perform QR factorization. In practice, it is performed recursively at every iteration; that is \( \overline{Q}_n \) and \( \overline{R}_n \) are determined from \( \overline{Q}_{n-1} \) and \( \overline{R}_{n-1} \), respectively. This facilitates regular checking of convergence at no extra cost, and thus the iteration process can be stopped as soon as convergence has been achieved.

### 3.2.4. Preconditioning

Incomplete lower-upper preconditioners comprise one of the most popular classes of preconditioners for Krylov subspace methods. Preconditioners from this class are constructed by approximately factoring the coefficient matrix of a linear system into a product of lower and upper triangular matrices. To reduce the computation time and the memory costs, the factorization is performed incompletely, i.e., elements are dropped whenever they satisfy certain structural and/or numerical criteria. Different ILU preconditioners are generated by using different forms of factorizations and/or different criteria for dropping elements. A standard ILU factorization of the coefficient matrix of the linear system (3.36) can be written as

\[
A = L \cdot D \cdot U + E,
\]

(3.46)

where \( L, U \in \mathbb{R}^{m \times m} \) are unit lower triangular matrices, \( D^{m \times m} \) is a diagonal matrix, and \( E^{m \times m} \) is the error matrix containing the elements dropped during the factorization. The linear system (3.36) can be preconditioned in different ways. For instance, applying the preconditioner \( M = L \cdot D \cdot U \) from the left gives

\[
M^{-1} A \cdot x = M^{-1} b
\]

(3.47)

Another possibility is right preconditioning, i.e.,

\[
A \cdot M^{-1} \cdot y = b,
\]

(3.48)

\[
x = M^{-1} \cdot y
\]

(3.49)
where (3.48) is the linear system to be solved and (3.49) is used to recover the solution of the original linear system (3.36). In addition, two-sided preconditioning is also possible. When a minimum residual method is selected as the Krylov subspace method, it is standard practice to apply the preconditioner from the right. This is because right preconditioning does not affect the commonly used convergence check of the residual norm during the iteration process. Since Krylov subspace methods use the coefficient matrix of a linear system only as matrix-vector products, the explicit computation of the matrix-matrix product $M^{-1}A$ or $A \cdot M^{-1}$ can be avoided. The iterative solution requires only matrix-vector products with $A$ and solutions of linear systems of the form $M \cdot x = r$. The convergence of Krylov subspace methods depends on how well the coefficient matrix of a linear system approximates the identity matrix. In case of right preconditioning, we have

$$A \cdot M^{-1} = I + E \cdot M^{-1}. \quad (3.50)$$

Most ILU preconditioners lack of robustness because they are constructed in such a manner that $\|E\|$ remains small. However, (3.50) shows that the magnitude of $\|E\|$ is just as important. Bollhöfer and Saad [16] recently proposed a novel ILU preconditioner that overcomes this drawback. A key feature of this preconditioner is the fact that the growth of the elements of the inverse triangular factors is controlled by an inverse-based pivoting and dropping approach during the factorization. For this approach to work properly, the coefficient matrix is preprocessed by scaling and reordering. Furthermore, the factorization is carried out in a hierarchical fashion within a multilevel environment. The multilevel inverse-based ILU preconditioner is available as software package ILUPACK [17].

**Multilevel Inverse-based ILU Preconditioning**

To enhance the convergence rate of the GMRES method, we preconditioned the linear system (3.36) with a multilevel inverse-based ILU factorization of its coefficient matrix [16]. The construction of the preconditioner was divided into the three steps: (i) matrix preprocessing, (ii) partial inverse-based ILU factorization, and (iii) recursive application of the first two steps to the approximate Schur complement.

**Preprocessing.** To equilibrate the magnitude of the elements of the coefficient matrix, first, its rows, and then, its columns were scaled in the infinity vector norm. The scaled coefficient matrix was reordered by a ddPQ strategy [72]. The reordering was performed to increase the diagonal dominance of the scaled coefficient matrix and the sparsity of its partial ILU factorization. The preprocessed coefficient matrix is given as

$$\tilde{A} = P^T \cdot D_1 \cdot A \cdot D_2 \cdot Q. \quad (3.51)$$

where $D_1, D_2 \in \mathbb{R}^{N \times N}$ are the diagonal matrices of the scaling factors and $P, Q \in \mathbb{R}^{N \times N}$ are the permutation matrices.

**Partial ILU Factorization.** Partial ILU factorization implies that ILU factorization was not performed until completion but was stopped at some intermediate step. The Crout algorithm was adopted to perform partial ILU factorization. At the $m$-th step of the Crout algorithm, the elements $(\tilde{A})_{i,j} \in \mathbb{R}$ (resp. $m$-th column $L_i$) and $(\tilde{A})_{m,j} \in \mathbb{R}$ (resp. $m$-th diagonal element of $D$ and $m$-th row of $U$) are computed using the previously computed rows and columns. Suppose that at the $m$-th step of the Crout algorithm, the following block structure is obtained:

$$\tilde{A} = \begin{bmatrix} B & F \\ E & C \end{bmatrix} = \begin{bmatrix} \tilde{L}_m \tilde{D}_m \tilde{U}_m + \tilde{E}_m \end{bmatrix}, \quad (3.52)$$

where

$$\tilde{L}_m = \begin{bmatrix} L_b & 0 \\ E & 1 \end{bmatrix}, \quad \tilde{D}_m = \begin{bmatrix} \tilde{D}_b & 0 \\ 0 & S \end{bmatrix}, \quad \tilde{U}_m = \begin{bmatrix} \tilde{U}_b & \tilde{U}_r \\ 0 & 1 \end{bmatrix}. \quad (3.53)$$

In (3.53), $\tilde{L}_b, \tilde{U}_b \in \mathbb{R}^{m \times m}$ are unit lower triangular matrices; $\tilde{D}_b \in \mathbb{R}^{m \times m}$ is a diagonal matrix; and $S \in \mathbb{R}^{(N-m) \times m \times m}$ is an approximate Schur complement. To limit the increase in the magnitude of the inverse triangular factors, an inverse-based pivoting approach was employed. For a schematic illustration of the factor-and-skip strategy, see Fig. 3.2. At the $m$-th step of the Crout algorithm, the inverses of $\tilde{L}_m$ and $\tilde{U}_m$ were estimated in the infinity norm using a modified condition number estimator for lower triangular matrices. The factorization was continued if these estimates were equal to or smaller than a prescribed bound, $\kappa$, i.e.,

$$\|\tilde{L}_m^{-1}\|_\infty \cdot \|\tilde{U}_m^{-1}\|_\infty \leq \kappa. \quad (3.54)$$

Otherwise, the $m$-th row and column of $\tilde{A}$ were permuted to its $N$-th row and column, respectively. After all the elements of $A$ had either been factorized or skipped, the partial factorization was stopped. During the course of the Crout algorithm, an approximation to the Schur complement was determined from the postponed updates using $S = C - L_b \cdot D_b \cdot U_r$.

To successfully control the magnitude of the inverse triangular factors during the factorization, it is necessary to apply a dropping strategy that is also inverse-based. At the $m$-th step of the Crout algorithm, any $m$-th element of $\tilde{L}_m$ and any $m$-th element of $U_r (i,j > m)$ was dropped if

$$\|\tilde{L}_m^{-1}_{ii}\|_\infty \cdot \|\tilde{U}_m^{-1}_{ij}\|_\infty < \frac{\tau}{\kappa^2}. \quad (3.55)$$

where $\tau$ denotes some threshold tolerance. A similar rule was applied to drop elements of $S$. Theoretical results state that by performing the dropping according to these inverse-based
rules, the perturbation of the preconditioned coefficient matrix with respect to the identity matrix is bounded by a constant time $\tau$. However, it is often the case in practice that many more elements can be safely dropped. A good compromise is to perform the dropping with respect to $\tau$ divided by the maximum of the estimated norms, $\kappa = \max(\|L^{-1}\|_{\infty}, \|U^{-T}\|_{\infty})$ [15].

Recursive Application. The multilevel framework of the ILU preconditioner was generated by recursively applying the first two steps, i.e., matrix preprocessing and partial ILU factorization, to the approximate Schur complement. Postponing the factorization to the next level may be necessary because the prescribed bound may otherwise be exceeded. With an increase in the factorization level, the Schur complement becomes increasingly dense. The recursive procedure was stopped as soon as it was no longer computationally profitable to postpone the factorization to the next level. The multilevel ILU factorization was completed by performing a full LU factorization of the remaining Schur complement.

Multilevel ILU Preconditioning. The application of ILU factorization as a preconditioner of some Krylov subspace method requires solutions of linear systems that have the result of ILU factorization as a coefficient matrix. Because of the multilevel structure of the inverse-based ILU preconditioner, these linear systems were solved level-wise by block forward and subsequent backward substitutions.

### 3.3. Validation

To validate the numerical method, we solved the benchmark problem for the UCM model and compared the solution obtained with the SFEM solution [13]. The Deborah number was set to $De = 7.24$. The cylinder system was configured using the parameter values $\varepsilon = 0.4$ and $\mu = 0.1$. The total number of collocation points corresponded to $CP_I = (M + 1) \times 2N = 55 \times 110$. The parameter values used in the inverse-based ILU preconditioned Newton-GMRES method are listed in Tab. 3.1. The Newton method must be started from some initial estimate. The initial estimate for the modified stream function field and the flux between the cylindrical boundaries was taken as the analytical solution for the Stokes flow [2]. The initial estimate for the modified conformation tensor field was taken as its analytical solution in the equilibrium state of rest. For the Newton method to converge at the parameter values of interest, a zero-order continuation procedure was applied. Every time the Newton method converged, the value of $De$ was incremented slightly. The solution of the previous continuation step was used as the initial estimate for the next continuation step. The starting vector for the GMRES method was a zero vector. The maximum number of allowed GMRES iterations was restricted to 200. The multilevel inverse-based ILU preconditioner was recalculated at every alternate Newton iteration.

Table 3.1: Parameters and their assigned values for the inverse-based ILU preconditioned Newton-GMRES method.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Newton method:</td>
<td></td>
</tr>
<tr>
<td>$\alpha$</td>
<td>$10^{-4}$</td>
</tr>
<tr>
<td>$\theta_{\min}$</td>
<td>$10^{-1}$</td>
</tr>
<tr>
<td>$\theta_{\max}$</td>
<td>$5 \times 10^{-1}$</td>
</tr>
<tr>
<td>$W$</td>
<td>1</td>
</tr>
<tr>
<td>$\tau$</td>
<td>$10^{-5}$</td>
</tr>
<tr>
<td>$\kappa$</td>
<td>$10^{-8}$</td>
</tr>
<tr>
<td>GMRES method:</td>
<td></td>
</tr>
<tr>
<td>$\eta$</td>
<td>$10^{-8}$</td>
</tr>
<tr>
<td>Inverse-based ILU preconditioner:</td>
<td></td>
</tr>
<tr>
<td>$\tau$</td>
<td>$10^{-5}$</td>
</tr>
<tr>
<td>$\kappa$</td>
<td>$10^{2}$</td>
</tr>
</tbody>
</table>

For ease of comparison with the SFEM solution, we present the results of the validation in the modified coordinate system $(\zeta, \theta)$ [13, 12]. Figure 3.3 shows profiles of the azimuthal normal stress at two different azimuthal positions, $\theta = 0$ and $\pi/2$. We observe that our solution agrees excellently with the SFEM solution; this excellent agreement is also apparent in regions where strong variations occur.
3. Eccentric Taylor-Couette Flow of Polymeric Solutions and Melts

3.4. Sample Calculations and Interpretation

The benchmark problem was also solved for the power-law EWM model. Two different values were used for $k$, namely, $k = -0.7$ (fluid 1) and $-2$ (fluid 2). The values of $De_0 = \lambda_0 V / R_1$ ranged between 1 and 5. A cylinder system with a large eccentricity ratio ($\varepsilon = 0.8$; $\mu = 1$) was chosen to allow flow recirculation to occur. The parameter values used in the preconditioned Newton-GMRES method were the same as those used in the validation. A zero-order parameter continuation procedure in $k$ and $De_0$ was applied to obtain their desired values. The total number of collocation points again corresponded to $CP_1 = (M + 1) \times 2N = 55 \times 110$.

To check the convergence of the simulations, we repeated the simulation for the set of model parameter values predicting the largest Deborah number in the eccentric annular flow field ($k = -0.7$; $De_0 = 5$). A total of $CP_2 = 50 \times 100$ and $CP_3 = 60 \times 120$ collocation points were used. For each grid of collocation points, the physical quantity $I \cdot \xi_1 / 3$ was computed at 100 different radial positions that were equally spaced across the narrowest part of the gap ($\theta = 0$). The relative difference between the solution computed on grid $CP_1$ and those computed on grids $CP_2$ and $CP_3$ was never larger than $10^{-4}$.

3.4.1. Fluid 1

In this subsection, we present the flow profiles for fixed $k = -0.7$ and varying $De_0$. For this $k$ value, the power-law EWM model predicts a mildly shear-thinning behavior.

Figure 3.4.1 shows the contour profile of the stream function for two different $De_0$ values: $De_0 = 1$ (Fig. 3.4.1(a)) and 5 (Fig. 3.4.1(b)). For both $De_0$ values, the stream function has the largest magnitude on the inner cylinder and a magnitude of zero on the outer one. Additionally, there exists a contour line where the stream function is zero. This contour line indicates the location where the flow is separated into a main flow around the inner cylinder and a recirculating flow in the large-gap region. For the larger $De_0$ value, the region of flow recirculation is smaller. The maximum intensity of recirculation, measured by the extremal value of the stream function in the center of flow recirculation, is lower. The recirculation center is shifted downstream (i.e., to larger $\theta$ values).

Figures 3.5 and 3.6 show profiles for three different $De_0$ values: $De_0 = 1$ (solid line), $De_0 = 3$ (dashed line), and $De_0 = 5$ (dotted line). Henceforth, we use the normalized variable $\xi = \xi (\xi - \xi_2) / (\xi_1 - \xi_2)$ to indicate the relative radial position within the flow field. Figure 3.5 shows profiles of the nontrivial components of the extra stress tensor (Fig. 3.5(a)-(c)) and the normalized first invariant of the conformation tensor (Fig. 3.5(d)) at a relative radial position $\xi = 0.3$. The first invariant of the conformation tensor has been normalized by the value
of this quantity in the equilibrium state of rest. The radial normal stress (Fig. 3.5(a)) is much smaller in magnitude than the other nontrivial components of the extra stress tensor. It exhibits two extrema that are caused by planar elongational flow. As opposed to the predictions for the Stokes flow, the extrema are not of equal magnitude. The maximum is larger in magnitude than the minimum. Furthermore, we observe that the maximum increases more strongly than the minimum as $D_e^0$ is increased. The asymmetry in the radial normal stress field and the sharp gradients in the diverging zone of the gap ($\theta/2\pi < 0.5$) are caused by the memory of the fluid. Large shear stresses (Fig. 3.5(b)) are restricted to the region of the main flow. Contrary to the predictions for the Stokes flow, the maximum shear stress is not at the narrowest part of the gap, but is pushed considerably into the diverging zone of the gap. Moreover, we find that the maximum of the shear stress increases strongly with $D_e^0$. The azimuthal normal stress (Fig. 3.5(c)) exhibits pronounced elastic effects. The maximum of the azimuthal normal stress is more than twenty times larger than that of the radial normal stress. It is slightly shifted into the converging zone of the gap ($\theta/2\pi > 0.5$). Note that there are two different types of shear acting on the fluid: the Taylor-Couette shear produced by the rotation of the inner cylinder and the Poiseuille shear produced by the azimuthal variation of the pressure gradient. As verified later, the latter strongly dominates at some locations in the small-gap region and causes pronounced macroscopic effects there. It should be noted that the conformation tensor incorporates information about the average deformation, extension, and orientation of the polymeric constituents of the fluid. The first invariant of the conformation tensor is a direct measure of the average extension of the polymers during flow. We observe that it behaves in a manner that is qualitatively similar to the azimuthal normal stress. Consequently, large polymer extension is restricted to the region of the main flow. In the region of flow recirculation, the polymers are relaxed and are almost in the equilibrium state of rest.
Figure 3.6 shows radial profiles of the shear stress (Fig. 3.6(a)) and the normalized first invariant of the conformation tensor (Fig. 3.6(b)) at the narrowest part of the gap. For $De_0 = 1$, the shear stress has a maximum on the outer cylinder. It decreases monotonically from the outer to the inner cylinder. For $De_0 = 3$ and 5, the shear stress also has a maximum on the outer cylinder. Contrarily, it decreases and then increases on approaching the cylinder. Additionally, we observe that the shear stress is larger for larger $De_0$ values. The analytical solution for the Stokes flow predicts that the shear stress is largest on the outer cylinder and that it decreases monotonically with an increase in $\xi^*$. Therefore, the maximum value of the shear stress on the outer cylinder cannot be due to the elasticity of the fluid. It must be due to the dominating Poiseuille shear flow. It is interesting to note that for $De_0 = 3$ and 5, the shear stress assumes values in the diverging zone of the gap (see Fig. 3.5(b)) that are larger than the shear stress on the outer cylinder at the narrowest part of the gap. Therefore, we relate the increase in the shear stress for larger $De_0$ values near the inner cylinder to its strong downstream convection. For $De_0 = 1$, the normalized first invariant of the conformation tensor is largest on the outer cylinder and decreases monotonically from the outer toward the inner cylinder. For $De_0 = 3$ and 5, the normalized first invariant of the conformation tensor is also largest on the outer cylinder. It also decreases in the vicinity of both cylinders as the value of $\xi^*$ is increased. Contrarily, there is a region of strong variation between the two cylinders; we attribute this to large differences in the local residence time of the fluid. Additionally, we find that the normalized first invariant of the conformation tensor is larger for larger $De_0$ values.

3.4.2. Fluid 2

In this subsection, we present the flow profiles for fixed $k = -2$ and varying $De_0$. Note that the power-law EWM model predicts a more pronounced shear-thinning behavior for $k = -2$ than for $k = -0.7$.

Figure 3.7 shows contour profiles of the stream function for two different $De_0$ values: $De_0 = 1$ (Fig. 3.7(a)) and 5 (Fig. 3.7(b)). The trends in the case of increasing $De_0$ are qualitatively similar to those observed for $k = -0.7$ (see Fig. 3.4.1). The shrinkage of the recirculation region, however, is greater. The maximum intensity of recirculation is lower. The recirculation center is pushed slightly less downstream. In the region of the main flow, the Deborah number is of the same magnitude for the set of parameter values $k = -2$ and $De_0 = 5$ as for the set of parameter values $k = -0.7$ and $De_0 = 1$. From this information, it is possible to distinguish whether each of these effects is caused by a decrease in memory and/or by a higher degree of shear-thinning. We find that it is the memory of the fluid that promotes the downstream shift of the recirculation center. Furthermore, we find that the shear-thinning character of the fluid promotes the other effects to a greater extent.

Figures 3.8 and 3.9 show profiles for three different $De_0$ values: $De_0 = 1$ (solid line), $De_0 = 3$ (dashed line), and $De_0 = 5$ (dotted line). Figure 3.8 shows profiles of the nontrivial components of the extra stress tensor (Fig. 3.8(a)–(c)) and the normalized first invariant of the conformation tensor (Fig. 3.8(d)) at a relative radial position $\xi^* = 0.3$. These profiles are qualitatively similar to those obtained for $k = -0.7$ (see Fig. 3.5). However, they are flatter and the magnitude of the plotted quantities is smaller. Whereas the flattening of the profiles is only due to the higher degree of shear thinning, the decrease in the magnitude of the plotted quantities is also partially due to a reduced relaxation time. Moreover, the maximum of the radial...
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and azimuthal normal stress components is shifted to larger and smaller $\theta$ values, respectively. Because the size of the recirculation zone differs considerably from that for $k = -0.7$, it is reasonable that the location of the maxima in the normal stress components must be shifted.

Figure 3.8: Azimuthal profiles calculated for power-law EWM model at $\xi^* = 0.3$: (a) radial normal stress, (b) shear stress, (c) azimuthal normal stress, and (d) normalized first invariant of conformation tensor. The model parameters are $k = -2$ and $D_0 = 1$ (solid line), 3 (dashed line), and 5 (dotted line).

Figure 3.9 shows radial profiles of the shear stress (Fig. 3.9(a)) and the normalized first invariant of the conformation tensor (Fig. 3.9(b)) at the narrowest part of the gap. Again, we observe that these profiles are flatter and that the magnitude of the plotted quantities is smaller for $k = -2$ than for $k = -0.7$ (see Fig. 3.6). The profiles are qualitatively similar for $D_0 = 1$. Contrarily, pronounced qualitative differences are found for $D_0 = 3$ and 5. We observe that the shear stress does not increase near the inner cylinder as $\xi^*$ is increased. Interestingly, the maximum of the shear stress in the diverging zone of the gap (see Fig. 3.8(b)) is also smaller than the shear stress on the outer cylinder in the narrowest part of the gap. Furthermore, the normalized first invariant of the conformation tensor has no pronounced local extrema between the cylinders. Clearly, these qualitative differences are due to a reduced relaxation time.

Figure 3.9: Radial profiles calculated for power-law EWM model at $\theta = 0$: (a) shear stress and (b) normalized first invariant of conformation tensor. The model parameters are the same as those given in the caption of Fig. 3.8.

3.5. Conclusions

In this work, we presented a new numerical approach to solve differential-type viscoelastic fluid models for a steady Taylor-Couette flow between eccentric cylinders. This numerical approach is special in that the nonlinear system of discretized algebraic flow equations is solved iteratively using an inverse-based ILU preconditioned Newton-Krylov method. The numerical approach has been validated by solving the benchmark problem for the UCM model at a large Deborah number. The successful calculation and the excellent agreement of the results with numerical data from literature encourage the application of the inverse-based ILU preconditioned Newton-Krylov method to more difficult viscoelastic fluid flow problems.

In addition, we performed a parameter study for the power-law EWM model. A large eccentricity ratio was chosen for the cylinder system to allow flow recirculation to occur. Several interesting phenomena that are caused by the large eccentricity ratio of the cylinder system as well as by the viscoelastic nature of the fluid were detected. The large eccentricity ratio of the cylinder system causes flow separation. Furthermore, it causes pronounced effects in the small-gap region. Interestingly, large shear and azimuthal normal stresses as well as large polymer extension are found in the upper middle part of the gap near the stationary outer cylinder. Pronounced elongation effects in the radial shear stress occur in the medium-gap region. The convection caused by the memory of the fluid destroys the symmetric structure
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of the flow profiles. From the nontrivial components of the extra stress tensor, the shear stress is most severely affected by the downstream convection. The memory of the fluids causes large gradients in the shear stress at positions located widely within the diverging zone of the gap. Furthermore, it promotes an increase in the shear stress in the vicinity of the rotating inner cylinder in the very-small-gap region. The shear-thinning nature of the viscoelastic fluid flattens the profiles of the nontrivial components of the extra stress tensor and of the polymer extension. The shear-thinning nature of the fluid decreases the magnitudes of these physical quantities, whereas the memory increases them. These interesting phenomena encourage further investigation of other polymeric fluids containing a more complex microstructure in an eccentric annular flow field.

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The predictions of the EWM model confirmed that the rheology and microstructure of complex fluids greatly influence each other in a steady eccentric Taylor-Couette flow. Therefore, we proceed with the investigation of a polymeric fluid having a more complex microstructure. In this chapter, we study a steady Taylor-Couette flow of an immiscible polymer blend with droplet morphology. Because the eccentric cylinder system has a capability for highly distributive and dispersive mixing, we believe that it is ideally suited to investigate the dynamics of multiple droplets, including breakup and coalescence.

For investigating the dynamics of the polymer blend, we adopted the EDE model. Compared to the EWM model, this model has more internal variables, not all of which are unconstrained. Furthermore, the degree of coupling between the state variables is significantly higher in the flow equations. Thus far, it has been solved only for non-benchmark flows [31, 21]. In these contributions, the flow problem was formulated as a BVP of ordinary differential equations, which was then solved using a shooting algorithm with adaptive step-size control. In the case of a real benchmark flow, the governing flow equations cannot anymore be reduced to ordinary differential equations. Consequently, more sophisticated numerical solution strategies must be adopted.

The remainder of this chapter is organized as follows. In Sec. 4.1, we introduce the dimensionless flow equations according to the EDE model. To solve them for a steady eccentric Taylor-Couette flow, we modified the numerical approach presented in the previous chapter. The modifications that were made are described in Sec. 4.2. Sample calculations are presented in Sec. 4.3. Conclusions drawn are presented in Sec. 4.4.

4.1. Dimensionless Flow Equations

The EDE model comprises some time evolution equations for the velocity vector \( \mathbf{v} \), the conformation tensor \( C \), the droplet shape tensor \( S \), the droplet number density \( n \), and an explicit
expression of the extra stress tensor, $\sigma$. Since the flow is assumed to be isothermal and incompressible, the scalar mass density is a constant and thus does not belong to the set of state variables. To obtain a complete set of flow equations, the divergence-free condition for the velocity vector must be additionally imposed when a specific flow problem is formulated. To render the flow equations dimensionless, we use the scalings

$$V = \sqrt{G \rho T}, \quad V = \sqrt{G \rho T}, \quad \nabla = \sqrt{G \rho T} \nabla, \quad S = \bar{n}, \quad n = n_{m}, \text{and} \quad \nabla = \sqrt{G \rho T} \nabla. \quad A \text{Deborah number consistent with these scalings is defined as } \text{De} = \sqrt{\text{De}_{\text{c}} \text{De}_{\text{o}}}. A \text{Deborah number consistent with these scalings is defined as } \text{De} = \sqrt{\text{De}_{\text{c}} \text{De}_{\text{o}}}.$$  

Under steady-state conditions, the complete set of dimensionless flow equations becomes

$$\nabla \cdot \vec{v} = 0, \quad (4.1)$$

$$- \nabla p + \nabla \cdot \vec{\sigma} = 0. \quad (4.2)$$

$$\text{De}_{\text{c}} \left(- \nabla \cdot \nabla \bar{S} - \frac{2}{3} \mathbf{I} \cdot \bar{S} + \mathbf{I} \cdot \bar{S} \right) - (1 - \phi) \left(\bar{C} - \mathbf{1}\right)$$

$$+ \frac{1}{3} \phi \frac{\Gamma}{G} \ln \bar{n} \nu \left(\frac{1}{3} \bar{S} - \frac{2}{3} r_{C} \mathbf{I}\right)$$

$$\times \left(1 + p^{*} \phi \left[- \left(1 - \frac{2}{3} \nu \rho T \right) \mathbf{I} \cdot \bar{S} - \frac{3}{2} r_{C} \mathbf{I}\right] - \frac{1}{3} \ln \bar{n} \nu \left(\frac{1}{3} \bar{S} - \frac{2}{3} r_{C} \mathbf{I}\right) \right) = 0, \quad (4.3)$$

$$\text{De}_{\text{o}} \left(- \nabla \cdot \nabla \bar{S} - \frac{2}{3} \mathbf{I} \cdot \bar{S} + \mathbf{I} \cdot \bar{S} \right)$$

$$- (1 + p^{*} \phi \left[- \left(1 - \frac{2}{3} \nu \rho T \right) \mathbf{I} \cdot \bar{S} - \frac{3}{2} r_{C} \mathbf{I}\right] + p^{*} \phi \left[- \left(1 - \frac{2}{3} \nu \rho T \right) \mathbf{I} \cdot \bar{S} - \frac{3}{2} r_{C} \mathbf{I}\right]$$

$$\times \left(\bar{C} + \bar{S} + \bar{S} \cdot \mathbf{I} - \frac{2}{3} \bar{S} \cdot \mathbf{I}\right) = 0, \quad (4.4)$$

$$\text{De}_{\text{c}} \nabla \vec{v} + \phi f_{h}, \quad \left(\bar{F}^{2} - \frac{1}{3} \pi \mathbf{I}^{2}\right) = 0, \quad (4.5)$$

where

$$\bar{\sigma} = (1 - \phi) \frac{\Gamma}{T} (\bar{C} - \mathbf{1})$$

$$+ \phi \frac{\Gamma}{G} \left[n \nu \left(\frac{1}{3} \bar{S} - \frac{2}{3} r_{C} \mathbf{I}\right) - \frac{1}{3} \ln \left(\frac{1}{3} \bar{S} + \frac{1}{3} r_{C} \mathbf{I}\right)\right]. \quad (4.6)$$

Equation (4.1) is the continuity equation for an incompressible medium. Equation (4.2) is the Cauchy momentum balance equation where the convective term has been neglected. Equation (4.3) is the conformation tensor equation describing the dynamics of the polymeric constituents of the matrix fluid. The dynamics of the shape and number density of the dispersed Newtonian droplets are described in (4.4) and (4.5), respectively. Equation (4.6) is the explicit expression for the extra stress tensor.

### 4.2. Numerical Methodology

In this subsection, we describe the numerical methodology that was used to solve dimensionless flow equations introduced in Sec. 4.1 for the flow in the benchmark geometry introduced in Sec. 3.1.2.

The flow conditions were the same as those described in Sects. 3.1.2 and 3.1.3. Because the numerical methodology is similar to that presented in the previous chapter, we restrict our focus to how it was modified.

The benchmark problem was also formulated in bipolar cylindrical coordinates using a modified stream function formulation. As the primary variables of the BVP, we selected a modified stream function $\bar{\psi}^*$, modified conformation tensor $\bar{C}^*$, modified droplet shape tensor $\bar{S}^*$, and droplet number density $\bar{n}$. The modified quantities from the set of primary variables are defined as

$$\bar{\psi}^* = \bar{\psi}_{X}, \quad \bar{C}^* = \bar{C}_{/X}, \quad \bar{S}^* = \bar{S}_{/X}. \quad (4.7)$$

If the extra stress tensor appearing in the Cauchy momentum balance equation (4.2) is expressed in terms of the primary variables, the subsequent elimination of pressure would result in a large number of terms. To avoid such an increase in the number of terms, a modified extra stress tensor $\bar{\sigma}^*$ was introduced as an intermediate primary variable as follows:

$$\bar{\sigma}^* = \bar{\sigma} / \bar{X}. \quad (4.8)$$

The extra stress tensor appearing in (4.2) was substituted by the intermediate primary variable. Considering the curl of the obtained equation gives

$$\left(\frac{\partial^2}{\partial \theta^2} + \frac{\partial^2}{\partial r^2} + 1\right) \bar{\sigma}_{\theta\theta} + \frac{\partial^2}{\partial \theta r} (\bar{\sigma}_{\theta r} - \bar{\sigma}_{r\theta}) = 0. \quad (4.9)$$

Adopting the procedure described in Sec. 3.1.3, we obtain the following integral equation that ensures the $2\pi$-periodicity of the scalar pressure in the $\theta$-direction:

$$\int_{0}^{2\pi} \left(\frac{\partial^2 \bar{\psi}_{\theta}}{\partial \theta \partial \xi} + \frac{\partial \bar{\psi}_{\theta}}{\partial \theta} \right) \bar{X} - \bar{\sigma}_{\theta r} \sin \theta + \bar{\sigma}_{\theta \theta} \sinh \xi \right) d\theta = 0. \quad (4.10)$$
The structural equations (4.3)–(4.5) and the equation for the extra stress tensor (4.6) were expressed in terms of the primary variables. These reformulated equations, the conservation balance equation (4.9), and the pressure periodicity condition (4.10), together with the boundary conditions (3.20) and (3.21) and the 2π-periodicity condition of the primary variables in the θ-direction, form a BVP. The pseudospectral collocation method described in Sec. 3.1.4 was used for the discretization of the BVP. The nonlinear system of discretized algebraic flow equations was solved iteratively using the preconditioned Newton-Krylov method introduced in Sec. 3.2. The elements of the Jacobian matrix originating from the partial derivatives of (4.9) and (4.10) with respect to the primary variables were determined numerically using a first-order finite difference approximation. In order to increase the computational efficiency of the numerical simulations, the following two measures were considered. (i) The computations of the nonlinear vector function and its Jacobian were parallelized using the software package OpenMP [62]. (ii) The Jacobian matrix and the inverse-based ILU preconditioner were recalculated at the first Newton iteration of every new continuation step.

### 4.3. Results and Discussion

Like in earlier numerical studies [31, 21], we assumed that the deformation of droplets into prolate or oblate configurations depends on the viscous properties of the blend components and we used the relationship $p^* = −η/\nu C_0$ for parameter $p^*$. Because we are interested in investigating the influence of the rheology of the matrix fluid on the droplet characteristics, we selected the values of the model parameters such that the macroscopic response of the blend is governed by the matrix fluid. The rheology of the matrix fluid was varied using three different values of $k$, namely, $k = −2$, $−2.5$, and $−3$, and $De_{CJ} = \lambda_{CJ}/R_1^2$ values ranging between 0.8 and 3. The values of the other model parameters were kept fixed at $\phi = 0.1$, $p^* = −4$, $\Gamma/\nu = 0.01$, $De_\nu = 3$, $De_c = 3$ and $f_{bc} = 1$, and $\theta = 0.001$. A cylinder system with a large eccentricity ratio ($ε = 0.8; \mu = 1$) was chosen to enable flow recirculation. The parameter values used in the preconditioned Newton-GMRES method were identical to those listed in Tab. 3.1. The initial estimates for the modified stream function field and flux between the cylindrical boundaries were taken as the analytical solution for the Stokes flow. The initial estimate for the other primary variables was taken as the analytical solution in the equilibrium state of rest. A zero-order parameter continuation procedure with $k$ and $De_{CJ}$ was applied to obtain their desired values. The starting vector for the GMRES method was a zero vector. The maximum number of GMRES iterations was restricted to 500. Further, to smoothen the sample calculations in the radial and azimuthal directions, a fourth-order exponential filter [44] was adopted. The total number of collocation points corresponded to $CP_1 = (M + 1) \times 2N = 55 \times 110$.

In order to verify the convergence of the simulations, we repeated the simulations for the set of model parameter values used to predict the largest Deborah number in the eccentric annular flow field ($k = −2; De_{CJ} = 3$). A total of $CP_2 = 50 \times 100$ and $CP_3 = 60 \times 120$ collocation points were used. For each grid of collocation points, the microstructural blend characteristics $\hat{\rho}^2/3$ with $\epsilon \in \{C, S\}$ and $\bar{n}$ were computed at 100 different radial positions that were equally spaced across the smallest part of the gap. The relative difference between the solution computed on grid $CP_1$ and the solutions computed on grids $CP_2$ and $CP_3$ was never larger than $4 \times 10^{-4}$.

#### 4.3.1. Qualitative Blend Behavior

In this subsection, we describe the qualitative blend behavior obtained for the set of model parameter values used to predict the largest Deborah number in the eccentric annular flow field ($k = −2; De_{CJ} = 3$). We begin by presenting macroscopic blend characteristics and then proceed to microstructural blend characteristics.

Figure 4.1 shows the contour profiles of macroscopic blend characteristics. Figure 4.1(a) shows the contours of the stream function. The profile has a highly symmetric structure. The stream function has the largest magnitude on the inner cylinder and a magnitude of zero on the outer one. In addition, there exists a contour line wherein the stream function is zero. This contour line indicates the location wherein the flow is divided into a main flow around the inner cylinder and a recirculating flow in the large-gap region. The contour profiles shown in Figs. 4.1(b)–(d) have become asymmetrical due to the memory of the matrix fluid. Figure 4.1(b) shows the contours of the shear stress. Large values of the shear stress are restricted to the region of the main flow around the inner cylinder. The Taylor-Couette shear induces a large shear stress near the inner cylinder in the medium- and large-gap regions. In the small-gap region, the Poiseuille shear dominates in the upper middle part of the gap near the outer cylinder. Moreover, the shear stress is strongly downstream convected by the matrix fluid memory. The contours of the first normal stress difference $\tilde{N}_1 = \tilde{\sigma}_{\theta\theta} − \tilde{\sigma}_{\theta\nu}$ are shown in Fig. 4.1(c). Simple shear causes large first normal stresses near the inner cylinder and across the entire gap in the small-gap region. In the region of flow recirculation, the first normal stress has small magnitudes. Negative magnitudes occur in the region of flow recirculation because the components of the extra stress tensor are represented in the bipolar cylindrical coordinate system and has no special physical significance. The contours of the second normal stress difference $\tilde{N}_2 = \tilde{\sigma}_{\nu\nu} − \tilde{\sigma}_{\theta\nu}$ are shown in Fig. 4.1(d). Planar extension induces significant effects on the small- and medium-gap regions. Furthermore, the memory of the matrix fluid causes the extremum in the diverging zone to be more than twice that in the converging zone of the gap.
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Figure 4.1: Contour profiles of macroscopic blend characteristics calculated for EDE model: (a) stream function, (b) shear stress, (c) first normal stress difference, and (d) second normal stress difference. The model parameters are $k = -2$, $D_{ECB} = 3$, $D_{CS} = 3$, $\phi = 0.1$, $p^* = -4$, $\Gamma/G = 0.01$, $f_{wc} = 1$, and $\theta = 0.001$.

4.3. Results and Discussion

Figure 4.2 shows the contour profiles of the microstructural blend characteristics. Figures 4.2(a) and (b) show the contours of the first invariants of the conformation and droplet shape tensors, respectively, normalized by their values in the equilibrium state of rest. Note that the first invariants of the conformation and droplet shape tensors are direct measures of the average extension of the polymeric constituents of the matrix fluid and dispersed Newtonian droplets, respectively. In the regime of the main flow, the contour profile of the normalized first invariant of the conformation tensor is qualitatively similar to that of the first normal stress difference; this implies that the extension of the polymeric constituents of the matrix fluid causes the first normal stress effects in the region of the main flow. The small absolute values of the first normal stress difference in the flow recirculation region (see Fig. 4.1(c)) can be explained by the fact that the polymeric constituents are relaxed and almost in the equilibrium state of rest. In the region of the main flow, the dynamics of the dispersed Newtonian droplets is qualitatively different from that of the polymeric constituents of the matrix fluid. This observation is not astonishing, since the dynamics of the former is governed by two additional mechanisms: breakup and coalescence. Interestingly, an isolated zone of large droplets is present near the outer cylinder in the narrow section of the gap. The droplet size in this zone has a magnitude that can only be found very close to the inner cylinder. Figure 4.2(c) shows the contours of the droplet number density; the largest values occur near the inner cylinder. The upper middle part of the gap in the medium-gap region contains a zone of increased droplet number density. Note that this delay in the droplet breakup is related to interfacial elasticity and also occurs if the matrix fluid is purely Newtonian [41]. The residence time of the droplets is not sufficiently large near the outer cylinder in the narrow section of the gap, and therefore, pronounced droplet breakup cannot occur there. Yet, owing to a reduced local contribution of the system dynamics to droplet break-up, it is also likely that the increased droplet number density in the diverging zone of the gap is lower than the average droplet number density close to the inner cylinder.

4.3.2. Tailoring of Droplet Characteristics

In the previous subsection, we had described the qualitative behavior exhibited by an immiscible blend of a shear-thinning viscoelastic matrix fluid and Newtonian droplets in a highly eccentric annular flow field. From an industrial perspective, it is desirable to understand how to tailor droplet characteristics. One possible strategy is to modify the rheology of the polymeric matrix fluid.

To examine the feasibility of this strategy, we plotted the droplet characteristics at a single highly simple shear-dominated location within the flow field, namely, on the outer cylinder in the narrowest section of the gap. Figures 4.3(a) and (b) show the normalized first invariant of the droplet shape tensor and the droplet number density, respectively, as a function of $D_{EC}$ at three different negative $k$ values. As $D_{EC}$ increases, the droplet size decreases and the droplet
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Figure 4.2.: Contour profiles of microstructural characteristics calculated for EDE model: (a) normalized first invariant of conformation tensor, (b) normalized first invariant of droplet shape tensor, and (c) droplet number density. The model parameters are identical to those given in the caption of Fig. 4.1.

4.4. Conclusions

To our knowledge, this work is the first computational investigation of the dynamics of multiple droplets dispersed in a polymeric matrix fluid in a steady eccentric Taylor-Couette flow field. We chose a large eccentricity ratio for the cylinder system in order to allow flow recirculation to occur. Furthermore, the macroscopic responses of the polymer blend were assumed to be governed by the dynamics of the polymeric matrix fluid. To describe the dynamics of the polymer blend, we adopted a differential-type constitutive model previously developed by other researchers using the generalized bracket approach. The benchmark problem was successfully solved using a modified version of the preconditioned Newton-Krylov method presented in the previous chapter.

number density increases. In this parameter study, the characteristic time scale of the matrix relaxation is smaller than the times of the interfacial relaxation, droplet breakup, and droplet coalescence. Our results confirm that the matrix relaxation can strongly impact the droplet dynamics under these conditions. Moreover, the droplet size decreases only very slightly whereas the droplet number density significantly increases with $k$. Such large variations in the droplet number density are due to the significant impacts of the different shear-thinning behaviors of the matrix fluid.

Figure 4.3.: Droplet characteristics calculated for EDE model as a function of $De_C$ for three different $k$ values, namely, $k = -2$ (squares), $-2.5$ (circles), and $-3$ (triangles): (a) normalized first invariant of droplet shape tensor and (b) droplet number density. The other model parameters are the same as those given in the caption of Fig. 4.1.
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The droplet characteristics were found to be strongly influenced by two different types of simple shear flows: the Taylor-Couette shear that resulted in a high number density of moderately sized droplets near the inner cylinder and the Poiseuille shear that formed an isolated zone of pronounced droplet extensions in a narrow section of the gap near the outer cylinder. Interestingly, the droplet number density was only moderate in this narrow section and enhanced droplet breakups were delayed until they widened in the diverging zone of the gap. In the flow recirculation region, the droplets were relaxed and almost in their equilibrium state of rest. In addition, the rheology of the matrix fluid was found to have large quantitative impacts on the droplet characteristics. The results suggest that modifying the rheology of the matrix of products is a feasible strategy to tailor their droplet characteristics to consumer expectations. It is important to note that because the droplet characteristics depend on many material parameters and flow conditions, our findings cannot be generalized. Further theoretical as well as experimental researches have to be conducted to develop a more complete understanding of the droplet dynamics of immiscible polymer blends under industrially relevant conditions.

5. Conclusions and Outlook

The objective of this dissertation was to gain a more fundamental understanding of the rheology and microstructural dynamics of polymeric solutions, melts, and blends with droplet morphology in a steady eccentric Taylor-Couette flow. The circular flow in the annular gap between eccentric cylinders is a complex flow composed of simple shear, planar elongation, and rigid body rotation. The relative importance of these different flow types can be adjusted by changing the configuration of the eccentric cylinder system. This particular benchmark problem is also attractive because its geometry can be described easily. Furthermore, exact analytical solutions exist for many fluid models in the limit of a vanishing eccentricity ratio, and thus, they can be used to verify the consistency of numerical codes. Moreover, the eccentric cylinder system is also of relevance for the industrial processing of complex fluids because of its capability for highly distributive and dispersive mixing.

In this dissertation, the rheology and microstructural dynamics of polymeric fluids were described by differential-type constitutive equations that other researchers had developed in the past using the generalized bracket approach. This approach is a suitable thermodynamic framework for modeling complex fluid systems having a microstructure. Models that have been developed using this approach have the advantage that they can be checked for thermodynamic admissibility. Microstructural information extracted from the model predictions can be guaranteed to be physically meaningful. If such models describe the dynamics of very complex systems, their applicability is often limited. With increasing number of state variables, the number of flow equations as well as the number of terms and the degree of coupling between the state variables increase in the flow equations. Numerical problems such as those due to the occurrence of multiple solutions or the limited amount of available computer arithmetic precision often prevent the solving of such type of models for benchmark flows, or if they can be solved only over a limited range of parameter values. To gain a more complete understanding of the dynamics of complex fluids, information from different length scales is necessary. This dissertation should encourage researchers to improve complex fluid models as well as develop novel ones in consideration of their subsequent application by using the generalized bracket approach.

A novel numerical approach was developed to perform benchmark simulations. The approach is unique in that the nonlinear system of discretized algebraic flow equations is solved iteratively using a Newton-Krylov method along with an inverse-based ILU preconditioner. The
key feature of this preconditioner is that the magnitude of perturbations in the inverse triangular factors is controlled during its construction. To validate the developed approach, the benchmark problem was solved for the upper-convected Maxwell model at a large Deborah number. The successful calculation of steep boundary layers and the excellent agreement of calculation results with numerical data from the literature encourages further application of the preconditioned Newton-Krylov method to complex viscoelastic fluid flow simulations. The scalability of the developed approach could be further improved by using a lower-order spatial discretization scheme and parallelizing the computation of the multilevel inverse-based ILU factorization. As has also been recognized by other researchers [39], the use of iterative linear solvers reduces the problem of over-solving of the linear Jacobian system.

The benchmark problem was also solved for other polymeric fluid models at different model parameter values. A large eccentricity ratio was selected for the cylinder system in order to allow flow recirculation to occur. Several interesting phenomena caused by the configuration of the cylinder system and the viscoelastic nature of the polymeric fluids were detected on different length scales. The calculations confirm that the eccentric annular benchmark problem is ideally suited to investigate effects of the Taylor-Couette and Poiseuille shear flows. It is important to realize that the Poiseuille shear is not present if the two cylinders of the cylinder system are positioned concentrically to each other. This benchmark flow is, however, only weakly extensional. Other benchmark flows such as contraction flows are better suited to study elongational effects. It should be noted that although rigid body rotation does not account for macroscopic effects, it plays an important role: it causes the polymeric constituents to rotate and thus allows flow separation to occur.

To conclude this dissertation, we gained a more fundamental understanding of the interrelationships between the rheology and the microstructural dynamics of polymeric fluids under industrially relevant flow conditions. To verify the model predictions, a comparison with experimental data would be needed. Thus far, we have considered only single-mode models. The incorporation of multiple modes would certainly improve the model predictions. The findings of this dissertation should motivate researchers to investigate other viscoelastic fluid models having a complex microstructure in an eccentric annular flow field. From an industrial point of view, it would be desirable to design and optimize products and processes in the future by the combined use of simulations and experiments. So that complex fluid models can be used for industrial tailoring purposes, they should be simple and still reflect the physics that is relevant for the system under investigation. We hope that someday, this goal can be achieved through modeling efforts, the development of software packages that could be used for their solution, the availability of advanced computer facilities, and the transfer of knowledge to non-specialists.

Bibliography


A. Polynomial Interpolation

Taken from Gottlieb et al. [45] and Peyret [67].

A.1. Lagrange Interpolation

Chebyshev-Gauss-Lobatto points:

\[ x_j = \cos\left(\frac{j\pi}{M}\right), \quad j = 0, \ldots, M. \quad (A.1) \]

Lagrange polynomials:

\[ g_j(x) = \frac{(-1)^{j+1} (1-x^2) \sin(M \arccos x)}{\tau_i M \sqrt{1-x^2}(x-x_j)}, \quad (A.2) \]

where \( \tau_0 = \tau_M = 2, \tau_j = 1 \) otherwise.

First-order differentiation matrix:

\[ (D^1_M)_{ij} = \begin{cases} \frac{2M^2 + 1}{6}, & i = j = 0, \\ \frac{2M^2 - 1}{6}, & i = j = M, \\ \frac{\tau_i}{2(1-x_i)}, & i = j; 1 \leq j \leq M - 1, \\ \frac{\tau_j (-1)^{j-1}}{\tau_j - x_j}, & i \neq j. \end{cases} \quad (A.3) \]
A. Polynomial Interpolation

Second-order differentiation matrix:

\[
\begin{cases}
\frac{M^4 - 1}{15},
& i = j; j = 0, M, \\

\frac{(M^2 - 1)(1 - x_i^2) + 3}{3(1 - x_j^2)^2},
& i = j; 1 \leq j \leq M - 1, \\

\frac{2(-1)^i(M^2 + 1)(1 - x_j) - 6}{3(1 - x_i^2)},
& i = 0; 1 \leq j \leq M, \\

\frac{2(-1)^i(M^2 + 1)(1 + x_j) - 6}{3(1 + x_i^2)},
& i = M; 0 \leq j \leq M - 1, \\

\frac{(-1)^i x_i^2 + s_i x_j - 2}{1 - x_i^2(x_j - x_i)},
& i \neq j; 1 \leq i \leq M - 1; 0 \leq j \leq M.
\end{cases}
\]  

(A.4)

A.2. Trigonometric Interpolation

Fourier points:

\[ y_j = \frac{\pi j}{N}, \quad j = 0, \ldots, 2N - 1 \]  

(A.5)

Trigonometric polynomials:

\[ h_j(y) = \frac{1}{2N} \sin \left[ N \left( y - y_j \right) \right] \cot \left( \frac{y - y_j}{2} \right). \]  

(A.6)

First-order differentiation matrix:

\[
\begin{cases}
\frac{1}{2N} (-1)^i \cot \left( \frac{y_i - y_j}{2} \right),
& i \neq j, \\
0,
& i = j.
\end{cases}
\]  

(A.7)

Second-order differentiation matrix:

\[
\begin{cases}
\frac{1}{7} \left( -1 \right)^i \sin^2 \left( \frac{y_i - y_j}{2} \right),
& i \neq j, \\
-\frac{2N^2 + 1}{6},
& i = j.
\end{cases}
\]  

(A.8)
### B. Curriculum Vitae

**Personal Data**

<table>
<thead>
<tr>
<th>Name</th>
<th>Natalie Germann</th>
</tr>
</thead>
<tbody>
<tr>
<td>Date of birth</td>
<td>June 18, 1982</td>
</tr>
<tr>
<td>Place of birth</td>
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<tr>
<td>Citizenship</td>
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**Education and Professional Experience**

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<tr>
<td>2011</td>
<td>Doctor of Science, ETH Zurich, Switzerland</td>
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<tr>
<td>2007–2011</td>
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<tr>
<td>2007</td>
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<tr>
<td>2002–2007</td>
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<tr>
<td>2002</td>
<td>Matura Type A (Latin and ancient Greek)</td>
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<tr>
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<tr>
<td>1989–1995</td>
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