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Abstract

A boundary integral method for the simulation of the deformation of axisymmetric compound non-Newtonian drops suspended in a Newtonian fluid which is subjected to an axisymmetric flow field is developed. The boundary integral formulation for Stokes flow is used and the non-Newtonian stress is treated as a source term. The latter yields an extra integral over the domain of the non-Newtonian material in the boundary integral formulation. By transforming the integral representation for the velocity to cylindrical coordinates we can reduce the dimension of the computational problem. Apart from a numerical validation of the method we present simulation results for a drop consisting of an Oldroyd-B fluid and a viscoelastic material. Moreover, we extend the method to compound drops, which are composed of a viscous inner core encapsulated by a viscoelastic material. The simulation results for these drops are consistent with theoretical results from the literature. Moreover, it is shown that the method can be used to identify the dominant breakup mechanism of compound drops and its relation to the specific non-Newtonian character of the drops.

Keywords: non-Newtonian drops, boundary integral method, Stokes flow.

AMS Classification: 76M15, 76D07.
1 Introduction

Deformable particles, such as gas bubbles, liquid drops and biological cells appear in a wide range of technological and medical applications. Examples include advanced material processing, food processing and pharmaceutical manufacturing. As the deformability of these particles strongly determines the macroscopic properties of the material it composes, much research emphasis has been put on the description of the deformation process of deformable drops (see e.g. Rallison [1] and Stone [2] for a survey). If the drops are sufficiently small, the dynamics of these drops can be analyzed through an investigation of the Stokes equations since the corresponding Reynolds number is small enough to justify the neglect of inertia forces. Most theoretical work has been directed towards the case that the drop and its surrounding fluid consist of different Newtonian fluids in which the interface between the drop and the surrounding fluid is considered infinitely thin and characterized by a constant surface tension. The velocity field can, in these cases, be expressed in terms of a boundary integral over the surface of the drop ([3], [4]). In this paper we extend this boundary integral approach for axisymmetric flow in two ways. First, we incorporate non-Newtonian fluids in the description, which gives rise to additional volume integrals. Secondly, we allow for compound drops, which are composed of multiple layers of (non-)Newtonian material. In this way the method can be used for non-Newtonian drops as well as for e.g. red blood cells or vesicles in which the membrane is modelled as an interface of finite thickness with certain non-Newtonian properties (Brunn [5] and Smeulders [6]). It will be shown that the specific non-Newtonian character of the layers has a significant effect on the dynamics and breakup mechanism of the drop.

The deformation of neutrally buoyant viscous drops with interfacial tension in viscous extensional flows at low Reynolds number was first studied analytically by Taylor [7]. In his analytical work he starts with the solution to the Stokes equation in terms of an expansion in spherical harmonics (Lamb [8]). By approximating the shape of the drop in terms of spherical harmonics one may find a first order approximation of the deformation of the drop and the viscosity of dilute suspension of drops. This technique was refined (Cox [9] and Barthès-Biesel [10]) and extended to other situations such as viscous drops surrounded by a viscoelastic shell (Brunn [5]) or double concentric viscous drops (Stone and Leal [11]). An extensive study of the effects of the interface properties of viscous drops on the rheology of a dilute emulsion was performed by Oldroyd ([12] and [13]). For this purpose a so called cell model was introduced, which enables the study of this problem in the frequency domain. The linear character of the flow problem in the frequency domain allowed Palierne [14] to extend the work of Oldroyd to linear viscoelastic drops with interfacial tension. A different approach was taken by Roscoe [15] who used the work of Jeffery [16] on solid ellipsoidal particles in a viscous flow to describe the deformation of viscoelastic particles.

Apart from theoretical studies the deformation of Newtonian drops in viscous extensional flows at low Reynolds number was first studied numerically by Youngren [17] using a boundary element method. This method has the advantage of reducing the dimension of the computational problem, which significantly reduces the computational cost. The method has extensively been used by others as well in order to simulate the behavior of Newtonian drops in simple flow fields. Apart from improvements in the numerical method (Huang [18]) recent studies have tackled more complicated flow problems (e.g. breakup of drops, Tjahjadi [19]) and incorporated additional physical phenomena (e.g. the effects due to surfactants, Stone [20] and the elasticity of the membrane, Li [27]). Recently the boundary element method was also used to simulate concentrated emulsions of viscous drops in shear flow (Xiofan Li et al. [21])
and Loewenberg and Hinch [22]) and double concentric viscous drops (Stone and Leal [11]).

In many applications, however, the drop consists of a non-Newtonian fluid. Examples include polymer blends, suspensions of vesicles and biological fluids such as blood. The development of the boundary integral method in these cases is much more complicated due to the domain integral which arises from the non-Newtonian contributions. This implies the introduction of a grid covering the layers of the drop containing non-Newtonian fluids, next to the definition of discrete points on the interface, and adds considerably to the numerical cost of simulations of compound non-Newtonian drops. However, compared to a more direct (finite difference) discretization of the Stokes equations, which would also require a grid covering the much larger Newtonian region exterior to the drop, the boundary integral method for non-Newtonian drops is more efficient. Moreover, in several applications the region containing a non-Newtonian fluid forms only a small portion of the flow-problem, e.g. a vesicle in which the drop is formed by a Newtonian fluid which is encapsulated by a non-Newtonian lipid bilayer [6], and the boundary integral method can be used effectively. Bush [23], [24] adopted the boundary element method to analyze extrusion experiments with non-Newtonian fluids. The extension of the method to non-Newtonian drops is valid provided certain assumptions on the smoothness of the non-Newtonian stress tensor are satisfied. These were numerically verified for a two-dimensional drop [25] and for a three-dimensional axisymmetric drop [26]. In the current paper the extension to compound non-Newtonian drops immersed in a Newtonian fluid in axisymmetric flow is considered. The method is illustrated by comparing simulation results for non-Newtonian drops with theoretical results from literature.

The organization of this paper is as follows. In section 2 we present the governing equations and express the velocity field in terms of a domain integral involving the non-Newtonian stress tensor and a boundary integral arising from the Newtonian contributions. Section 3 is devoted to the numerical method used to simulate the deformation of a compound drop. Simulation results for both Newtonian and non-Newtonian compound drops are presented in section 4. Finally, we summarize our findings in section 5.

2 Governing equations

In this section we present the basic equations governing Stokes flow of compound non-Newtonian drops and arrive at the fundamental expression for the velocity-field corresponding to axisymmetric deformation processes.

We consider an isotropic axisymmetric compound non-Newtonian drop placed in an unbounded Newtonian fluid with viscosity $\eta$, which is subjected to a linear elongational flow. The axis of symmetry of the drop is assumed to coincide with the axis of symmetry of the external flow field which is defined by:

$$
\begin{align*}
    u_1^\infty &= G x_1, \\
    u_2^\infty &= -\frac{1}{2} G x_2, \\
    u_3^\infty &= -\frac{1}{2} G x_3,
\end{align*}
$$

with $u_j^\infty$ the $j$–th component of the velocity field with respect to a Cartesian coordinate frame $\{e_1, e_2, e_3\}$ and $G$ the magnitude of the flow. The compound drop consists of $K$ concentric layers, each containing some (non-)Newtonian material. The volumes occupied by the unbounded Newtonian fluid outside the drop and the different layers, counted from the outside to the inside, are denoted by $V^{(0)}$, $V^{(1)}$, $V^{(2)}$, $\ldots$, $V^{(K)}$ respectively (see figure 1). Along the interface $S^{(l)}$ between the domains $V^{(l-1)}$ and $V^{(l)}$, $l = 1, \ldots, K$ there acts a constant interfacial tension $\sigma^{(l)}$. The interface $S^{(1)}$ corresponds to the outer drop interface.
The fluid is incompressible and buoyancy is considered to be absent, i.e. we assume that the densities of all fluids composing the drop are equal. The initial configuration of the drop is taken to be a set of concentric spheres with radii $R^{(l)}$, $l = 1, \ldots, K$. Throughout we will work with dimensionless variables: all lengths are scaled with the initial outer drop radius $R^{(1)} = a$, velocities by $aG$ and viscosities by $\eta$ (Li et al. [27]). In changing to dimensionless variables it is convenient to introduce the following dimensionless parameters:

\begin{equation}
C^{(l)} = \frac{\eta Ga}{\sigma^{(l)}}, \quad \lambda^{(l)} = \frac{\eta^{(l)}}{\eta}, \quad \kappa^{(l)} = \frac{R^{(l)} - R^{(l+1)}}{a}; \quad l = 1, \ldots, K,
\end{equation}

where $C^{(l)}$ is the capillary number of the $l$-th interface, which is a measure of the ratio between the viscous and interfacial tension stresses. The parameter $\lambda^{(l)}$ is the ratio between the viscosity $\eta^{(l)}$ in the $l$-th domain and the exterior viscosity $\eta$ and $\kappa^{(l)}$ the ratio of the thickness of the undeformed spherical layer $V^{(l)}$ and the outer radius of the drop, where for convenience we introduced $R^{(K+1)} = 0$. In order to characterize the degree of distortion of the interfaces due to the external velocity field one commonly defines a deformation parameter $D^{(l)}$ as:

\begin{equation}
D^{(l)} = \frac{r_{\text{max}}^{(l)} - r_{\text{min}}^{(l)}}{r_{\text{max}}^{(l)} + r_{\text{min}}^{(l)}}, \quad l = 1, \ldots, K,
\end{equation}

where $r_{\text{max}}^{(l)}$ and $r_{\text{min}}^{(l)}$ denote the longest and shortest principal axes of the $l$-th deformed interface.

The flow in and around a compound non-Newtonian drop as introduced above is governed by the Stokes equations if the Reynolds number $Re = (\rho a^2 G)/\eta$ (with $\rho$ the density) is sufficiently small (Pozrikidis [28]). In particular the basic equations read:

\begin{equation}
\partial_j u_j = 0; \quad \partial_j \pi_{ij} = 0
\end{equation}

with $\pi_{ij}$ the total stress tensor and $\partial_j = \partial/\partial x_j$. In equation (2.4), as in the rest of this paper, the summation convention is adopted which implies summation over repeated indices. The Stokes equations given above are further supplemented with suitable matching conditions at the interfaces $S^{(l)}$ and asymptotic conditions as $|x| \to \infty$. As the different layers of the drop can consist of different types of non-Newtonian materials it is convenient to distinguish between a set of local stress tensors $\pi_{ij}^{(l)}$, $l = 1, \ldots, K$, corresponding to the total stress tensor in the domain $V^{(l)}$, i.e. $\pi_{ij}^{(l)} = \pi_{ij}$ if $x \in V^{(l)}$. The superscript $(l)$ will be used for the other flow quantities as well to indicate the domain of definition.

In the domain $V^{(l)}$ the total stress tensor $\pi_{ij}$ is decomposed as:

\begin{equation}
\pi_{ij}^{(l)} = -\delta_{ij} P^{(l)} + \lambda^{(l)} \dot{\gamma}_{ij}^{(l)} + \tau_{ij}^{(l)}, \quad i, j = 1, \ldots, 3,
\end{equation}

where $P^{(l)}$ is the isotropic pressure field and $\dot{\gamma}_{ij}^{(l)}$ the rate-of-strain tensor:

\begin{equation}
\dot{\gamma}_{ij}^{(l)} = \partial_i u_j^{(l)} + \partial_j u_i^{(l)}.
\end{equation}

The non-Newtonian part of the stress tensor $\tau_{ij}^{(l)}$ in (2.5) obeys a certain constitutive equation which describes the non-Newtonian character of the fluid motion. In this paper we focus on the Maxwell model and a specific elastic model, although the method presented here can be
extended to quite general rheological models. The Maxwell model finds its origin in polymer rheology and contains two parameters: a relaxation time \( t_p^{(l)} \) and the polymer contribution to the zero-shear-rate viscosity \( \eta_p^{(l)} \). The constitutive equation for this model is given by:

\[
\mu_p^{(l)} D_t \tau_{ij}^{(l)} + \tau_{ij}^{(l)} = \lambda_p^{(l)} \ddot{\epsilon}_{ij}^{(l)}
\]

where \( D_t \) denotes the upper-convected time derivative (Bird [29]), \( \mu_p^{(l)} = G t_p^{(l)} \) the dimensionless relaxation time and \( \lambda_p^{(l)} = \eta_p^{(l)}/\eta \). The total stress tensor \( \tau_{ij}^{(l)} \) for this model corresponds to the well-known Oldroyd-B model (Bird [29]). The constitutive equation for the elastic model considered here is given by:

\[
\ddot{\tau}_{ij}^{(l)} = c_1^{(l)} B_{ij} + c_2^{(l)} B_{ij}^{-1},
\]

where \( c_1^{(l)} \) and \( c_2^{(l)} \) are arbitrary constants. In expression (2.7) \( B_{ij} \) is the so-called left Cauchy-Green-strain tensor which is defined as:

\[
B_{ij} = \frac{\partial x_i}{\partial X_l} \frac{\partial x_j}{\partial X_j},
\]

where \( x \) describes the present and \( X \) the initial configuration of the material. The total stress tensor \( \tau_{ij}^{(l)} \) for this model is the so-called left Cauchy-Green-strain tensor which is defined as:

\[
\tau_{ij}^{(l)} = \dot{C}_{ij}^{(l)} + \frac{1}{2} \delta_{ij} \dot{\epsilon}^{(l)},
\]

where \( \dot{C}_{ij}^{(l)} \) is the rate of deformation tensor and \( \dot{\epsilon}^{(l)} \) is the strain rate tensor.

The identification of the non-Newtonian stress contribution to \( \tau_{ij}^{(l)} \) is not unique. This can be used to simplify the subsequent formulation by incorporating a part of the Newtonian stress into the non-Newtonian stress. In particular, by redefining the non-Newtonian stress as:

\[
\ddot{\tau}_{ij}^{(l)} \rightarrow \ddot{\tau}_{ij}^{(l)} + (\lambda^{(l)} - 1) \ddot{\epsilon}_{ij}^{(l)},
\]

we can use \( \lambda^{(l)} = 1 \) in (2.5) without loss of generality (Toose et al. [25] and [26]). In the remainder of the paper the redefined non-Newtonian stress tensor will be referred to as the extra stress tensor. Assuming that the Reynolds number is small the fluid motion in the \( l \)-th domain is governed by the inhomogeneous Stokes equations:

\[
\begin{aligned}
\partial_j u_i^{(l)} - \partial_i P^{(l)} &= -\partial_j \ddot{\tau}_{ij}^{(l)}, \\
\partial_j u_j^{(l)} &= 0.
\end{aligned}
\]

The velocity and the total stress tensor satisfy the following matching and asymptotic conditions:

\[
\begin{aligned}
[u_i]_{S^{(l)}} &= 0, \quad C^{(l)} \left[ \tau_{ij} n_j \right]_{S^{(l)}} = n_i^{(l)} \partial_j n_j^{(l)}, \quad \text{for } l = 1, 2, \ldots, K \\
\big[ u_i \big]_{S^{(l)}} &= 0, \quad \big[ u_i \big]_{S^{(l)}} \rightarrow \big[ u_i \big]_{\infty}, \quad \text{as } |x| \rightarrow \infty,
\end{aligned}
\]

where \( n^{(l)} \) is the outward unit normal on \( S^{(l)} \), \( \partial_j n_j^{(l)} \) is the boundary curvature and \( \big[ \cdot \big]_{S^{(l)}} \) denotes the jump of the quantity between the brackets over the interface \( S^{(l)} \) counted in the direction \( n^{(l)} \):

\[
[f]_{S^{(l)}} = f^{(l)}(x) - f^{(l-1)}(x), \quad x \rightarrow S^{(l)}.
\]
As the time-dependence does not appear explicitly in (2.9) we use a kinematic constraint which describes the fluid motion in the different domains. The motion of the domain \( V(t) \) and its boundaries is modelled by considering the domain as a set of material points. The trajectories of these points can be followed using a Lagrangian representation of the velocity:

\[
d_t x_i = u_i^{(l)} (x), \quad \forall x \in V^{(l)} (t),
\]

with \( d_t \) the material time derivative. For the evolution equations (2.6) and (2.12) initial conditions need to be specified. For the interfaces \( S^{(l)} \) we start with a spherical shape, whereas for the non-Newtonian contribution to the stress tensor we assume an isotropic stress distribution:

\[
\tau_{ij} (0) = Q \delta_{ij},
\]

with \( Q \) constant.

Assuming that the non-Newtonian stress tensor is known at time \( t \), and interpreting the right hand side of (2.9) as a source term we can construct a solution for the velocity at time \( t \) in terms of boundary integral equations (Lorentz [3]). Following Ladyzhenskaya [4] it can be shown that the integral representation for the velocity is given by (Toose et al. [26]):

\[
u_k (x) = \nu_k^{\infty} (x) + \sum_{l=1}^{K} \int_{V^{(l)}} J_{ik} (r) \partial_j z_{ij}^{(l)} (y) \, dy + \sum_{l=1}^{K} \int_{S^{(l)}} J_{ik} (r) q_i^{(l)} (y) \, dS_y,
\]

with \( r = x - y \), \( J_{ik} (r) \) the Green’s function for the Stokes problem [3] and \( q_i^{(l)} \) a surface force which is defined as:

\[q_i^{(l)} = \left[ \tau_{ij} \right]_{S^{(l)}} n_j^{(l)} - \frac{1}{C^{(l)}} \partial_i \bar{n}_j^{(l)}.
\]

From the latter definition we see that the interfacial tension and the discontinuity of the extra stress tensor across the surface \( S^{(l)} \) lead to the surface force \( q_i^{(l)} \).

The flow problem defined above remains axisymmetric in time and hence we can reduce the dimension of the computational problem by transforming the integral equation (2.14) to cylindrical coordinates (Toose et al. [26]). To perform this transformation it is convenient to define cylindrical coordinates:

\[
\begin{align*}
x_1 &= \bar{x}_1 \\
x_2 &= \bar{x}_2 \cos(\bar{x}_3) \\
x_3 &= \bar{x}_2 \sin(\bar{x}_3),
\end{align*}
\]

where \( \bar{x}_1, \bar{x}_2 \) and \( \bar{x}_3 \) indicate the axial, radial and azimuthal components respectively. Observe that we use \( (\bar{x}_1, \bar{x}_2, \bar{x}_3) \) instead of the more common \( (x, r, \phi) \) in view of the summation convention. In the sequel we denote corresponding components with respect to cylindrical coordinates with an over-bar.

By transforming the integral expression (2.14) to cylindrical coordinates and performing the integration over the azimuthal direction analytically we obtain the following integral equation [26]:

\[
\tilde{u}_\alpha (\bar{x}) = \tilde{u}_\alpha^{\infty} (\bar{x}) + \sum_{l=1}^{K} \int_{V^{(l)}} M^\beta_\alpha (\bar{r}) \partial_\gamma \tilde{z}_{\beta \gamma}^{(l)} (\bar{y}) \bar{y}_2 \, d\bar{y}_2 d\bar{y}_1 + \sum_{l=1}^{K} \int_{S^{(l)}} M^\beta_\alpha (\bar{r}) \tilde{n}_{\beta \gamma}^{(l)} (\bar{y}) \bar{y}_2 \, d\ell_y,
\]

where \( \tilde{V}^{(l)} \) and \( \tilde{S}^{(l)} \) are the integration domains in cylindrical coordinates, which correspond to \( V^{(l)} \) and \( S^{(l)} \) respectively. In the expression above \( d\ell_y = \{1 + (\partial_1 \bar{y}_2)^2\}^{1/2} d\bar{y}_1 \) is the differential
arclength, \( M^\alpha_\alpha(\mathbf{r}) \) the transformed Green’s function and \( \tilde{\partial}_\gamma \tilde{\tau}^{(l)}_{\beta \gamma}(y) \) the divergence of the extra stress tensor in cylindrical coordinates which is defined as:

\[
\tilde{\partial}_\gamma \tilde{\tau}^{(l)}_{\beta \gamma}(y) = g^{\alpha \beta} \left( \partial_\alpha \tilde{\tau}^{(l)}_{\beta \gamma} - \left\{ \frac{\lambda}{\beta \alpha} \right\} \tilde{\tau}^{(l)}_{\alpha \gamma} - \left\{ \frac{\lambda}{\gamma \alpha} \right\} \tilde{\tau}^{(l)}_{\beta \lambda} \right),
\]

where the coefficients \( \left\{ \frac{\lambda}{\alpha \beta} \right\} \) are the Christoffel symbols and \( g^{\alpha \beta} \) is the metric tensor of the transformation. In expression (2.16) we reduced the three dimensional problem to a two dimensional one. The presence of the non-Newtonian stress, however, makes it impossible to reduce the problem to one dimension as is possible for a purely Newtonian drop [11]. This implies that the computational effort required to solve non-Newtonian problems is considerably higher than for corresponding Newtonian problems. However, since only the volume of the drop needs to be discretized an efficient method can be arrived at compared to a conventional finite difference method.

With equation (2.16) we have expressed the solution for the velocity field in the entire flow domain in terms of boundary and domain integrals, provided that the non-Newtonian stress tensor and the shape of the interface are given. In the next section we will describe the complete method for solving the full time-dependent problem and show how the integral equation (2.16) is incorporated in this method.

3 Numerical method

In this section the numerical method used to evaluate the boundary and domain integrals, the non-Newtonian stress tensor and the evolution of all surfaces \( S^{(l)}_l, l = 1, \ldots, K \) are given in detail. In subsection 3.1 the complete simulation method for the evolution of a drop consisting of \( K \) concentric layers of (non-)Newtonian material is described. The description of the geometry is provided in subsection 3.2. The numerical method to calculate the velocity field, i.e. the evaluation of (2.16) is provided in subsection 3.3. In subsection 3.4 the time integration of the evolution equations (2.6) and (2.12) is presented.

3.1 Numerical algorithm

In anticipation of the numerical evaluation of the boundary integral formulation, a set of discrete collocation points is introduced in the internal domains \( V^{(l)} \) and on the interfaces \( S^{(l)} \). Using these discrete points we introduce boundary elements to describe the shape of the interfaces and internal grid cells to cover the internal domains \( V^{(l)} \). In order to describe the state of the drop both the non-Newtonian stress and the velocity need to be specified at these collocation points. At the start of the simulation the stress is given by its initial conditions, whereas the velocity corresponding to the initial stress can be calculated using the boundary integral formulation for the velocity.

The complete procedure used to simulate the evolution of the non-Newtonian axisymmetric drop can be sketched as follows. Using the velocity given at time \( t \) and the evolution equation (2.12) we can calculate the new positions of the collocation points, and consequently the shape of the interfaces of the drop at \( t + \Delta t \). Moreover, depending on the non-Newtonian model chosen, we either integrate (2.6) in time or calculate (2.7) explicitly in order to obtain the non-Newtonian stress tensor at the new time level. With this new stress tensor and shape of the interfaces, a new velocity field at time level \( t + \Delta t \) can be calculated from (2.16). At
this point all the elements which describe the state of the drop, i.e. the collocation points describing the shape of the drop and the velocity and the non-Newtonian stress at these points, have advanced one time step. Repeating this explicit time integration procedure gives the evolution of the stress tensor, the velocity field and the shape of all the boundaries $S^{(l)}$.

After imposing the initial condition for the non-Newtonian stress and the interfaces and after calculating the velocity field corresponding to this initial geometry and stress, the full algorithm can be summarized as follows:

1. update positions of the collocation points by integrating (2.12) over one time step to yield $S^{(l)}(t + \Delta t), \ l = 1, \ldots, K$,

2. calculate the stress tensor at $t + \Delta t$ by solving (2.6) given $\bar{u}_\alpha$ at time $t$ or by evaluating (2.7) using the new positions at time $t + \Delta t$

3. calculate the velocity field at $t + \Delta t$ using (2.16) given $\bar{\tau}_{\beta\gamma}$ and $\bar{S}^{(l)}$ from step 1 and 2.

In the next three subsections we will discuss the representation of $S^{(l)}$ and $V^{(l)}$, the calculation of the velocity field and the time integration of both the non-Newtonian stress tensor and the boundaries respectively.

3.2 Geometry Description

The numerical implementation of the integrals in (2.16) requires an accurate representation of the interfaces $\tilde{S}^{(l)}$ into $N_b^{(l)}$ boundary elements and the subdivision of the internal domains $\tilde{V}^{(l)}$ into $N_c^{(l)}$ internal cells. For reasons of convenience we drop the superscript $(l)$ whenever possible in this subsection.

Since the curvature of the interface is required in order to calculate the surface force $\vec{q}$, we use higher order boundary elements to represent the interfaces, whereas for the internal cells we adopt quadrilateral elements. In order to be able to use standard integration techniques for the calculation of the domain integral we map these quadrilateral elements to a standard square element using a bilinear transformation (Farin [30]):

$$\vec{X}(s_1, s_2) = A_1(1 - s_1)(1 - s_2) + A_2s_1(1 - s_2) + A_3s_1s_2 + A_4(1 - s_1)s_2,$$  
(3.1)

where $A_1$, $A_2$, $A_3$ and $A_4$ are the vertices of the quadrilateral element and $s_1$ and $s_2$ are the local coordinates of the corresponding standard element. For the higher order boundary elements we use piecewise $C^2$ cubic B-spline elements (DeBoor [31] and Farin [30]). The use of cubic B-spline elements requires a suitable parametrisation of the curve along the collocation points which is obtained with a chord length parametrisation:

$$v[0] = 0 \quad v[i] = v[i - 1] + \| \vec{x}_i - \vec{x}_{i-1} \|, \quad i = 1, \ldots, N_b,$$

where $\vec{x}_i$ is the $i$-th collocation point on the interface $S$. On this set of nodes we introduce a cubic B-spline curve $\vec{s}$ as (DeBoor [31]):

$$\vec{s}(v) = \sum_{j=-3}^{N_b-1} \vec{d}_j B_j(v),$$  
(3.2)
where \( B_j(v) \) is a cubic B-spline and \( \{d_j\} \) the control vertices of the B-spline curve such that \( \bar{s}(v[i]) = \bar{x}_i \). It is noted that this interpolation problem can be fully specified if the symmetry of the drop is taken into account. The interpolation problem for \( \bar{s}(v) \) leads to a sparse matrix problem for \( \{d_j\} \) which is solved using a band matrix solver.

From the control vertices \( \{d_j\} \) we can calculate first and second order derivatives as well as points on the interface \( S \) using the DeBoor scheme (DeBoor [31]). The advantage of the DeBoor scheme is that we only use combinations of these control vertices, and do not have to evaluate the B-spline functions explicitly. From the first derivatives of the B-spline curve \( \bar{s} \) we can calculate the components of the normal vector using:

\[
\bar{n}_1(v) = -\frac{1}{J} \partial_v \bar{s}_2, \quad \bar{n}_2(v) = \frac{1}{J} \partial_v \bar{s}_1,
\]

where \( J \) is the differential arclength of the curve:

\[
J(v) = \sqrt{(\partial_v \bar{s}_1)^2 + (\partial_v \bar{s}_2)^2} \quad (3.3)
\]

The curvature \( \partial_\alpha \bar{n}_\alpha \) of the interface \( \bar{S} \) of the drop is equal to the sum of the values of the curvatures in two arbitrary but mutually perpendicular directions. In case of an axisymmetric body described by cylindrical coordinates it is most convenient to consider the curvatures in the meridional and azimuthal directions:

\[
\partial_\alpha \bar{n}_\alpha(v) = \frac{1}{(J)^2} \left\{ \partial_{vv} \bar{s}_1 \bar{n}_1 - \partial_{vv} \bar{s}_2 \bar{n}_2 \right\} + \left| \frac{n_2}{\bar{s}_2} \right|,
\]

where we have used a signed curvature [30] for the meridional direction.

### 3.3 Calculation of the velocity

In order to calculate the velocity field we have to solve the integral equation (2.16) at all the collocation points. If viscosity ratios in the layers differ from one we incorporate a part of the Newtonian stress in the non-Newtonian stress by redefining the non-Newtonian stress tensor as described in section 2. However, by this procedure the extra stress tensor becomes an explicit function of the velocity which implies that we solve (2.16) iteratively at each time step. In this iteration we assume a trial velocity field which is used to calculate the Newtonian part of the extra stress tensor. Application of (2.16) yields a new velocity field, which serves as the trial velocity field for the next iteration step. This iteration procedure is applied, until the residual \( R^n \) obtained after \( n \) iterations is smaller than a prespecified small number. The residual is defined as the discrete \( L_2 \) norm of the difference in the velocity at all collocation points in two successive iterations:

\[
R^{(n)} = \| \bar{u}^{(n)} - \bar{u}^{(n-1)} \| \equiv \left\{ \frac{1}{N_p} \sum_{i=1}^{N_p} |\bar{u}_i^{(n)} - \bar{u}_i^{(n-1)}|^2 \right\}^{1/2}, \quad (3.4)
\]

where \( \bar{u}^{(n)} \) is the velocity in the \( n \)-th iteration level and \( N_p \) is the total number of collocation points. Computations showed that the iteration process can be stopped if the residual is smaller than \( 10^{-6} \): more iterations do not effect the results. To reduce the required number of iterations, we set the initial trial velocity equal to the converged velocity obtained in the
The constants $a$ and $b$ are determined such that the following conditions are met:

$$v(-1) = v^{(l)}[m], \quad v(1) = v^{(l)}[m + 1], \quad \frac{\partial v}{\partial \omega} |_{\hat{v}^{(l)}} = 0.$$  \hfill (3.7)

where $\hat{v}^{(l)}$ is defined as:

$$\hat{v}^{(l)} = \left\{ v^{(l)} \in v^{(l)} \mid \| \mathbf{x} - \mathbf{s}^{(l)}(\hat{v}^{(l)}) \| = \text{minimal} \right\}.$$  

Transformation (3.6) leads to:

$$I_2(\mathbf{x}; S_m^{(l)}) = \int_{-1}^{1} M_\alpha^\beta(\mathbf{r}) \tilde{n}_\beta^{(l)}(v(\omega)) \tilde{s}_2^{(l)}(v(\omega)) J^{(l)}(v(\omega)) \partial_\omega v \, d\omega.$$  \hfill (3.8)

It is noted that the Jacobian $\partial_\omega v$ removes the singularity or quasi-singularity if $\mathbf{x}$ is located either in or outside the integration interval. The removal of the quasi-singularity leads to a higher accuracy, which is especially useful for drops with very thin layers. The resulting
transformed integral in (3.8) is evaluated numerically using Gauss quadrature (Patridge [33] and Evans [34]).

In the remaining part of this section we discuss the numerical evaluation of the domain integral. The domain integral \( \mathcal{I}_1(\bar{x}; \bar{V}_m^{(l)}) \) can be written as:

\[
\mathcal{I}_1(\bar{x}; \bar{V}_m^{(l)}) = \int_{\bar{V}_m^{(l)}} M^\beta_\alpha(\bar{r}) \frac{\partial}{\partial y_2} \bar{v}_{\beta g}(\bar{y}) d\bar{y}_2 d\bar{y}_1
\]

\[
= \int_0^1 \int_0^1 M^\beta_\alpha(\bar{r}) \frac{\partial}{\partial y_2} \bar{v}_{\beta g}(\bar{X}(s_1, s_2)) \bar{X}_2(s_1, s_2) J_B^{(l)}(s_1, s_2) ds_2 ds_1,
\]

where we have used the bilinear representation (3.1) of the \( m \)-th quadrilateral element and the Jacobian \( J_B^{(l)} \) is given by:

\[
J_B^{(l)} = \det \left( \frac{\partial \bar{X}_2(1/2, 1/2)}{\partial s_\beta} \right).
\]

Unlike the interface integrals, we now do distinguish between two cases; (a) the internal cell does not contain the point \( \bar{x} \) and (b) the internal cell does contain the point \( \bar{x} \). In case (a) the domain integrand is regular and a normal Gauss quadrature is used to evaluate the integral. In case (b) the integrand is singular and a special treatment of the singularities in the kernels is required. To remove the singularity we use the same quadratic transformation as was used for the boundary integral for both integration variables \( s_1 \) and \( s_2 \). After the removal of the singularity a normal Gauss quadrature is used in both integration directions to evaluate the integral numerically.

With these numerical techniques we have devised a second order accurate method to calculate the velocity of the stationary problem (2.16) in all collocation points. In the next subsection we discuss the coupling of this solution with the time dependent problem.

### 3.4 Time integration

In this subsection we describe the method to find the non-Newtonian stress tensor and the shape of the interface \( \bar{S}^{(l)} \) at the new time level.

Updating the shape of the interface and the grid covering the \( l \)-th layer \( \bar{V}^{(l)} \) requires the calculation of the position of all the collocation points at the new time level. This calculation can be performed by time integration of (2.12) with an Euler forward scheme:

\[
\bar{x}_i(t_{n+1}) = \bar{x}_i(t_n) + \Delta t \, \bar{u}_i,
\]

with \( t_n = n\Delta t \) where \( \Delta t \) is a constant time step and \( \bar{u}_i \) the velocity at the \( i \)-th collocation point \( \bar{x}_i \). Moving the grid points in this way, however, may lead to a clustering of both internal and boundary grid points in certain regions and hence a highly deformed grid develops, leading to inaccurate results. The clustering arises directly from the fact that there are no restrictions on the stress tensor in tangential direction implying that the points can move freely along the interface in the direction of the external velocity field. To prevent the clustering of the collocation points we redistribute the boundary points every timestep. To this end we introduce an auxiliary set of collocation points \( \bar{y}_i \) at the interface by:

\[
\bar{y}_i(t_{n+1}) = \bar{y}_i(t_n) + \Delta t \left( \bar{u}_i + \alpha_{i}^{(l)} t_i^{(l)} \right), \quad \bar{y}_i \in \bar{S}^{(l)},
\]

\[
(3.10)
\]
where \( \mathbf{t}_i \) is the unit tangent vector along the \( l \)-th interface (no summation over the index \( i \)).

The coefficients \( \alpha_i^{(l)} \) are determined such that:

\[
\| \mathbf{y}_{i+1} - \mathbf{y}_i \| \left( \partial_\gamma n_i^\gamma (\mathbf{y}_i) + \partial_\nu n_i^\nu (\mathbf{y}_{i+1}) \right),
\]

is constant for all \( i \) at the new time level. By using this grid movement scheme the points are redistributed in such a way that areas with high curvature have a somewhat higher concentration of grid points then areas with low curvature. The interior collocation points are found subsequently by generating a new grid at each time step. This is done by interpolation between the new auxiliary interface points on the interfaces \( \tilde{S}^{(l)} \) and \( \tilde{S}^{(l+1)} \).

For a layer consisting of a viscoelastic model, however, we simply use the Euler scheme (3.9) since the non-fading elasticity of the stress tensor compensates the viscous stress in tangential direction which prevents clustering of the collocation points.

The new non-Newtonian stress tensor in the numerical algorithm is obtained either by explicitly evaluating the stress (2.7) at the new time level or by integration of the constitutive equation (2.6) depending on the chosen model. In order to calculate the stress for the viscoelastic model we need to evaluate the Cauchy-Green tensor (2.8) which requires the derivatives of the deformed grid with respect to a reference grid. For the reference grid we use the grid at the initial time level, where the partial derivatives are evaluated using a second order accurate finite difference scheme. Details concerning finite difference methods for curvilinear coordinates can be found in Thompson [35]. In case the non-Newtonian stress tensor is prescribed by a differential constitutive equation we integrate the stress in time. To perform this integration for the Maxwell model we have to evaluate the upperconvected time derivative using a partial or a material time derivative. The use of the material time derivative, however, leads to a convective term which is somewhat difficult to calculate. For this problem it is more convenient to use the material time derivative which does not require an explicit calculation of the convective term. This implies, that the new non-Newtonian stress tensor \( \tilde{\tau}_{\beta\gamma} \) will be defined on the grid which is convected according to (3.9). Integration of (2.6) with an Euler forward scheme leads to:

\[
\tilde{\tau}_{\beta\gamma}(t_{n+1}) = \tilde{\tau}_{\beta\gamma}(t_n) + \Delta t \left( g^{\nu\mu} \left[ \partial_\nu \tilde{u}_\mu(t_n) \tilde{\tau}_{\beta\gamma}(t_n) + \tilde{\tau}_{\beta\mu}(t_n) \tilde{u}_\gamma(t_n) \right] \right) + \tilde{R}_{\beta\gamma}(t_n) \tag{3.11}
\]

with \( g^{\nu\mu} \) the metric tensor and \( \tilde{R}_{\beta\gamma} \) given by:

\[
\tilde{R}_{\beta\gamma}(t_n) = \frac{\lambda^{(p)}}{\mu} \left( \partial_\beta \tilde{u}_\gamma(t_n) + \partial_\gamma \tilde{u}_\beta(t_n) \right) - \frac{1}{\mu} \tilde{\tau}_{\beta\gamma}^{NN}(t_n) \tag{3.12}
\]

The covariant derivative \( \partial_\beta \) in (3.11) and (3.12) is defined as (Bird [29]):

\[
\partial_\beta \tilde{u}_\gamma = \frac{\partial \tilde{u}_\gamma}{\partial x_\beta} - \left\{ \begin{array}{c} \gamma \\ \beta \end{array} \right\} \tilde{u}_\xi, \tag{3.13}
\]

As an alternative to the first order Euler scheme a second- or higher-order Runge-Kutta scheme can be used.

Due to the Lagrangian approach the new stress tensor \( \tilde{\tau}_{\beta\gamma}(t_{n+1}) \) resulting from (3.11) is defined on the grid whose positions are given by (3.9). In order to find it on the new grid \( \tilde{y}_i \) the stress tensor is interpolated by:

\[
\tilde{\tau}_{\beta\gamma}(\tilde{y}_i) \approx \frac{1}{\sum_{j \in J} 1/(D_{ij})^2} \sum_{j \in J} \frac{\tilde{\tau}_{\beta\gamma}(\tilde{x}_j)}{(D_{ij})^2}, \tag{3.14}
\]

11
with \( D_{ij} = |\vec{y}_i - \vec{x}_j| \) and the summation \( j \in J \) involves all the indices of the nearest neighbors of the grid point \( \vec{y}_i \). To ensure that this interpolation is sufficiently accurate we introduce a time step restriction:

\[
\Delta t = \epsilon \min_n \left\{ \frac{\Delta \vec{x}_n}{\vec{u}_n} \right\},
\]

where \( \Delta \vec{x}_n \) is the shortest side of the \( n \)-th internal cell, \( \vec{u}_n \) the mean velocity over this element and \( \epsilon \) is of the order 0.1.

\section{Results and discussion}

In this section we present the results of numerical calculations to validate and illustrate the method. For the validation of the numerical method we perform a grid refinement study and compare the numerical results with available analytical results known from literature. Although the method can be used for drops with many layers, here we focus on drops with either one or two layers (i.e. \( K = 1 \) or \( K = 2 \) respectively). The one layer drop is discussed in section 4.1, whereas the results for the two layer drop are presented in section 4.2. In order to remove the effects of the deformation history of the drop, we use the relaxed state of the non-Newtonian stress tensor at \( t = 0 \) (i.e. \( Q = 0 \) in equation (2.13)).

\subsection{Singe layer non-Newtonian drop}

In this subsection we study the response of an axisymmetric drop containing an Oldroyd-B fluid or a viscoelastic material to an elongational flow. Before we turn to the validation of the method, we first show a typical example of a deformed drop and the corresponding velocity field.

In figure 2 a vector plot of the steady state velocity field of a fluid like drop (i.e. viscous or Oldroyd-B drop) at \( C = 0.1 \) is drawn. In the velocity field we can distinguish two vortices, which are also drawn schematically in the lower part of the figure. For the viscoelastic drops no vortices exist since the velocity becomes zero in the steady state due to the non fading elasticity of the viscoelastic material.

We investigate the order of accuracy of the numerical method using grid refinement (Toose [25]). Suppose we have numerically obtained the solution at a certain time \( t_0 \) on a given grid. The deformation \( D(t_0) \) should asymptotically converge to its analytical value at a specific rate in case the grid is refined. The order of accuracy of the method can be obtained from the values of \( D(t_0) \) on subsequent refinement levels of the grid. Especially the values of the convergence rate \( \rho_i \), defined by:

\[
\rho_i = \frac{D_{i+1} - D_i}{D_{i+2} - D_{i+1}}, \text{ for } i \geq 0,
\]

on the set of grids is of importance in this respect. Here \( D_{i} \) is the deformation of the drop on the \( i \)-th refinement level at time \( t_0 \). The grid is usually refined with a Romberg sequence, in which the grid spacing is halved at each refinement. It is, however, more efficient to use a Bulirsch-sequence consisting of two intertwined Romberg sequences (Stoer [36]). In table 5 we present typical grid refinement results for an Oldroyd-B drop with \( \lambda_p = 5 \) and \( \mu_p = 1 \) using the deformation at \( t_0 = 1 \) at two different values of the capillary number. Analyzing the convergence behaviour of the deformation we find that the method is second order accurate.
in space for this specific case. Besides a rapid convergence, the method also gives very good results for relatively coarse grids. Using a similar refinement study for the timestep we find that accuracy in time is first order for the Euler and second order for the compact storage four stage Runge-Kutta scheme (Jameson [37]). To provide a way to verify the correctness of the numerical method we compare our numerical results with theoretical results from literature and investigate several limiting cases of the non-Newtonian stress tensor.

The Oldroyd-B model contains three independent parameters, the viscosity ratios $\lambda$ and $\lambda_p$ and the dimensionless relaxation time $\mu_p$. If either $\lambda_p$ or $\mu_p$ approaches zero, an essentially Newtonian behavior results (Toose et al. [26]). Numerical calculations showed that the limiting behaviour of the Oldroyd-B drop is correctly recovered. To verify the results for moderate values of $\lambda_p$ and $\mu_p$ we use theoretical results obtained by Delaby et al. [38]. The theoretical background for these results is based on the linear theory for viscous emulsions of Oldroyd [12], which has recently been extended to viscoelastic emulsions by Palierne [14]. This linear theory starts with the solution to the Stokes equation in terms of an expansion in spherical harmonics (Lamb [8]). By approximating the shape interface in terms of spherical harmonics one can use the boundary conditions of the problem to find the leading-order shape correction. The amplitude of this correction for the radial displacement of the interface, is given by:

$$A(\omega) = \frac{5G_d^*(19G_d^* + 16G_o^*)}{(2G_d^* + 3G_o^*)(19G_d^* + 16G_o^*) + C(G_o^* + G_d^*)}, \quad (4.2)$$

where $G_d^*$ and $G_o^*$ are the complex moduli of the drop fluid and outer fluid respectively. In case of an Oldroyd-B drop placed in a viscous fluid the complex moduli $G_d^*$ and $G_o^*$ are given by:

$$G_d^* = i\omega \lambda + \frac{i\omega \lambda_p}{1 + i\omega \mu_p}, \quad G_o^* = i\omega,$$

with $i$ the imaginary unit and $\omega$ the frequency of the oscillations of the external flow. The amplitude $A(\omega)$ can also be written in the form:

$$A(\omega) = A_\infty \left( 1 - \sum_j \frac{B_j}{1 + i\omega \mu_j} \right),$$

where the coefficients $B_j$, relaxation times $\mu_j$ of the deformation process and $A_\infty$ are evaluated numerically for particular values of the parameters. The response of the deformation to an elongational velocity is then given by:

$$D(t) = \frac{3A_\infty C}{4} \left( \left( 1 - \sum_j B_j \right)t + \sum_j \frac{B_j \mu_j}{1 - e^{-t/\mu_j}} \right). \quad (4.3)$$

In figure 3 we show a plot of expression (4.3) for three values of the relaxation time $\mu_p$ and $\lambda_p = 5$ of the Oldroyd-B fluid and some corresponding numerical results. The numerical results were generated using a grid with 48 boundary elements and 624 cells in the internal domain. For small values of the relaxation time of the Oldroyd-B model, the analytical and numerical results are in good agreement. The steady state deformation of the analytical solution for small relaxation times of the Oldroyd-B model lies somewhat below the numerically obtained values. Comparison with a second order theory for viscous drops (Barthes-Biesel [10]) reveals that the numerically obtained steady state deformation is correct and that the linear theory
slightly underpredicts this deformation. For higher values of $\mu_p$ there are some discrepancies between the analytical and numerical results which cannot be explained by shortcomings in the first order theory. In this case the differences are caused by the nonlinear character of the upperconvected time derivative in the constitutive equation for the Maxwell model. In order to establish this we considered a simulation performed with a linear time derivative and observed that the numerical and analytical results were in better agreement. Analyzing this equation we observe that the nonlinearity increases with increasing relaxation time of the model which explains why the discrepancies increase with increasing relaxation time. The overall agreement, however, is very satisfactory.

The viscoelastic model, given in (2.7), contains two independent parameters, $c_1$ and $c_2$. In this subsection we consider two cases: the viscoelastic I and the viscoelastic II model. To verify our results we use theoretical results obtained by Roscoe [15] for viscoelastic spheres. Assuming that the shape of the drop remains ellipsoidal in time, the longest axis $r_{\text{max}}$ of the drop follows from the following nonlinear equations [15]:

$$
\begin{align*}
\frac{5}{4}g_2(r_{\text{max}}^2 - r_{\text{max}}^{-1}) &= \frac{5\eta G}{2c_1} \quad \text{for } c_2 = 0, \\
\frac{5}{4}g_2(r_{\text{max}} - r_{\text{max}}^{-2}) &= \frac{5\eta G}{2c_2} \quad \text{for } c_1 = 0,
\end{align*}
$$

(4.4)

where $g_2$ is defined as (Jeffery [16]):

$$
g_2 = \int_0^\infty \frac{z \, dz}{(r_{\text{max}}^2 + z)(r_{\text{max}}^{-1} + z)(r_{\text{max}}^{-1} + z)^2}^{1/2}.
$$

By evaluating $g_2$ numerically for several values $r_{\text{max}}$ we can find the relation between the deformation $D$ and the constants $c_1$ or $c_2$ since $D$ can be calculated from $r_{\text{max}}$, the assumption that the shape remains ellipsoidal and the incompressibility of the fluid. In figure 4 we have plotted the numerical and analytical results for the steady state deformation of a drop consisting of a viscoelastic I or II material. For the numerical calculations we used a grid with 16 boundary elements and 128 cells in the internal domain. For all values of $c_1$ and $c_2$ the analytical and the numerical results are in very good agreement. Both models have the same behaviour for small deformations, (i.e. large values of $c_1$ or $c_2$) and differ strongly for large deformations. Especially the viscoelastic II drop shows strong nonlinear behaviour for larger deformation, which is well captured by the numerical method.

From this we can conclude that the method gives correct results for a drop consisting of an Oldroyd-B fluid or a viscoelastic material. We also find that the method is capable of dealing with large deformations and strong nonlinear behaviour of the non-Newtonian stress tensor. In the next subsection we will concentrate on a drop composed of two layers.

### 4.2 Double layer non-Newtonian drop

In this subsection we present some simulation results for a drop consisting of two layers of material. The internal layer $V^{(2)}$ consists of a viscous fluid with $\lambda^{(2)} = 1$ and the outer layer $V^{(1)}$ of the drop contains a (non-)Newtonian material. At the present moment only analytical results for a drop where $V^{(2)}$ consists of a viscous or a viscoelastic material are known to the authors. For this reason we restrict ourselves to these two situations and do not discuss a drop where $V^{(2)}$ contains an Oldroyd-B fluid, although the method is perfectly suitable for this situation as well.
As for the single layer drop we first turn our attention to a typical example of a double layer drop. In figure 5 the steady state velocity field and grid of a viscous drop with $C^{(1)} = C^{(2)} = 0.025$ and $\lambda^{(1)} = 1$ is plotted. In the second layer two vortices are present which are also drawn schematically in the lower part of the figure. It is seen that these vortices deform the inner interface in a direction opposite to the deformation of the outer interface. Due to this mechanism the layer $V^{(1)}$ is thicker at the tops of the drop and thinner in the middle part of the drop. Stone and Leal [11] suggest that a contact of the interfaces leads to breakup of the drop. The steady state deformation of a drop where $V^{(1)}$ consist of a viscoelastic I material with $c^{(1)}_1 = 80$ differs substantially from a viscous drop as can been seen in figure 6. In this figure no velocity field is plotted as it is zero in the steady state situation. Due to the elastic nature of the material there is almost no variation of the thickness of the layer $V^{(1)}$, and hence breakup is not caused by contact of the two interfaces. At this moment a breakup mechanism based on critical stresses in the viscoelastic layer is developed. Results concerning this mechanism will be published in the near future.

Using grid refinement it is found that the order of accuracy for the double layer drop is the same as for the single layer drop, i.e. second order in space and first or second order in time, depending on the time integration method used. To verify the results for a drop consisting of two viscous layers we use small deformation results obtained by Stone and Leal [11]. The theory leading to these results is based on the work of Taylor [7] for single layer drops and shows strong resemblance with the theories discussed in the previous subsection. In this case we obtain expressions for the amplitude of the radial displacement of both interfaces from which the deformation can be derived. In figure 7 we plotted the analytical and numerical steady state deformation of both interfaces as a function of the initial shell thickness $\kappa^{(1)}$ at $C = 0.025$. The numerical results were generated with 32 boundary elements along both interfaces and no internal cells since there is no domain integral present in this particular case. In the figure we have also plotted the steady state shapes of the drop for three different values of the thickness $\kappa^{(1)}$. The numerical and analytical results are in excellent agreement for small deformations. For larger deformations, i.e. smaller $\kappa^{(1)}$, there are some differences which can be explained by the limited validity of the linear theory. The shapes in this figure clearly show that breakup can already occur for very small deformations of the outer interface, as was also remarked by Stone and Leal [11].

The results for a drop consisting of a viscous inner layer and a viscoelastic I outer layer are verified using analytical first order results by Brunn [5]. In this theory the Navier-Stokes and Navier equations are used to describe the displacement fields of the viscous and elastic layer respectively. The derivation of the amplitude of the radial displacement of both interfaces is analogous to the theories described above. In figure 8 we have plotted the analytical and numerical results for the steady state deformation as a function of the initial thickness of the viscoelastic I layer with $c^{(1)}_1 = 80$. In order to keep the aspect ratio of the internal cells limited, we used different grids for different values of $\kappa^{(1)}$ (e.g. $N^{(1)}_{cb} = N^{(2)}_{cb} = 32$, $N^{(1)}_{cc} = 160$ for $\kappa^{(1)} = 0.1$ and $N^{(1)}_{cb} = N^{(2)}_{cb} = 16$, $N^{(1)}_{cc} = 256$ for $\kappa^{(1)} = 0.9$). It is seen, similar to a completely viscous double layer drop, that the numerical and analytical results are in good agreement for small deformations. Additional computations which were performed for higher values of $c^{(1)}_1$ (i.e. smaller deformations) lead to smaller differences at $\kappa^{(1)} = 0.1$ indicating that the discrepancies are due to the limited validity of the linear theory. In figure 8 we also plotted the shapes of the drop for several values of $\kappa^{(1)}$. From these plots it is seen that the interfaces deform uniformly for all $\kappa^{(1)}$.
From this we can conclude that the method developed in sections 2 and 3 gives correct results for a double layered (non-)Newtonian drop. We also find very interesting results concerning the thickness of the outer layer pointing towards essentially different breakup mechanisms for different composing materials. This will be studied in more detail in the near future.

5 Conclusions

In this paper a boundary integral method for axisymmetric multilayered non-Newtonian drops immersed in a viscous fluid subjected to an axisymmetric flow was presented. The non-Newtonian contribution was treated as a source term, leading to a domain integral in the boundary integral representation of the solution. By transforming the integral representation for the velocity to cylindrical coordinates we can reduce the dimension of the computational problem. The integral equation for the velocity remains of the same form and the Green’s functions are transformed explicitly to cylindrical coordinates. Simulations show that the numerical method which was developed is second order accurate in space and first or second order accurate time depending on the time integration scheme used.

The numerical results for a single and double layered (non-)Newtonian drop have been compared with analytical results known from literature. The time-dependent behaviour of a single layer Oldroyd-B drop is in good agreement with analytical results for small values of the relaxation time (Delaby et al. [38]). For a drop containing an elastic material we examined the steady state behaviour. It was found that the numerical and analytical results are in excellent agreement with each other and that strong nonlinear behaviour of the drop is well captured by the method. The numerical results for the steady state behaviour of a double layer drop consisting of a viscous inner layer and a viscous or viscoelastic outer layer is in good agreement with analytical results. The thickness of a viscous outer layer varies strongly along the interface whereas the thickness of a viscoelastic layer remains almost uniform. This implies that the breakup mechanism due to contact of the interfaces is unlikely to be relevant for viscoelastic layers. In this case the introduction of a critical breakup stress in the viscoelastic layer seems a more plausible mechanism. This will be studied in the future.

The boundary integral method developed in this paper is well suited for compound non-Newtonian drops although computational times are much longer than in the Newtonian case due to the domain integral that appears in the formulation. The advantage of the method over a more direct (finite difference) discretization of the Stokes equations lies in the fact that only the non-Newtonian layers of the drop have to be discretized and that calculations using relatively few points already give very accurate results. In case of thin layers the advantages of the method can be fully exploited since only small part of the total flow domain has to be discretized.

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References


Figure Captions

Figure 1: Schematic illustration of the different domains and interfaces.

Figure 2: The velocity profile and grid of a single layer viscous drop with $\lambda^{(1)} = 1$ at $C^{(1)} = 0.1$. In the lower part of the figure the vortices which are present in the velocity profile are drawn schematically.

Figure 3: The deformation of an Oldroyd-B drop at $C^{(1)} = 0.025$ for three values of the relaxation time $\mu_p^{(1)}$. The viscosity ratio $\lambda_p^{(1)}$ is kept constant at 5. The dash-dotted, dashed and solid lines represent the analytical results for $\mu_p^{(1)} = 1, 0.25$ and 0.0625 respectively. The circles, pluses and asterisks display the corresponding numerical results.

Figure 4: Steady state deformation of a viscoelastic drop with $\lambda^{(1)} = 1$ for a constant value of either $c_1^{(1)}$ or $c_2^{(1)}$. The solid and dashed lines represent the analytical results for $c_2^{(1)} = 0$ and $c_1^{(1)} = 0$ respectively. The asterisks and circles display the corresponding numerical results.

Figure 5: The velocity profile and grid of a double layer viscous drop with $\lambda^{(1)} = \lambda^{(2)} = 1$. The initial thickness of the outer interface is 0.2 and the capillary number at both interfaces is equal to 0.1. In the lower part of the figure the vortices which are present in the outer viscous layer are drawn schematically.

Figure 6: The grid of a two layer drop with a viscous ($\lambda^{(1)} = 1$) inner and a visco-elastic outer layer ($\lambda^{(1)} = 1$, $c_1^{(1)} = 80$ and $c_2^{(1)} = 0$). The initial thickness of the outer interface is 0.2.

Figure 7: Steady state deformation of a two layer viscous drop with $\lambda^{(1)} = \lambda^{(2)} = 1$ at $C^{(1)} = 0.025$ versus the initial interface thickness $\kappa^{(1)}$. The circles and crosses represent the numerically obtained deformations of the inner and outer interface respectively. The solid and dashed lines display the corresponding analytical results. The steady state configurations for $\kappa^{(1)} = 0.1$, $\kappa^{(1)} = 0.5$ and $\kappa^{(1)} = 0.9$ are also contained in the plot.

Figure 8: Steady state deformation of a two layer drop with a viscous interior $\lambda^{(2)} = 1$ and a viscoelastic ($\lambda^{(1)} = 1$, $c_1^{(1)} = 80$ and $c_2^{(1)} = 0$) outer layer at $C^{(1)} = 0.025$ versus the initial interface thickness $\kappa^{(1)}$. The circles and crosses represent the numerically obtained deformations of the inner and outer interface respectively. The solid and dashed lines display the corresponding analytical results. The steady state configurations for $\kappa^{(1)} = 0.1$, $\kappa^{(1)} = 0.5$ and $\kappa^{(1)} = 0.9$ are also contained in the plot.
Figure 2.
Figure 4:

\[ G = c_{1,2} \]

\[ D = \frac{\eta G}{c_{1,2}} \]
Figure 6:
Table 5: Grid refinement results of the deformation of an Oldroyd-B drop ($\lambda_p^{(1)} = 5$ and $\mu_p^{(1)} = 1$) at $t_0 = 1$ for two different capillary numbers ($C^{(1)} = 0.01$ and $C^{(1)} = 0.1$).

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