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Slip Mechanisms in Complex Fluid Flows

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Abstract

The classical no-slip boundary condition of Fluid Mechanics is not always a valid assumption for the flow of several classes of complex fluids including polymer melts, their blends, polymers solutions, microgels, glasses, suspensions and pastes. In fact, it appears that slip effects in these systems is the rule and not the exemption. The occurrence of slip complicates the analysis of rheological data, although it provides new opportunities to understand their behavior in restricted environments delineating additional molecular mechanisms i.e. entropic restrictions due to limitations in the number of molecular conformations. This article discusses these complexities and provides future research opportunities.

I. Introduction

Slip has been considered by several scientists in the early stages of development of fluid mechanics including Bernoulli, Coulomb, Poiseuille, Girard, Maxwell, Navier and Stokes are among those who considered the possible effects of slip in Newtonian fluid dynamics. Simply experimental observations were found consistent with the assumption of no-slip and therefore slip phenomena have received little further attention (see [1] for a summary).

Unlike Newtonian fluids, several classes of complex exhibit phenomena inconsistent with the assumption of no-slip, including polymer melts [2-32], elastomers [33-34], polymer solutions [35-39], suspensions [40-52], dispersions [53-55], gels [56-62], colloidal dispersions/glasses [63-65], pastes [66-80] and foams [81-84]. Excellent reviews can be found in [25, 85-88] for polymer melts, in [74] for suspension and pasty materials, in [89] for polymer solutions, emulsions and particle suspensions and in [90] for general complex fluids. Slip plays a significant role in correctly determining the rheology of polymers by correcting the data for slip effects [11, 25] and explaining mismatch of rheological data obtained from various rheometers utilizing different geometries [25, 88]. Interfacial (slip) constitutive laws are also needed to simulate these flows either from a macroscopic [40, 41, 62, 63, 66, 69, 72, 75, 77, 91-99], microscopic [59, 60, 100-106] and/or molecular point of view [100-112].

This article discusses a number of important issues related to flows of several complex systems and in particular the behavior of these systems at solid boundaries; (i) molecular mechanisms of slip in representative complex fluids including polymer melts, solutions, suspensions and gels; (ii) molecular mechanisms of slip in the case of immiscible and/or partially miscible/immiscible polymer blends, where depending on the relative magnitudes of the three interfacial tensions (two polymer components and the solid wall), the wall might be preferentially coated by one of the components, thus providing lubrication [113-120]; (iii) migration effects of polymer molecules of entropic origin complicates the analysis of slip data in the case of polymer melts and solutions as well as in other cases such as suspensions and emulsions where concentration gradients might form due to thermodynamically driven particle motions [30, 112, 121-124]; (iv) complications arising in deriving/developing interfacial rheological laws for transient flows where relaxation effects are to be considered. In these cases the relaxation times of molecules and/or network of particles (suspensions and gels) are much shorter arising from conformation restrictions due to the presence of the wall [91, 93, 98]; (v) the slip behavior in the presence of wall roughness [17, 125-127] or the presence of low surface energy coatings (fluoropolymer-based coatings) [127-137]. In particular, the different behavior over solid surfaces with macroscopic roughness versus micro/nano patterned roughness (dual scale) that renders surfaces superhydrophobic (lotus effect). In the latter case, the length scales of dual scale roughness might be of the same order with that of particles or even length scales of polymer molecules; (vi) slip at polymer-polymer interfaces [138-139].

Moreover another goal of this paper is to raise awareness that the rheometry of complex fluids is not straightforward as it might seem, particularly in the nonlinear flow regime, where the high levels of stresses cause complexities inconsistent with the classical no-slip boundary condition of fluid mechanics.

II. Polymer Melts

Several studies have considered slip in flows of linear polymer melts including high-density polyethylene (HDPE) [2, 6, 7, 12-14, 22, 24, 29, 30], linear low-density polyethylene (LLDPE) [3, 4], fluoropolymer (FEP) [116, 132], polyisoprene (PI) [140], polybutadiene (PBD) [5, 16-18, 28, 31, 32, 141], polydimethylsiloxane (PDMS) [9, 10, 142-143], polystyrene (PS) [11, 19], polycarbonate (PC) [144], polyisobutelene (PIB) [20, 145], polypropylene (PP) [26, 94, 146, 147], poly(ethylene-vinyl) acetate) (PEVA) [148], polyvinylchloride (PVC) [151-153], polyethylene-propylenediene-monomer (EPDM) and other rubbers/elastomers [33-35, 152].

Regarding the mechanism of slip in the case of polymer melts, everything occurs within the first monolayer of macromolecules adsorbed at the wall as seen in Figure 1. These chains form loops and they are adsorbed on the wall at several sites along their backbone (Figure 1a). The adsorbed chains are also entangled with chains in the bulk again at several sites also shown in Figure 1a. Under the influence of flow, the following possibilities exist: (i) chain detachment/desorption of the chains adsorbed at the wall that leads directly to weak slip. This is referred to as true/adhesive slip. This is the case when low surface energy coating are on the surface such as fluoropolymers (fluoropolymer-based coatings) or other low surface energy polymer-based coatings [117, 128-130, 135-137]; (ii) partial chain disentanglement of the bulk from chains, which are strongly adsorbed on the wall that leads also to weak slip. This is referred to as cohesive slip. This is the most usual mechanism of slip in flows of polymer melts over the metallic surfaces and it is of most interest to polymer processing [2, 3, 7, 16-18, 22, 29, 30-32]. In this case interfacial rheological (slip) laws are needed to simulate these processes [91-106]; (iii) under strong flow and due to orientation of chain loops adsorbed on the wall towards the directions of flow, sudden disentanglement occurs that leads to a transition from weak slip to strong slip (nearly plug flow) and therefore to a discontinuous flow curve [14, 29].

The slip of polymer molecules is influenced by a number of factors:

Temperature and pressure dependency: Many reports study the effects of temperature (consistent with the time-temperature superposition) [6, 7, 14, 29] and pressure [4, 7, 102, 103] on slip velocity. In fact, it has been found that slip increases with increase of temperature [4, 6, 7, 29] and decreases with increase of pressure [4, 7, 103].

Molecular weight dependence: The slip velocity [V_S] also increases with decrease of molecular weight, M_n ($V_S \propto M_n^{-a}$, where the exponent "*a*" has been found to be about 2 for PBDs) [16, 99], 2 for HDPEs [99] and about 3.2 for PSs [21, 99]. Using a double reptation, Ebrahimi et al [99] have proposed a method to calculate the slip of polydisperse polymers solely based on the slip of their monodisperse counterparts, consistent with experimental findings [99]. More work is required in this area for other polymeric systems to establish these slip relations, scaling laws and methodologies.

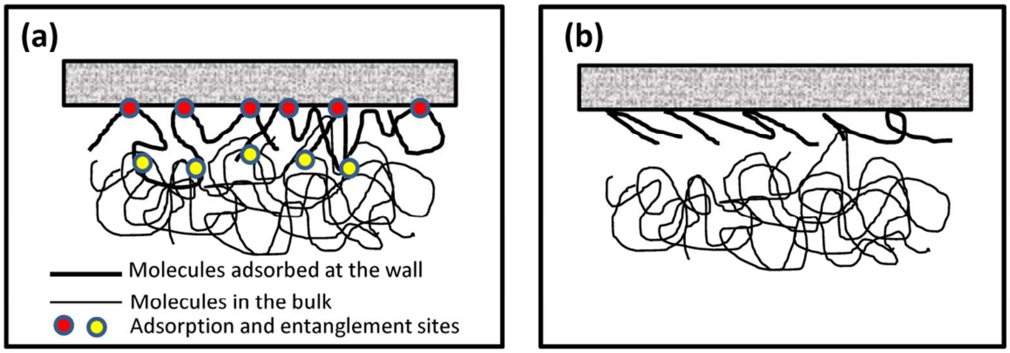

Figure 1: Mechanisms of slip in polymer melts (a) weak slip either by desorption/detachment of polymer chains off the wall (adhesive slip) or by partial disentanglement of the bulk from the molecular layer strongly adsorbed to the wall (b) strong slip (complete disentanglement of the bulk from the molecular layer strongly adsorbed to the wall)

Molecular architecture (Long chain Branching Effects): The presence of long chain branching at least in the case of polyethylenes suppresses the transition from partial slip to strong slip [153-156]. Similarly, linear polymers of significantly wide molecular weight distribution do not exhibit this transition [154, 155]. As a result, the flow curve of such polymers is a continuous rather than a discontinuous (two-branched) one. It is more striking that this transition is obtained in polymer flows in capillary and slit dies but not in annular ones, still an open question in the literature [157].

Migration/Fractionation Effects: In pressure driven flows of broad molecular weight distribution polymers migration effects occur that complicate the slip analysis significantly [30, 112, 121-124]. It has been reported that the molecular fractionation along the radius of a capillary could occur in the extrusion of polymer melts [121], with the molecular weights of the surface sections of the extrudates were lower than those of the whole extrudates [122, 123]. While flow-induced fractionation may be difficult to occur over the finite distance of a capillary die, flow-induced diffusion or molecular fractionation over a small distance from the solid wall might be possibly happening [30]. The origin of this fractionation is of entropic origin (minimization of Gibbs free energy) as molecular dynamics simulations have recently shown [112].

Interfacial conditions (roughness): Surface roughness has a profound effect on slip by decreasing it and delaying the transition from weak slip to strong slip to higher rates [17, 125-127]. Typical roughness of length scales used in these studies are of a few μm, which are much larger than the coil size of a typical polymer chain, which is of the order of a few nm. Therefore many chains can be trapped in the space between the asperities and thus the polymer/wall interface is replaced with corresponding polymer/polymer interface. It would be interesting to see how slip is affected in the presence of well-defined asperities of pitch size (distance between asperities) similar to molecular coil lengths. In this case molecules cannot diffuse in the space between the asperities due to steric effects and significant portion of the polymer/wall interface might be replaced by corresponding polymer/air interface. Such technologies exist to manufacture such substrates in order to perform experiments [158, 159].

Dependence on coatings (low surface energy): The presence of low surface energy coating suppresses polymer adsorption and slip becomes easier. Many studies exist and in most cases fluoropolymer-based coatings are used [127-137]. In general the magnitude of the slip velocity in these cases scales with the work of adhesion of these interfaces [103, 128, 129, 133, 134].

Flow of immiscible blends with one component at small amounts (<1 wt%): Two component immiscible blends with one component in proportion of less than 1 wt%, have been observed to slip significantly [113-120]. Typical examples of such systems are: EPDM/Viton(fluoropolymer) [115], polyethylene/fluoropolymer [116], polylactide (PLA)/polycaprolactone (PCL) [119]. In these cases the minor components gradually coats the wall with the major component slipping over this layer due to the incompatibility of the two components (absence of adsorption sites which otherwise are present in flow of polymers over metallic surfaces). It has been suggested that for this to occur, the interfacial tension between the two polymeric components should be less that the interfacial tension between either component with the metallic wall that implies a competition with interfacial forces [116, 117, 119].

III. Polymer Solutions and Suspensions

Polymer solutions exhibit phenomena that are incompatible with the no-slip boundary condition of fluid mechanics [35-39]. Similar effects are obtained in the case of suspensions [40-52] and dispersions [53-55]. A good review for the case of polymer solutions can be found in [36-38, 74] and for suspension in [74, 89]. This is due to a very thin low-viscosity layer (slip or depletion layer) near the wall that is formed due to migration of the solute (polymer solutions) or solid particles (suspensions) away from the wall that is of entropic origin (minimization of Gibbs free energy). Figure 2 shows a possible velocity profile for tube flow of a polymer solution and/or suspension. The slip layer may vary between 0.1 to 2 μm or higher, depending on the concentration and particle size. This type of slip is referred to as apparent slip.

For the case of polymer solutions, the slip velocity has been reported to depend on polymer concentration [36-38], polymer adsorption [36], wetting properties of the wall [hydrophobic versus hydrophilic] and type of polymer molecules [37]. For the case of suspensions, the apparent slip depends on temperature [45, 46], concentration [48, 89], particle or fiber size [47, 48, 89] and wetting properties of the wall [74]. Sandblasted, serrated and other roughened walls have been reported to eliminate these effects which are summarized in [53, 74, 89]. More work in this area is needed in particular the development of slip laws that underlying the mechanisms of these phenomena either phenomenologically (practical applications) or theoretically.

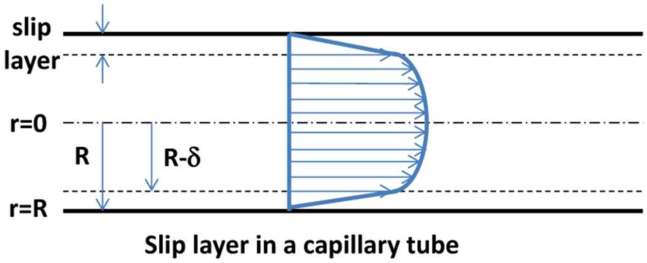

Figure 2: Velocity profile for tube flow of a polymer solution and/or suspension showing the formation of a thin layer (slip or depletion layer) close to the wall having a smaller viscosity due to migration effects.

IV. Gels, Microgels and Colloidal Gels/Glasses

A distinctly different class of complex fluids is the case of gels, microgels (concentrated suspensions of deformable particles) and colloidal gels/glasses that exhibit slip phenomena. In particular, slip has been reported for the cases of a hydroxypropyl guar gel [56], microgels of deformable particles [57-60], skin/hair care gels [61], a viscoplastic hydrogel [62] and gels of colloidal particles [63-65]. Generally speaking most of these systems exhibit a generic slip behavior that is discussed next [56-62].

Meeker et al. [58, 60] have comprehensively studied the slip behavior of a microgel including deformable particles. They have shown that microgels display a generic unusual slip behavior. First, they have shown that in parallel-plate geometry when both plates are rough, slip is suppressed at low stresses (Figure 3a). However, when these materials are sheared on smooth surfaces, they exhibit slip at the wall. Three slip regimes were identified (Figure 3a). (i) at stresses below a yield stress, σ_y the flow is solely due to slip (ii) at stresses, $\sigma_y < \sigma < 1.5\sigma_y$ the flow is due to deformation and slip and (iii) at higher shear stresses, the slip decreases with increase of slip, a counterintuitive observation. They have also developed a clever, non-contact elasto-hydrodynamic slip model that considers various types of short-range interactions (dispersion forces, hydrophilic/ hydrophobic interactions, electrostatic forces) [60]. The slip model seems to describe the slip behavior quite well (Figure 3b). This is referred to as lubricating or hydrodynamic slip illustrated in Figure 3c and it is due to the deformation of particles forming a lubricating layer.

The slip velocity in the case of gels depends on the surface energy of the wall (hydrophobic versus hydrophilic surfaces) [60], roughness [56, 58, 59] and shear history since many of these fluids exhibit thixotropic behavior [61, 56]. Similar slip phenomena have been discussed for the case of colloidal gels/glasses [63-65], where slip has been shown to depend on the concentration of colloidal particles as well as on the polymer concentration present in these systems (adsorbed on the particle surface).

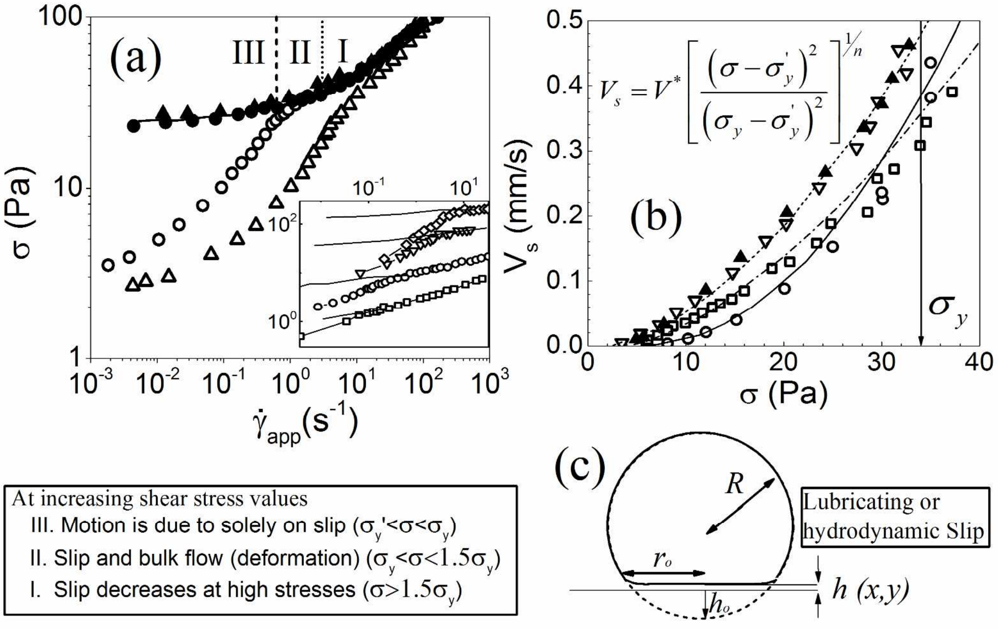

Figure 3: Slip in microgels (a) flow curve of a microgel in parallel plate using rough (filled symbols) and smooth plates (unfilled symbols) where three regimes can be identified summarized below fig 3a (b) slip velocity model fits to experimental data of the microgel studied in [58, 59] (c) mechanism of slip in microgels identified as lubricating or hydrodynamic slip due to fluid layer between the wall and the microgel particle [58, 60].

Pastes

Significant work of slip in the case of paste rheology has been reported [66-80]. Examples include potato pastes [66, 73], starches [68, 69, 70], mayonnaise [67], polytetrafluoroethylene (PTFE) pastes [71, 75, 76, 80], bread dough [77], and ice cream [78, 79] amongst others. The general methodology to study such systems and how to prevent slip can be found in [74]. The general mechanism of slip for these systems is similar to that illustrated in Figure 2 for the case of polymer solutions and suspensions, that is the formation of a depletion layer.

Factors affecting slip in these systems include, the method used to prepare the sample since many of these systems are sensitive, the sample thickness the shear history, surface wettability, surface roughness and lubricant volume fraction [67, 71, 74]. The Mooney technique has been applied to determine slip, which has been found inconsistent in many cases, resulting negative deformation once the rheological data are corrected for slip effects. A modified Mooney technique can be applied in these cases known as the Jastrzebski method that generates slip velocity data that depend on the characteristic dimension of the flow field used i.e. diameter in tube flow. Martin and Wilson [72] have critically assessed this method and they have managed to provide consistency in many sets of data (pastes, foams and polymers) analysed by this method by using a Tikhonov regularization together with the Mooney analysis. They have proposed that further research is required to understand the capillary flow of pastes which does not conform to the classic Mooney analysis. Since the Jastrzebski method lacks physical grounds, it should no longer be used to analyse experimental data to calculate slip [72].

Foams fall into the category of pasty materials [74]. Slip in such systems has been studied to a certain extent [81-84]. Bertola et al., [82] have examined in detailed the slip behaviour of a commercial shaving cream showing that slip is significant for stresses less than its yield stress and decreases or is eliminated at high enough shear stresses (similar to microgels described above). Denkov et al [84] have discussed in detail the slip of several foams and proposed a mechanistic model for the occurrence of slip. It is due to lubricating film that forms between the bubbles of the foam and the solid wall, similar to the lubricating film formed in the case of microgels (Figure 3c).

V. Conclusions

As a concluding remark, one may simple state that “everything slips”. However, different classes of complex fluids slip by a different mechanism, including: (i) adhesive slip of polymer melts on low surface energy walls, (ii) cohesive slip of polymer melts over high surface energy walls (metallic), (iii) apparent slip in the cases of polymer solutions, solid/fiber suspensions, dispersions and several pasty materials by the formation of a slip or depletion layer and (iv) lubricating slip in the case of gels, microgels and colloidal suspension (glasses). From the above discussion it is evident that more systematic work is needed in several areas. Here a few of these areas are listed: (i) dependence of slip of polymer melts on molecular parameters for different classes of polymers other than HDPEs, PSs and PBDs (ii) migration effects observed in the case of broad molecular weight polymer melts and associated slip models to capture such effects in pressure-driven flows (iii) systematic slip studies for several classes of complex fluids to address effects of particle size, concentration and interactions with wetting properties of walls. Appropriate models to describe slip effects in these classes of fluids are also needed.

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