Journal of Non-Newtonian Fluid Mechanics 240 (2017) 1–14

Contents lists available at ScienceDirect

Journal of Non-Newtonian Fluid Mechanics

journal homepage: www.elsevier.com/locate/jnnfm

Dynamics of spreading thixotropic droplets☆

A.S. Uppal[a,*], R.V. Craster[b], O.K. Matar[a]

[a] *Department of Chemical Engineering, Imperial College, London*
[b] *Department of Mathematics, Imperial College, London*

A R T I C L E   I N F O

Article history:
Received 17 October 2016
Revised 4 January 2017
Accepted 7 January 2017
Available online 9 January 2017

Keywords:
Thixotropy
Lubrication theory
Droplets
Spreading

A B S T R A C T

The effect of thixotropy on the two-dimensional spreading of a sessile drop is modelled using lubrication theory. Thixotropy is incorporated by the inclusion of a structure parameter, λ, measuring structure build-up governed by an evolution equation linked to the droplet micromechanics. A number of models are derived for λ coupled to the interface dynamics; these range from models that account for the cross-stream dependence of λ to simpler ones in which this dependence is prescribed through appropriate closures. Numerical solution of the governing equations show that thixotropy has a profound effect on the spreading characteristics; the long-time spreading dynamics, however, are shown to be independent of the initial structural state of the droplet. We also compare the predictions of the various models and determine the range of system parameters over which the simple models provide sufficiently good approximations of the full, two-dimensional spreading dynamics.

1. Introduction

Thixotropy is of central importance in a variety of applications, due to its presence in a wide range of fluids, which include natural muds, slurries, clay suspensions, greases, paints, gels and adhesives [1]. The mechanism underlying thixotropy is normally attributed to the interactions of polymers, particles and colloids, for instance, within the fluid capable of forming a microstructure [2]; the evolving microstructure modifies the internal stress of the fluid and consequently alters the rheological response. While in this paper, we will model thixotropy by the direct inclusion of a structure parameter, it has been shown that thixotropy and yield stress behaviour can be the natural limit of viscoelastic behaviour, when the relaxation time is large, [3–5]. Structure parameter models can be seen to be a natural extension of viscoelastic thixotropy models when you take the structure parameter to be the trace of the conformation tensor. Thixotropy can have a dramatic effect upon the flow behaviour as exemplified by the chaotic regimes observed in numerical studies of a highly thixotropic fluid displaced by a Newtonian fluid [6]. Similarly, fingering instabilities are seen to grow exponentially, rather than algebraically during the injection of a thixotropic fluid into a porous medium [7]. Additional flow regimes have also been found when gravity-driven flows of thixotropic fluids have been studied [8–10] Finally, in capillary-driven levelling of a thixotropic fluid [11], large variations in viscosity across a fluid layer were measured. In the present work, we will restrict our analysis to capillary-driven flows where inertia is negligible.

The aforementioned studies have all used the lubrication approximation as a key simplification which exploits the naturally occurring small aspect ratio; this permits solution for the depth dependence of the velocity field, and the derivation of an evolution equation for the interface [12]. It is not possible, however, to remove the depth dependence completely in the presence of thixotropy using lubrication theory. Typically, one is left with a so-called “1.5D model” [13] characterised by a one-dimensional (1D) equation for the interface coupled to a two-dimensional equation for the structure parameter, that must be integrated over the depth. Additional simplifications have been proposed to reduce this further through an averaged structure [10] or ‘fluidity’ [7]. This is further built upon by Livescu et al. [14] where a depth profile is assumed for the fluidity that is then linked to the structure parameter at the substrate and the interface, yielding three 1D evolution equations. Alternatively, one can assume a depth profile for the structure parameter in the form of a polynomial [15]. Furthermore, an asymptotic approach is considered by Pritchard et al. [16] where the thixotropic properties are considered weak and enter at higher order. Our goal here is to solve a typical problem of interest, the evolution of a spreading droplet, and then assess which, if any, of the above simplifying approximations are appropriate via com-

☆ This document is a collaborative effort.

* Corresponding author.
E-mail address: a.uppal14@imperial.ac.uk (A.S. Uppal).

http://dx.doi.org/10.1016/j.jnnfm.2017.01.001
0377-0257/

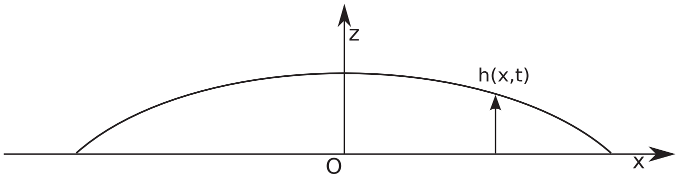

Fig. 1. Schematic illustration of the coordinate system used to model the spreading.

parisons with the solution of the 1.5D model through a parametric study. It is noted by Pritchard et al. [16] that reduced models can be insufficient for confined thixotropic flows, due to the presence of strong transverse gradients in the microstructure. However, this may not hold true for unconfined flows with free surfaces. In this present work, we seek to verify the validity of reduced models with respect to free-surface flows in the presence of moving contact lines.

The rest of this paper is organised as follows. In Section 2, we provide details of the problem formulation, in which we set out the rheological model under consideration via a microstructure derivation of the model. We also derive the governing 1.5D model using the lubrication approximation, and demonstrate its connection to simpler models. In Section 3, we discuss our numerical results, focusing on the comparison of the predictions provided by the various models derived in Section 2. Finally, concluding remarks are provided in Section 4.

2. Formulation

2.1. Governing equations

We consider a slender droplet of density ρ and viscosity $\hat{\mu}$ lying on a horizontal, rigid and impermeable substrate; the overlying gas phase is assumed to be hydrodynamically passive and its dynamics are neglected. We use a Cartesian coordinate system $(\hat{x}, \hat{z})$ with $\hat{x}$ and $\hat{z}$ orientated parallel and normal to the substrate, respectively and with origin at the centreline of the droplet, such that the interface is given by $\hat{z} = \hat{h}(\hat{x}, \hat{t})$, as shown in Fig. 1. The velocity is given by $\hat{\mathbf{u}} = (\hat{u}, \hat{w})$, where $\hat{u}$ and $\hat{w}$ are the components in the $\hat{x}, \hat{z}$ directions, respectively; the hat decoration designates dimensional quantities.

We neglect inertial and gravitational forces such that the governing equations are given by

$$\hat{u}_{\hat{x}} + \hat{w}_{\hat{z}} = 0, \tag{1}$$

$$\hat{\nabla}\hat{p} = \hat{\nabla} \cdot \hat{\tau}, \tag{2}$$

where the stress tensor is expressed by $\hat{\tau}_{ij} = \hat{\mu}(\lambda)\hat{\dot{\gamma}}_{ij}$, $\hat{p}$ denotes the pressure, the rate of strain tensor is given by $\hat{\dot{\gamma}}_{ij} = \partial_j \hat{u}_i + \partial_i \hat{u}_j$, and $|\hat{\dot{\gamma}}| = \sqrt{\hat{\dot{\gamma}}_{ij}\hat{\dot{\gamma}}_{ij}}$ is the second invariant of $\hat{\dot{\gamma}}_{ij}$. Crucially, we assume that the viscosity depends on a dimensionless structure parameter λ that describes the evolving structure within the fluid. We enforce no-slip and no-penetration conditions at the substrate, $\hat{u} = \hat{w} = 0$ at $\hat{z} = 0$ and the kinematic and stress boundary condi-

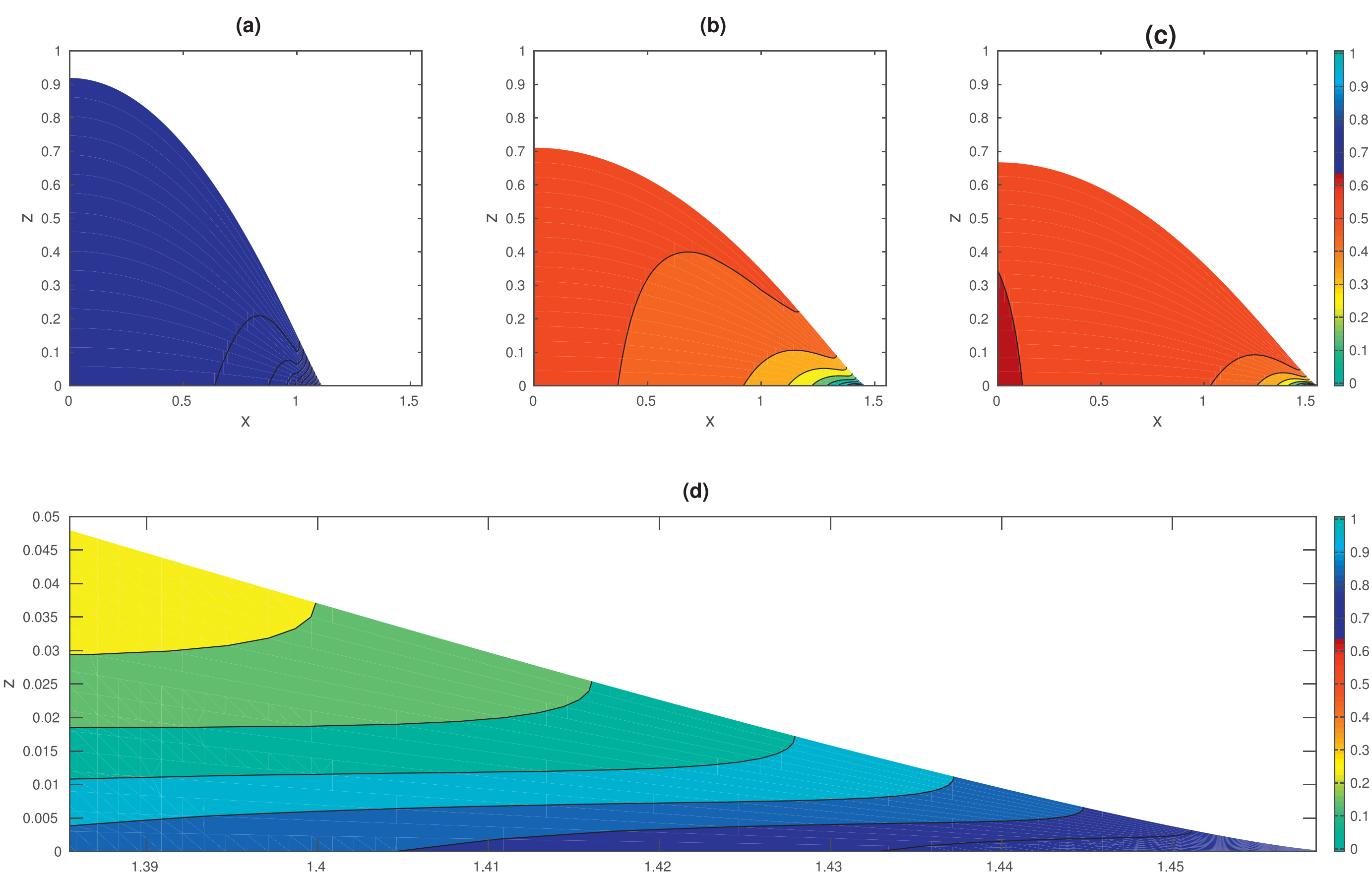

Fig. 2. Results from the numerical solution of Eqs. (20) and (21), computed with $\Lambda = 0$, $B = 5$, $D_b = 10$, and $\delta = 9$: droplet evolution for $t = 1, 25, 50$ shown in (a)-(c), respectively. Panel (d) shows a zoomed in view of the contact line at $t = 25$. In this and subsequent figures, the colour reflects the degree of structure build-up, where red and blue represent high and low values of λ, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)