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To Anne-Claire, Ofelia and Françoise
Preface

Over the last decades, an increasing number of functional and structural parts, made so far with metals, has been progressively reengineered by replacing metallic materials by polymers, reinforced polymers, and composites. The motivation for this substitution may be the weight reduction, the simpler, cheaper or faster forming process, or the ability to exploit additional functionalities.

The fillers usually employed cover a broad range involving many scales: (i) the nanometer scale (e.g., carbon nanotubes, graphene, fullerene, nanodiamonds); (ii) the micrometer to the millimeter scale (particles and short fibers); (iii) the centimeter scale of fibers used in SMC and BMC composite processes; and finally (iv) the macroscopic scale where fibrous reinforcements are made of continuous fibers arranged in bundles.

When load-bearing capacities are especially looked for, continuous fiber reinforced polymers are selected. In that case, the impregnation of the reinforcement with a low viscosity polymer involves the flow of a Newtonian or non-Newtonian fluid in the complex multi-scale microstructure related to the fiber and tow arrangement. Reinforced polymers are selected instead of high performance polymers of equivalent properties since the latter are generally more expensive.

When looking for functional properties, the use of nano-charges opens a wide spectrum of possibilities but also raises new challenges, such as dispersion of charges into the polymer matrix and occurrence of aggregation and disaggregation mechanisms. Suspensions of practical interest involve many scales and many concentration regimes, the latter ranging from dilute to highly concentrated.

In the present monograph, we survey modern developments related to the multi-scale modeling and simulation of polymers (Chap. 1, by Roland Keunings), reinforced polymers that involve a flowing microstructure (Chap. 2, by Francisco Chinesta), and continuous fiber-reinforced composites, wherein the fluid flows inside a nearly stationary multi-scale microstructure (Chap. 3, by Christophe Binetruy).

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Chapter 1
Multi-scale Modeling and Simulation of Polymer Flow

Abstract The flow of polymeric fluids in complex geometries relevant to processing applications can be simulated numerically using a wide variety of theoretical models. Simple mathematical models have a purely macroscopic nature and focus only on the non-linear relationship between shear viscosity and shear rate. More advanced models address the viscoelastic character of polymeric fluids, either in a macroscopic or multi-scale framework. These advanced models and the related numerical approaches are the subject of this first chapter, wherein we build upon and update our previous reviews of the field [29, 31].

1.1 Introduction

1.1.1 Rheology and Structure

Polymer solutions and melts, like other rheologically-complex fluids, exhibit a variety of non-Newtonian flow properties [3, 13, 49, 52]. A first departure from Newtonian behaviour is that the shear viscosity of most polymeric fluids is not a constant material property (at a given temperature), but rather a non-linear function of the shear rate. This property can have dramatic effects in shear-dominated flows, such as flows in cylindrical pipes. A wide variety of macroscopic phenomenological constitutive equations are available that offer a very good fit, with few material parameters, of the non-Newtonian shear viscosity of polymeric fluids [3]. In combination with the conservation principles of continuum mechanics (for mass, linear momentum and energy), these simple constitutive models yield generalized Navier-Stokes equations that can be solved numerically with rather obvious extensions of well-known techniques for Newtonian fluids [52]. It is much less obvious to take account of the many other non-Newtonian properties of polymeric liquids related in general to memory effects. Indeed, polymeric liquids are viscoelastic materials, meaning that the stress endured by a fluid element depends upon the history of the deformation experienced.
by that element. Viscoelastic properties include non-linear shear viscosities, but also the presence of normal stresses in viscometric flows, high resistance to elongational deformation, and various memory effects (such as stress relaxation) related to the elasticity of the material. They are responsible for numerous flow phenomena of scientific and industrial relevance [5] that need to be predicted, understood, and possibly controlled by means of a combination of suitable physical models and numerical techniques.

The rheological properties of viscoelastic liquids are governed by the flow-induced evolution of their internal microstructure [18, 19, 34]. In flowing polymers, the relevant microstructure is the conformation of the macromolecules, namely their orientation and degree of stretch. Each macroscopic fluid element contains a large number of polymers with a statistical distribution of conformations. While the flow alters the polymer conformations along the fluid trajectories, the macroscopic stress carried by each material element is itself governed by the distribution of polymer conformations within that element (Fig. 1.1).

Furthermore, the frozen-in microstructure which develops in processing flows dictates the physical properties of the final product. One thus faces a challenging non-linear coupling between flow-induced evolution of molecular configurations, macroscopic rheological response, flow parameters (such as geometry and boundary conditions), and final product properties. Theoretical modeling and methods of computational rheology are needed to elucidate this coupling. Clearly, the purely macroscopic approach of continuum mechanics will not be able to fully address the problem at hand. Rather, a suitable multi-scale approach is called for.

1.1.2 A Hierarchy of Modeling Approaches

Modern research in computational materials science exploits, sometimes in a coupled fashion, a hierarchy of theoretical models ranging from quantum mechanics to continuum mechanics. The difficulty with polymers is of course the gigantic number of microstructural degrees of freedom and the broad range of time and length scales separating the relevant atomistic and macroscopic processes [41]. A modeling approach based on quantum mechanics and related ab initio computational techniques must
1.1 Introduction

clearly be ruled out at the outset. Indeed, atomistic modeling is probably the most detailed level of description that could realistically be applied in rheological studies, by means of techniques of non-equilibrium molecular dynamics (e.g. [51]).

Atomistic flow simulations have been conducted to study the behaviour of polymers near walls and geometrical singularities such as re-entrant corners (e.g. [7, 10, 33]). In view of the significant computer resources involved in such calculations, the atomistic approach is currently restricted to very coarse models for the polymer and to flow geometries of molecular dimensions. While it is doubtful that it may be feasible soon (and even necessary, for that matter!) to solve flow problems of macroscopic size using the atomistic approach, the potential of atomistic simulations is great, however, in helping us resolve difficult issues such as wall slip. Consideration of macroscopic flows found in processing applications calls for less detailed mesoscopic models, such as those of kinetic theory.

The next level of description of a polymeric fluid is that of kinetic theory [4, 14]. Here, one ignores atomistic processes altogether and focuses rather on the evolution of a more or less coarse-grained model of the polymer conformations. For example, a linear polymer chain in a viscous solvent can be described by several models of decreasing complexity: the freely jointed, bead-rod Kramers chain made of a number of beads (of order 100) connected linearly by rigid segments, the bead-spring chain made of a smaller number of beads (of order 10) connected by entropic springs, or the single dumbbell, namely two beads connected by a spring. Clearly, these models of kinetic theory are not meant to describe the chemical structure of the polymer. They do, however, display in a more or less detailed fashion the important features needed to describe the evolution of polymer conformations in a macroscopic flow. Kinetic theory models for polymer solutions or melts are most naturally exploited numerically by means of stochastic simulation or Brownian dynamics methods [44], but a powerful alternative method known as the Proper Generalized Decomposition (PGD) has been developed recently [9], as we shall discuss below. Use of a model of kinetic theory in the numerical simulation of complex flows, in combination with the macroscopic conservation laws, constitutes the so-called micro-macro approach to computational rheology. This emerging methodology is the main theme of this chapter.

Finally, besides atomistic modeling and kinetic theory lies the macroscopic approach of continuum mechanics [3, 52]. Here, details of the fluid microstructure are not taken into account, at least explicitly. Rather, the stress experienced by the macroscopic fluid elements is related to the deformation history through a suitable constitutive equation. Combined with the conservation laws, the constitutive model yields a set of partial differential (or integro-differential) equations that can be solved by means of a suitable grid-based numerical method, such as the finite element technique. The vast majority of research publications in computational rheology has followed the macroscopic approach [29, 45, 52]. Very much like atomistic modeling and kinetic theory, continuum mechanics offers a hierarchy of constitutive models of diverse predictive ability. It should be stressed that most constitutive equations