

Non-Newtonian effects on the slip and mobility of a self-propelling active particle

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Janus particles propel themselves by generating concentration gradients along their active surface. This induces a flow near the surface, known as the diffusio-osmotic slip, which propels the particle even in the absence of externally applied concentration gradients. In this work, we study the influence of viscoelasticity and shear-thinning (described by the second-order-fluid and Carreau model, respectively) on the diffusio-osmotic slip on an active surface. Using matched asymptotic expansions, we provide an analytical expression for the modification of slip induced by the non-Newtonian behavior. The results reveal that the modification in slip velocity, arising from polymer elasticity, is proportional to the second tangential derivative of the concentration field. Using the reciprocal theorem, we estimate the influence of this modification on the mobility of a Janus sphere. The current study also has direct implications on the understanding of the transport of complex fluids in diffusio-osmotic pumps.

1. Introduction

Synthetic active particles are micron and submicron sized colloidal particles which can propel themselves along predictable trajectories. The self-propulsion arises from the generation of local concentration gradient at the surface, which is induced by variation in the surface activity such as adsorption or reaction (Anderson 1989; Golestanian *et al.* 2005, 2007; Jülicher & Prost 2009). These active particles play an important role in biomedical research; they offer possible applications in drug-delivery micromachines and controlled studies of microbial infections through bio-sensing (Gao & Wang 2014; Su *et al.* 2019). Furthermore, a suspension of active particles represents a non-equilibrium system which exhibits characteristics such as enhanced fluid mixing in complex fluids (Gomez-Solano *et al.* 2016) and inertia-less turbulence, which are also observed in microbial suspensions and granular matter (Patteson *et al.* 2016). Therefore, understanding such novel self-propelled systems provides insights which extend to a wide range of physical phenomena, making it intriguing from a scientific standpoint.

The notion of self-propulsion at the micro-scale was introduced via exploitation of electrochemical-mechanical transduction mechanism i.e. conversion of electrochemical energy (stored in electrolytes) to power mechanical motion. Paxton *et al.* (2004, 2006) and Fournier-Bidoz *et al.* (2005) fabricated bimetallic micro rods by coating the two halves with platinum and silver. When immersed in an aqueous solution of hydrogen peroxide, the oxidation process generates excess electrons and protons (H^+) at the platinum end. The electrons are transported to the other end through conduction, where they are consumed to reduce hydrogen peroxide. The asymmetric distribution of protons drives the autonomous motion of the micro-rod, which was later called ‘self-electrophoresis’.

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Golestanian *et al.* (2005); Howse *et al.* (2007) studied the second mechanism of self-propulsion: conversion of chemical energy (arising from molecular interactions such as van der Waals) to power the mechanical motion in a non-electrolytic medium. Golestanian *et al.* (2007) proposed the first continuum framework to understand the underlying mechanism which drives the self-propulsion of Janus spheres. On the basis of existing experimental studies (Golestanian *et al.* 2005; Fournier-Bidoz *et al.* 2005), three major assumptions were made: (i) the interaction layer is asymptotically thin in comparison to the particle radius ($\epsilon \ll 1$, where ϵ is the ratio of the thickness of interaction layer to particle radius); (ii) the active (catalytic) surface adsorbs (or desorbs) the solute molecules at a fixed rate and diffusion time scale is much shorter than the reactive time scale; (iii) advective effects are negligible. Borrowing insights from the seminal works of Derjaguin *et al.* (1947), Anderson, Prieve and co-workers (Anderson *et al.* 1982; Anderson 1989), they showed that the interaction between the particle and the solute molecules creates a pressure gradient inside the thin interaction layer. This pressure gradient is balanced by the viscous stresses which is the driving mechanism for self-propulsion. An external variation in solute concentration around the colloid can trigger the pressure gradient which generates a surface slip. Consequently, the freely suspended particle moves in the direction of chemical gradient, known as diffusio-phoresis: a macro-scale motion manifested through an asymmetry at the micro-scale. A Janus sphere (possessing a chemically active cap coated on an inert particle) can create and sustain this pressure gradient as its asymmetrically active surface facilitates a tangential concentration gradient and is therefore also called a self-diffusiophoretic particle.

The continuum description assumes that the solute molecules do not occupy volume and therefore loses its validity for nanometer sized Janus spheres. For such cases, Córdova-Figueroa & Brady (2008); Brady (2011) provided a colloidal perspective to diffusio-phoresis: solute molecule and particle interact both energetically (through hard-sphere and van der Waals forces) and hydrodynamically with each other. Later Sharifi-Mood *et al.* (2013) demonstrated that the results derived by Brady (2011) (for diffusio-phoretic velocity) can be obtained through a continuum description, in the limit of asymptotically small solute molecules. They relaxed the assumption of thin interaction layer and incorporated the effects of an irreversible reaction (characterized by Damkhlér number) at the active surface which was found to dampen the propulsion.

Michelin & Lauga (2014) explored the effects of finite advection on self-propulsion. They found that the particles exhibiting attractive interaction with solute molecules show a maxima in swimming speed at a finite Péclet number ($Pe \sim O(1)$, here Pe is the ratio of diffusive to advective time scales); whereas for repulsive interactions, the swimming speed always reduces with increase in advection. In agreement with Sharifi-Mood *et al.* (2013), they also reported a monotonic decrease in swimming velocity with an increase in Damkhlér number. Utilizing matched asymptotic expansions, they showed that the diffusive flux at the surface of the Janus sphere is equal to that at the outer edge of the interaction layer, provided $\epsilon Pe \ll 1$.

All the aforementioned studies assume the surrounding medium to be Newtonian. Since a majority of potential applications of the synthesized active particles lie in drug-delivery and other areas of biological research (Patteson *et al.* 2016; Su *et al.* 2019), understanding the influence of complex rheology on self-propulsion is essential. There has been limited progress towards this, both experimentally and theoretically. Table 1 summarizes the theoretical studies which analyzed self-propulsion in various regimes. The experimental and theoretical analysis of Gomez-Solano *et al.* (2016); Aragonés *et al.* (2018) demonstrated that the translational swimming of a Janus particle is coupled to the rotational motion in a viscoelastic medium. Recently, Saad & Natale (2019) studied

| Investigation | Regime | Description |
|--|--|---|
| Golestanian <i>et al.</i> (2005, 2007) | $\epsilon \rightarrow 0$ $Da \rightarrow 0; Pe \rightarrow 0$ | Built the first continuum-level framework using the foundation laid by Anderson <i>et al.</i> (1982). |
| Brady (2011) Sharifi-Mood <i>et al.</i> (2013) | $\epsilon \sim O(1)$ $Da \ll 1; Pe \rightarrow 0$ | Bridged continuum and colloidal perspectives. |
| Michelin & Lauga (2014) | $\epsilon \ll 1$ $Da > O(1); Pe > O(1)$ | Studied advective and reactive effects. Showed well-posedness of flux boundary condition at the macro-scale. |
| | | |
| Zhu <i>et al.</i> (2012) De Corato <i>et al.</i> (2015) | $\epsilon \rightarrow 0$; two modes | Swimming kinematics and hydrodynamics of swimmers in complex media. Zhu <i>et al.</i> (2012) $\rightarrow De \sim O(1)$ De Corato <i>et al.</i> (2015) $\rightarrow De \ll 1$ |
| Datt <i>et al.</i> (2015, 2017) Pietrzyk <i>et al.</i> (2019) | $\epsilon \rightarrow 0$ $\chi \ll 1; De \ll 1$ | Extended Golestanian <i>et al.</i> (2007) & showed the importance of higher swimming modes and asymmetry of surface activity, assuming the swimming gait to be Newtonian. |
| Natale <i>et al.</i> (2017) | $\epsilon \rightarrow 0$ $Da \sim O(1); Pe \sim O(1)$ $De \ll 1$ | Extended Datt <i>et al.</i> (2017) to include advective and reactive effects. FEM simulations revealed sharp gradient in stresses due to viscoelasticity. |
| This work | $\epsilon \ll 1$ $Da \rightarrow 0$ $De \ll 1; \chi \ll 1$ | Extends Anderson <i>et al.</i> (1982) to include the effects of complex rheology. Uses Golestanian <i>et al.</i> (2007) framework to study non-Newtonian effects on the swimming gait and mobility of Janus sphere for $Pe \rightarrow 0$ & $\epsilon Pe \ll 1$. |

TABLE 1. A summary of continuum-level studies in the past towards the modeling of a non-Brownian active particle; first section of the table describes the studies in Newtonian medium in various regimes; second section summarizes the studies which explored the effects of Non-Newtonian behavior. Here, ϵ is the interaction layer thickness, normalized by the particle radius; Da is the Damkhler number which represents the ratio of time scale associated with diffusion to that of reaction; Pe is the Pcllet number which is the ratio of time scale associated with diffusion to that of advection; De is the Deborah number which governs the ratio of time scale of polymer relaxation to that of advection; χ represents the deviation of infinite shear rate viscosity from that of zero shear rate (the difference is normalized with zero shear rate viscosity).

the effect of polymer entanglements on active motion and showed that the particle can escape the entangled confinements at time-scales significantly shorter than the polymer relaxation time.

There have been a few theoretical studies in the past which have used the continuum-level framework and studied the effect of bulk non-Newtonian stresses on active propulsion. Zhu *et al.* (2012) employed the Giesekus model to study squirming in a viscoelastic fluid, considering the first two modes of swimming. They reported that swimming speed (and the swimming power required) is always lower than that in Newtonian fluids, with a minimum at a moderate value of Weissenberg number. Using a second-order fluid model, De Corato *et al.* (2015) also studied locomotion of a two-mode squirmers. They reported that ‘pullers’ are slowed, ‘pushers’ are hastened and ‘neutral’ squirmers are unaffected in

viscoelastic fluids. Later Datt *et al.* (2015) showed that it is essential to consider higher modes while modeling squirmers in a non-Newtonian medium as the constitutive equation is non-linear. Using a Carreau-Yasuda model, they demonstrated that the swimming may be faster or slower in shear-thinning fluids (compared to Newtonian fluids) depending upon the rate of actuation i.e. strength of the swimming modes. Their subsequent study (Datt *et al.* 2017) extended the analysis of Golestanian *et al.* (2007) to understand the self-propulsion of Janus sphere in complex media. Through an approach based on reciprocal theorem (Elfring & Goyal 2016), they showed the effect of bulk non-Newtonian stresses on the swimming velocity, assuming a prescribed slip velocity. They found: (i) swimming is always slower in weakly shear-thinning fluids; (ii) for weakly viscoelastic fluid, a Janus sphere swims faster if surface coverage of activity is more than half ($> \pi/2$) and vice-versa. However, they have acknowledged that the assumption of Newtonian slip in a non-Newtonian medium may not hold true. A change in slip velocity may significantly alter self-propulsion and therefore entails further investigation. A recent study by Natale *et al.* (2017) extended the work of Datt *et al.* (2017) to investigate the effects of advection and reaction (i.e. finite Peclet and Damkhler numbers) on self-diffusiophoresis through complex media. Their FEM simulations demonstrated the presence of an extensional flow across the point of discontinuity in the surface activity. This behavior was triggered due to large gradients in viscoelastic stresses. The origin of these viscoelasticity-triggered extensional flows is currently unknown. Given the recent interest in self-propulsion through complex medium with several open questions in the literature (Datt *et al.* 2017; Natale *et al.* 2017; Pietrzyk *et al.* 2019), it is natural to ask: how does the slip change in complex fluids and what is its relationship to the concentration field?

The importance of considering non-Newtonian effects in the thin interaction region around a diffusio-phoretic particle can be realized by considering the progress in the field of electrophoresis. Khair *et al.* (2012) demonstrated that the modification in slip due to shear-thinning can alter the mobility of an electrophoretic particle and the flow field around it. Very recently, through the use of various continuum-level rheological models (Oldroyd-B, Giesekus, FENE-P, and FENE-CR), Li & Koch (2020) showed that the electrophoretic particle (similar to a neutral squirmer in Newtonian fluid) behaves like a puller-type squirmer at low Weissenberg numbers. Such behavior arises primarily due to the elastic effects in the thin electrical double layer. These studies show that the high shear rate inside the thin layer generates significant polymer extension and deformation which needs to be accounted in the prediction of phoretic motion. Motivated by these recent developments in the context of electrophoresis (Khair *et al.* 2012; Zhao & Yang 2013; Li & Koch 2020), we aim to investigate the modification due to complex rheology in both slip and mobility of a self-propelling active particle.

In this work, we study the influence of viscoelasticity (second-order-fluid model) and shear-thinning (Carreau-fluid model) on the slip and mobility of an axisymmetric Janus particle. Using matched asymptotic expansions, we provide an analytical expression for the modification of the diffusio-osmotic slip due to complex rheology. Employing the reciprocal theorem, we evaluate the modification in the swimming velocity. Our results are applicable to a general diffusio-osmotic flow of complex fluid and hence can be used to model the transport of biological fluids through narrow channels and confinements, where applying pressure drop is undesirable (Michelin *et al.* 2015; Lisicki *et al.* 2016; Michelin & Lauga 2019).

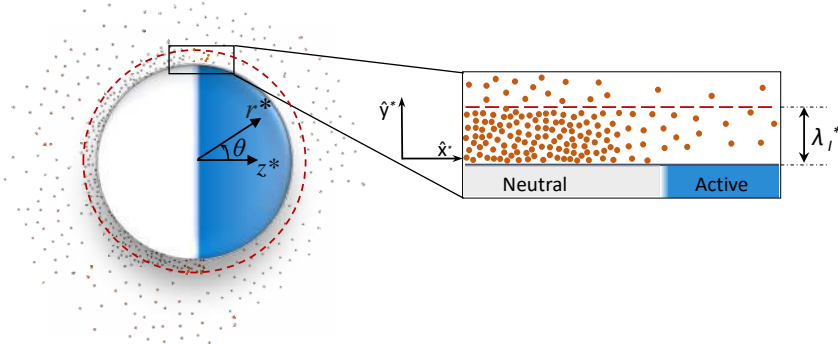


FIGURE 1. Schematic of an axisymmetric Janus particle suspended in a non-Newtonian medium with uniform external solute concentration. Zoomed-in view shows the solute in the thin interaction layer above a surface with varying activity. λ_I^* is the interaction layer thickness.

2. Active particle in a second-order-fluid

We consider an active particle surrounded by a non-Newtonian fluid medium consisting of a solute at uniform concentration C_∞^* . The solute, treated as a continuum, interacts with the active particle of radius a^* . The short-range interaction potential is governed by

$$\mathcal{P}^*(r, \theta) = k_B^* T^* \psi(r, \theta), \quad (2.1)$$

which acts over the length scale corresponding to the interaction layer thickness (λ_I^*), where $\lambda_I^* \ll a^*$. Here $*$ denotes the dimensional variables. ψ is interaction potential energy between the solute molecules and particle surface, scaled with the thermal energy ($k_B^* T^*$). This interaction generates a pressure field in the thin interaction layer, which decays to zero far away from the surface. The particle surface is partially active; a fixed-flux adsorption (\mathcal{A}^*) varies as a step function in the tangential direction. Since the interaction layer (λ_I^*) is very thin compared to the particle radius (a^*), we model this as a the diffusio-osmotic flow over a flat surface (see fig.1). This approximation neglects the curvature effects and introduces an error of $\mathcal{O}(\lambda_I^*/a^*)$ and has been employed earlier in studies involving particle electrophoresis (Anderson 1989; O'Brien 1983).

We assume the rheology inside and outside the interaction layer to follow the second-order fluid (SOF) model (Bird *et al.* 1987). This assumption is valid for slow and weakly non-Newtonian flows, i.e. polymers with low molecular weight ($\sim 10^4$) and radius of gyration smaller than the interaction layer thickness which is typically 1–10 nm (Sharifi-Mood *et al.* 2013). The qualitative insights obtained through such continuum models can also be extended to the cases of larger polymers as their segments would experience strong shear, which can result in modification of the dynamics in the interaction layer (Li & Koch 2020). We also assume the solute molecules, inside and outside the interaction layer, to follow Fickian diffusion with constant diffusivity. It has been reported that the presence of polymers significantly affects the diffusive mass transport in stagnant polymeric medium (Maldonado-Camargo & Rinaldi 2016; Makuch *et al.* 2020). To our knowledge, there has been no study conducted on the mass transport in sheared complex flows. As a first step, we focus on gaining qualitative insights and assume that the solute molecules and polymers are in the dilute concentration regime and the associated effects, such as volume exclusion and variable diffusivity, do not affect the system at leading order.

Following Anderson *et al.* (1982); Michelin & Lauga (2014), we represent the system

using the following non-dimensional equations

$$\nabla \cdot \mathbf{u} = 0, \quad -\nabla p + \nabla^2 \mathbf{u} + De \nabla \cdot \mathbf{S} - \frac{1}{\epsilon^2} (c + C_\infty) \nabla \psi = \mathbf{0}, \quad (2.2a)$$

$$Pe(\mathbf{u} \cdot \nabla c) = \nabla \cdot [\nabla c + (c + C_\infty) \nabla \psi]. \quad (2.2b)$$

The characteristic scales are

$$C_{ch} = \frac{|\mathcal{A}^*| a^*}{D^*}, \quad U_{ch} = \frac{k_B^* T^* \lambda_I^{*2} C_{ch}}{\mu^* a^*}, \quad p_{ch} = \frac{\mu^* U_{ch}}{a^*}, \quad y_{ch} = x_{ch} = a^*. \quad (2.3)$$

Here, D^* is the solute diffusion coefficient, $|\mathcal{A}^*|$ is the maximum magnitude of activity, c denotes the disturbance to the uniform concentration C_∞ (defined as: $c = C - C_\infty$). In eq. (2.2a), the polymeric stress $\mathbf{S} = \mathbf{A} \cdot \mathbf{A} + \delta \mathbf{B}$, where \mathbf{A} is the rate of strain tensor ($\nabla \mathbf{u} + (\nabla \mathbf{u})^T$) and \mathbf{B} is the steady-state Rivlin-Ericksen tensor (covariant derivative of rate of strain tensor \mathbf{A}) (Bird *et al.* 1987).

$$\mathbf{B} = \mathbf{U} \cdot \nabla \mathbf{A} + \mathbf{A} \cdot \nabla \mathbf{U}^T + \nabla \mathbf{U} \cdot \mathbf{A}. \quad (2.4)$$

Here T denotes the transpose. The dimensionless quantities are defined as

$$Pe = \frac{U_{ch} a^*}{D^*}, \quad De = \frac{\Psi_1^* + \Psi_2^*}{\mu^*} \left(\frac{U_{ch}}{a^*} \right), \quad \delta = \frac{-\Psi_1^*}{2(\Psi_1^* + \Psi_2^*)} \text{ and } \epsilon = \frac{\lambda_I^*}{a^*}, \quad (2.5)$$

where Pe (Peclet number) is the ratio of diffusive to advective time scales; De (Deborah number) is defined as the ratio of viscoelastic time scale to that based on shear arising due to macro-scale movement of the Janus sphere; δ is a viscometric parameter which compares first and second normal stress coefficients (Ψ_1^* and Ψ_2^* , respectively); ϵ is the dimensionless thickness of the interaction layer.

The boundary conditions are

$$\left(\frac{\partial c}{\partial y} \Big|_{y=0} + (c + C_\infty) \frac{\partial \psi}{\partial y} \Big|_{y=0} \right) = \frac{\mathcal{A}^*(x)}{|\mathcal{A}^*|} = \mathcal{K}(x) \text{ and } \mathbf{u} = \mathbf{0} \quad \text{at } y = 0; \quad (2.6a)$$

$$p \rightarrow 0, \quad \psi \rightarrow 0 \text{ and } c \rightarrow 0 \quad \text{as } y \rightarrow \infty. \quad (2.6b)$$

In the next sections, we investigate the influence of non-Newtonian effects on the diffusio-osmotic slip on a partially active surface using matched asymptotic expansions (MAE).

2.1. Evaluation of the diffusio-osmotic slip

For an asymptotically thin interaction region ($\epsilon \ll 1$), we expand the field variables (\mathbf{u}, c, p, ψ) as

$$f(x, y) = f^{(0)}(x, y) + \epsilon f^{(1)}(x, y) + \dots \quad (2.7)$$

Following Michelin & Lauga (2014), we divide the domain into an ‘inner’ ($0 \ll y \ll 1$) and ‘outer’ region ($y \gg 1$). We use MAE to replace the solution of the inner region with coarse-grained boundary conditions for the outer region.

2.1.1. Outer region

Neglecting the rapidly decaying interaction potential in the outer region i.e. $\psi = 0$ (Sharifi-Mood *et al.* 2013; Michelin & Lauga 2014), the leading order equations read

$$\nabla \cdot \mathbf{u}^{(0)} = 0, \quad -\nabla p^{(0)} + \nabla^2 \mathbf{u}^{(0)} + De \nabla \cdot \mathbf{S}^{(0)} = \mathbf{0}, \quad (2.8a)$$

$$Pe(\mathbf{u}^{(0)} \cdot \nabla c^{(0)}) = \nabla^2 c^{(0)}, \quad (2.8b)$$

subject to the following boundary conditions

$$\mathbf{u}^{(0)} \rightarrow \mathbf{0}, \quad p^{(0)} \rightarrow 0 \quad \text{and} \quad c^{(0)} \rightarrow 0 \quad \text{as} \quad y \rightarrow \infty, \quad (2.9a)$$

$$\mathbf{n} \cdot \nabla c^{(0)} = \mathcal{K}(x) \quad \text{at} \quad y \rightarrow 0. \quad (2.9b)$$

The concentration boundary condition (2.9b) has been shown to be well-posed, provided $\epsilon Pe \ll 1$ (Michelin & Lauga 2014, p. 580). To obtain the macro-scale boundary condition for velocity at the *surface* (i.e. $y \rightarrow 0$), the outer solution must be matched with the inner solution.

2.1.2. Inner region

We first rescale the variables to derive the equations in the inner layer. The variables in the inner layer are defined as \hat{f} . In the thin interaction layer limit ($\epsilon \ll 1$), using boundary layer principles, we define the characteristic scales for y^* – direction, vertical velocity (v^*) and pressure (p^*) as λ_I^* , ϵU_{ch} and $p_{ch} \epsilon^{-2}$, respectively. Using (2.7) and (2.2), we obtain the leading order equations in the inner region as

$$\frac{\partial \hat{u}^{(0)}}{\partial \hat{x}} + \frac{\partial \hat{v}^{(0)}}{\partial \hat{y}} = 0, \quad (2.10a)$$

$$-\frac{\partial \hat{p}^{(0)}}{\partial \hat{x}} + \frac{\partial^2 \hat{u}^{(0)}}{\partial \hat{y}^2} + De \left(\frac{\partial \hat{S}_{xx}^{(0)}}{\partial \hat{x}} + \frac{\partial \hat{S}_{yx}^{(0)}}{\partial \hat{y}} \right) = (\hat{c}^{(0)} + C_\infty) \frac{\partial \hat{\psi}^{(0)}}{\partial \hat{x}}, \quad (2.10b)$$

$$-\frac{\partial \hat{p}^{(0)}}{\partial \hat{y}} + De \left(\frac{\partial \hat{S}_{yy}^{(0)}}{\partial \hat{y}} \right) = (\hat{c}^{(0)} + C_\infty) \frac{\partial \hat{\psi}^{(0)}}{\partial \hat{y}}, \quad (2.10c)$$

$$\epsilon^2 Pe \left(\hat{u}^{(0)} \frac{\partial \hat{c}^{(0)}}{\partial \hat{x}} + \hat{v}^{(0)} \frac{\partial \hat{c}^{(0)}}{\partial \hat{y}} \right) = \frac{\partial}{\partial \hat{y}} \left(\frac{\partial \hat{c}^{(0)}}{\partial \hat{y}} + (\hat{c}^{(0)} + C_\infty) \frac{\partial \hat{\psi}^{(0)}}{\partial \hat{y}} \right), \quad (2.10d)$$

subject to the following surface boundary conditions at the leading order

$$\mathbf{u}^{(0)} \Big|_{\hat{y}=0} = \mathbf{0} \quad \text{and} \quad \left(\frac{\partial \hat{c}^{(0)}}{\partial \hat{y}} \Big|_{\hat{y}=0} + (\hat{c}^{(0)} + C_\infty) \frac{\partial \hat{\psi}}{\partial \hat{y}} \Big|_{\hat{y}=0} \right) = 0. \quad (2.11)$$

For low to moderate advective effects (i.e. $Pe \ll \epsilon^{-2}$), we can neglect the LHS of (2.10d). This decouples the solute concentration field from the hydrodynamics. The pressure scaling in the inner region suggests a decay condition as $\hat{y} \rightarrow \infty$ (Michelin & Lauga 2014, p.579). The components of the polymeric stress tensor (\mathbf{S}) are rescaled as:

$$S_{xx} = \frac{\hat{S}_{xx}}{\epsilon^2} = \frac{1}{\epsilon^2} \left(\frac{\partial \hat{u}}{\partial \hat{y}} \right)^2, \quad S_{yy} = \frac{\hat{S}_{yy}}{\epsilon^2} = \frac{1}{\epsilon^2} (1 + 2\delta) \left(\frac{\partial \hat{u}}{\partial \hat{y}} \right)^2, \quad (2.12a)$$

$$S_{xy} = S_{yx} = \frac{\hat{S}_{xy}}{\epsilon} = \frac{\hat{S}_{yx}}{\epsilon} = \frac{2\delta}{\epsilon} \left(\frac{\partial \hat{u}}{\partial \hat{y}} \frac{\partial \hat{u}}{\partial \hat{x}} + \frac{\hat{v} \frac{\partial^2 \hat{u}}{\partial \hat{y}^2} + \hat{u} \frac{\partial^2 \hat{u}}{\partial \hat{y} \partial \hat{x}}}{2} \right). \quad (2.12b)$$

We now perform a perturbation expansion in Deborah number (De) i.e. accounting for weakly non-linear viscoelastic effects such that $\epsilon \ll De \ll 1$. The field variables for each term in (2.7) can be further expanded as

$$f^{(i)}(x, y) = f_0^{(i)}(x, y) + De f_1^{(i)}(x, y) + \dots \quad (2.13)$$

Here f represents the velocity and pressure field. The concentration field is not expanded in De as it is decoupled from velocity field in the inner region.

$\mathcal{O}(De^0)$ solution: Since our objective is to obtain leading order change in the diffusio-osmotic slip, for convenience, we temporarily drop the superscript (0) from all the variables. The solution to leading order (i.e. $\mathcal{O}(De^0)$) governing equations is obtained as

$$\hat{c}(\hat{x}, \hat{y}) = \mathcal{I}(\hat{x})e^{-\hat{\psi}(\hat{x}, \hat{y})} - C_\infty, \quad \hat{p}_0(\hat{x}, \hat{y}) = \mathcal{I}(\hat{x}) \left(e^{-\hat{\psi}(\hat{x}, \hat{y})} - 1 \right), \text{ and} \quad (2.14a)$$

$$\hat{u}_0(\hat{x}, \hat{y}) = -\mathcal{I}'(\hat{x}) \int_0^{\hat{y}} \int_t^\infty \left(e^{-\hat{\psi}(\hat{x}, s)} - 1 \right) ds dt + \mathcal{J}_0(\hat{x}) \hat{y}. \quad (2.14b)$$

Here, $\mathcal{I}(\hat{x})$ and $\mathcal{J}_0(\hat{x})$ are to be determined through matching, $\mathcal{I}'(\hat{x})$ represents $d\mathcal{I}/d\hat{x}$. The leading order velocity field (\hat{u}_0) is represented in a form different from that reported by Anderson *et al.* (1982); Michelin & Lauga (2014), as it helps in the evaluation of higher order velocity field. It can be seen in fig. 2(a) that this expression is equivalent to that provided in the literature.

$\mathcal{O}(De^1)$ solution: For ease of calculation, we assume the interaction potential ($\hat{\psi}$) to be independent of the tangential direction (x). The flow field at $\mathcal{O}(De)$ is governed by

$$\frac{\partial \hat{u}_1}{\partial \hat{x}} + \frac{\partial \hat{v}_1}{\partial \hat{y}} = 0, \quad (2.15a)$$

$$-\frac{\partial \hat{p}_1}{\partial \hat{x}} + \frac{\partial^2 \hat{u}_1}{\partial \hat{y}^2} + \frac{\partial \hat{S}_{xx0}}{\partial \hat{x}} + \frac{\partial \hat{S}_{xy0}}{\partial \hat{y}} = 0, \quad (2.15b)$$

$$-\frac{\partial \hat{p}_1}{\partial \hat{y}} + \frac{\partial \hat{S}_{yy0}}{\partial \hat{y}} = 0. \quad (2.15c)$$

Using the pressure decay condition ($\hat{p} \rightarrow 0$ as $\hat{y} \rightarrow \infty$), the solution to (2.15c) yields $\hat{p}_1 = \hat{S}_{yy0} - \mathcal{J}_0(\hat{x})^2(1 + 2\delta)$. Simplifying (2.15b), we obtain

$$\frac{\partial^2 \hat{u}_1}{\partial \hat{y}^2} = -\delta \left\{ -\frac{\partial \hat{u}_0}{\partial \hat{y}} \frac{\partial^2 \hat{u}_0}{\partial \hat{y} \partial \hat{x}} + \frac{\partial \hat{u}_0}{\partial \hat{x}} \frac{\partial^2 \hat{u}_0}{\partial \hat{y}^2} + \hat{v}_0 \frac{\partial^3 \hat{u}_0}{\partial \hat{y}^3} + \hat{u}_0 \frac{\partial^3 \hat{u}_0}{\partial \hat{x} \partial \hat{y}^2} \right\} + \frac{\partial \hat{p}_1}{\partial \hat{x}}. \quad (2.16)$$

Here, \hat{v}_0 is found by substituting (2.14b) in the continuity equation and integrating it over \hat{y} direction. Substituting (2.14b) and \hat{v}_0 into (2.16), integrating twice over \hat{y} direction and using the no-slip condition, we obtain †

$$\begin{aligned} \hat{u}_1 = \mathcal{J}_1(\hat{x})\hat{y} - \delta \mathcal{I}'(\hat{x})\mathcal{I}''(\hat{x}) \int_0^{\hat{y}} dp \int_p^\infty \left\{ \left(\int_r^\infty \mathcal{F}(s) ds \right)^2 + 2\mathcal{F}(r) \left(\int_0^r s\mathcal{F}(s) ds + r \int_r^\infty \mathcal{F}(s) ds \right) \right. \\ \left. + \hat{\psi}'(r)e^{-\hat{\psi}(r)} \left(\int_0^r \left(r - \frac{s}{2} \right) s\mathcal{F}(s) ds + \frac{r^2}{2} \int_r^\infty \mathcal{F}(s) ds \right) \right. \\ \left. - \left(\frac{\mathcal{J}_0(\hat{x})}{\mathcal{I}'(\hat{x})} + \frac{\mathcal{J}'_0(\hat{x})}{\mathcal{I}''(\hat{x})} \right) \left(r\mathcal{F}(r) + \int_r^\infty \mathcal{F}(s) ds \right) \right. \\ \left. + \frac{\mathcal{J}_0(\hat{x})}{\mathcal{I}''(\hat{x})} \mathcal{F}'(r) \frac{r^2}{2} + \frac{\mathcal{J}_0(\hat{x})\mathcal{J}'_0(\hat{x})}{\mathcal{I}'(\hat{x})\mathcal{I}''(\hat{x})} - \frac{2\mathcal{J}_0(\hat{x})\mathcal{J}'_0(\hat{x})(1+2\delta)}{\delta \mathcal{I}'(\hat{x})\mathcal{I}''(\hat{x})} \right\} dr. \end{aligned} \quad (2.17)$$

Here, $\mathcal{F}(\xi) = -1 + e^{-\hat{\psi}(\xi)}$, \mathcal{J}_0 , \mathcal{J}_1 , and \mathcal{I} are to be determined through matching.

2.1.3. Matching

For any field variable f , the matching condition at $\mathcal{O}(\epsilon^0)$ is

$$\lim_{y \rightarrow 0} (f_0^{(0)} + Def_1^{(0)} + \dots) = \lim_{\hat{y} \rightarrow \infty} (\hat{f}_0^{(0)} + De\hat{f}_1^{(0)} + \dots). \quad (2.18)$$

† Details of the derivation can be found in the supplementary material.

The matching condition for the concentration field (\hat{c}) yields $\mathcal{I}(\hat{x}) = \lim_{y \rightarrow 0} c^{(0)}(x, y) + C_\infty$. We substitute (2.14b) and (2.17) in the above matching condition and obtain: $\mathcal{J}_0 = \mathcal{J}_1 = 0$ at the leading order. At the present order of approximation, the total slip velocity at the outer edge of the interaction layer is

$$u^{(0)} \Big|_{y=0} = c_x^{(0)} \Big|_{y=0} \left(M_0 + De \delta M_1 c_{xx}^{(0)} \Big|_{y=0} \right), \quad (2.19)$$

where M_0 and M_1 are the mobility coefficients representing the Newtonian and non-Newtonian contribution, respectively:

$$M_0 = - \int_0^\infty \int_t^\infty \mathcal{F}(s) ds dt \quad \text{and} \quad M_1 = - \int_0^\infty \int_p^\infty \mathcal{G}(r) dr dp, \quad (2.20)$$

$$\text{where } \mathcal{G}(r) = \left\{ \left(\int_r^\infty \mathcal{F}(s) ds \right)^2 + 2 \mathcal{F}(r) \left(\int_0^r s \mathcal{F}(s) ds + r \int_r^\infty \mathcal{F}(s) ds \right) + \hat{\psi}'(r) e^{-\hat{\psi}(r)} \left(\int_0^r \left(r - \frac{s}{2} \right) s \mathcal{F}(s) ds + \frac{r^2}{2} \int_r^\infty \mathcal{F}(s) ds \right) \right\}. \quad (2.21)$$

Modification to the diffusio-osmotic slip velocity (arising from the non-Newtonian effects) is found to be proportional to the first and second tangential derivative of the bulk-scale concentration at the particle *surface*. The dependency of viscoelastic effects on the second derivative can be intuitively understood by realizing that the polymers stretch only when there exists a spatial variation (or gradient) in the flow. In the current physics, as the flow itself is generated by the chemical gradient, an appearance of second derivative at the leading order is intuitive. The proportionality to $De \delta$ and the definitions described in (2.5) suggests that the effect of viscoelasticity is solely due to first normal stress difference. Furthermore, the dimensional form of the slip velocity reveals that the first order slip has a dependency on the characteristic length scale (a^*), whereas the Newtonian slip is independent of it.

2.1.4. Velocity profile in the inner region

Fig. 2(a) shows the Newtonian component of the velocity field (2.14b) inside the thin interaction layer for an exponentially decaying solute-surface attraction (Anderson *et al.* 1982; Sharifi-Mood *et al.* 2013). The velocity monotonically grows away from the surface and approaches an asymptotic value, which upon multiplication with the tangential concentration gradient provides the Newtonian slip velocity. The viscoelastic component of the velocity field ($\hat{u}_1^{(0)}$), shown in Fig. 2(b), also grows monotonically away from the surface and attains an asymptotic value. The magnitude of this asymptotic value increases with the magnitude of interaction (Φ_0).

The interactions between solute molecules and the particle surface can be either attractive or repulsive; if the solute is more attracted to the surface than the solvent, the interaction coefficient (Φ_0) is negative and vice-versa. To show that the current analysis is valid for other forms of potential, we evaluate the velocity field inside the interaction layer for (i) an exponentially repulsive and (ii) a long-range van der Waals type interaction (arising primarily from the dipolar forces $\sim 1/r^6$). (i) For repulsive interactions, the thin ‘layer’ is characterized by an absence of solute concentration and the direction of motion (and slip) is opposite to that observed for an attractive interaction (Michelin & Lauga 2014). Fig 3 (a) shows that while the Newtonian slip is reversed (c.f. fig.2a), the contribution from viscoelasticity is in the same direction as the case of attraction potential (c.f. fig.2b). (ii) To incorporate long-range attractive van der Waals interactions, we follow

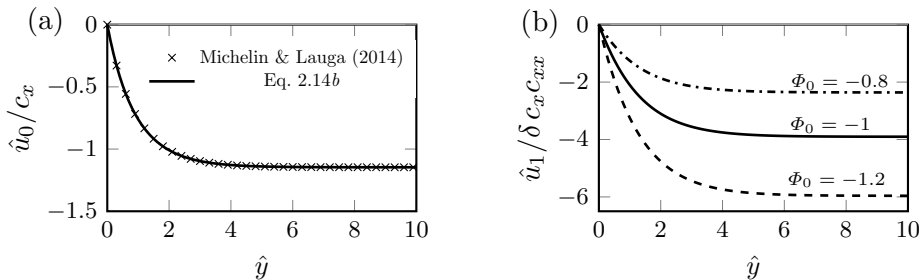


FIGURE 2. (a) Velocity field of Newtonian fluid inside the interaction layer (for $\Phi_0 = -1$). (b) Variation of the modification to the velocity field. The results are for an exponentially decaying solute-surface interaction ($\hat{\psi}(\xi) = \Phi_0 e^{-\xi}$), where a negative Φ_0 represents an attractive interaction. Here c_x and c_{xx} are the concentration gradients at the outer edge of the inner region i.e. $c_x|_{y=0}$ and $c_{xx}|_{y=0}$.

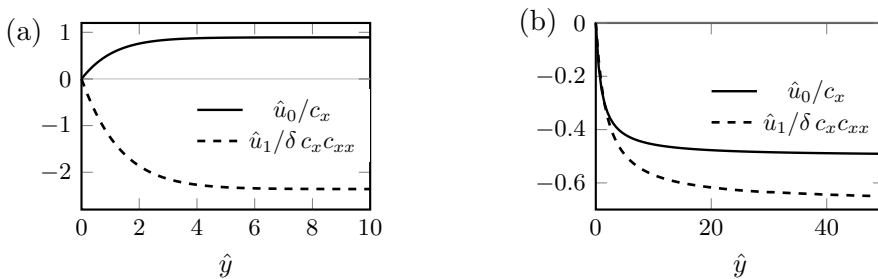


FIGURE 3. Velocity field inside the interaction layer for: (a) repulsive exponential interaction ($\Phi_0 = +1$); (b) attractive long-range van der Waals interaction ($\Phi_0 = -1$). Here c_x and c_{xx} are the concentration gradients at the outer edge of the inner region i.e. $c_x|_{y=0}$ and $c_{xx}|_{y=0}$.

Anderson *et al.* (1982, p. 112) and assume the dipolar interactions between the solute molecules to be pairwise additive. The potential is defined as

$$\hat{\psi}(\xi) = \Phi_0 \left(-\frac{1}{(1+\xi)^9} + \frac{1}{(1+\xi)^3} \right). \quad (2.22)$$

The velocity fields (Newtonian and viscoelastic) remain qualitatively similar to that obtained from the attractive exponential interactions. As (2.22) decays slower than the exponential interaction, the asymptotic value (i.e. slip velocity) is obtained at larger distances from the surface.

The above results demonstrate that the velocity fields inside the inner region approach an asymptotic value, the slip velocity. The Newtonian component of this slip is equal to the product of concentration gradient ($c_x|_{y=0}$) and mobility coefficient M_0 . Similarly, the viscoelastic component of velocity field involves M_1 (see eq.2.19). The magnitude of mobility coefficients depends on the nature of interaction (exponential or van der Waals) and magnitude of attraction or repulsion (Φ_0). Fig. 4 quantifies the effect of the type and magnitude of interaction on M_0 and M_1 . Fig. 4(a) shows that the sign of Newtonian mobility coefficient is opposite for repulsive interactions ($\Phi_0 > 0$) as compared to attractive ones ($\Phi_0 < 0$), for both the type of interactions i.e. short-range exponential or long-range van der Waals solute-surface interaction. Fig. 4(b) shows that the sign of M_1 is always negative and does not depend on the nature of interaction. Furthermore,

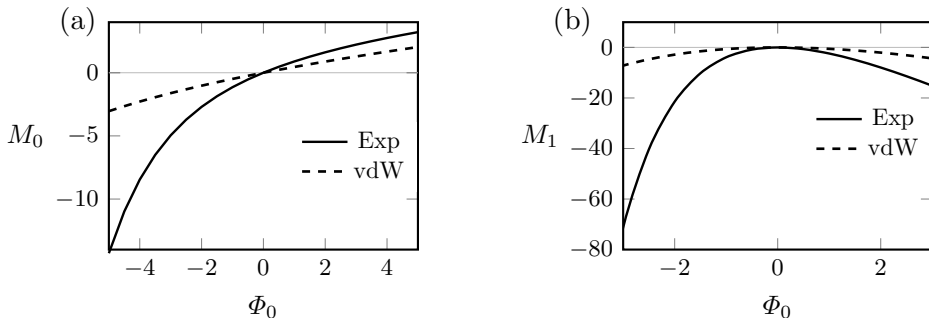


FIGURE 4. Variation of mobility coefficient (a) M_0 and (b) M_1 with respect to Φ_0 , for exponential and van der Waals interactions.

for repulsive interactions, the magnitude of mobility coefficients (M_0 and M_1) is reduced because the adsorption coefficient in such cases is generally lower than that of attractive interactions (Anderson 1989, p.69).

The above results, valid for moderate advective effects ($\epsilon Pe \ll 1$), are also applicable to diffusio-osmotic flows of complex fluids in micro-channels, arising from externally imposed concentration gradients or generated due to active ‘patches’ (Michelin *et al.* 2015; Michelin & Lauga 2019).

Several experimental studies (Ebbens & Howse 2011; Baraban *et al.* 2012; Ke *et al.* 2010) have shown that the particles are propelled with catalytic surface oriented at the rear-end, suggesting an attractive interaction between the solute molecules and active particle. Thus, we focus primarily on the exponentially attractive interaction because of its simplicity and ease of computation. We follow Sabass & Seifert (2012); Sharifi-Mood *et al.* (2013) and assume $|\Phi_0| = 1$ in the results which follow.

2.2. Diffusio-osmotic slip on an active particle

We now extend the results of previous section (eq.2.19) to an axisymmetric Janus particle. Here, direction normal to the surface is r ; the tangential direction is polar angle θ , varying from 0 to π . The surface activity \mathcal{K} follows a step function (Golestanian *et al.* 2007; Michelin & Lauga 2014; Natale *et al.* 2017)

$$\mathcal{K}(\theta) = \begin{cases} 1 & \theta < \theta_c \\ 0 & \theta > \theta_c, \end{cases} \quad (2.23)$$

where θ_c is the angle at which the activity undergoes a step change; it represents the surface coverage of activity. To evaluate the slip velocity, we require solution to the concentration field (c) in the outer region. In the absence of advection ($Pe \rightarrow 0$), the solution to (2.8b) is sought in terms of an expansion in Legendre polynomials. Following Golestanian *et al.* (2007), we obtain the solution to bulk-scale concentration field as

$$c(r, \theta) = \sum_{n=0}^{\infty} \frac{-\mathcal{K}_n}{(n+1)} \frac{P_n(\cos \theta)}{r^{n+1}}, \quad (2.24)$$

where P_n is the n^{th} order Legendre polynomial. Here \mathcal{K}_n are the spectral coefficients of the activity distribution:

$$\mathcal{K}(\theta) = \sum_{n=0}^{\infty} \mathcal{K}_n P_n(\cos \theta). \quad (2.25)$$

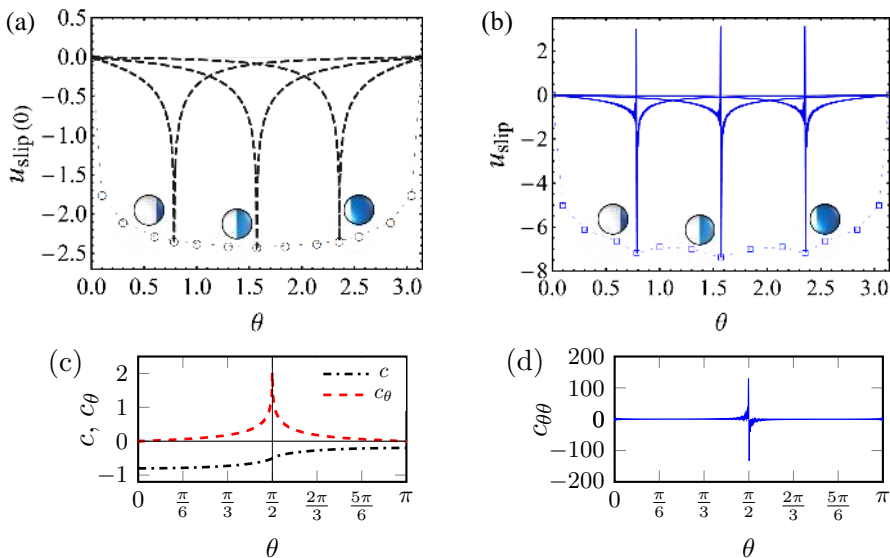


FIGURE 5. (a) The dashed line represents the Newtonian slip velocity along the polar angle for three different surface coverages ($\theta_c = \pi/4, \pi/2, 3\pi/4$). The empty circular markers represent the maximum magnitude of the slip velocity for a range of surface coverage: $\theta_c \in (0, \pi)$. (b) The solid line represents the total slip velocity (for three different surface coverages) along the polar angle for $\hat{\psi}(\xi) = -e^{-\xi}$, $De = 0.01$, and $\delta = -0.5$. The empty square markers represent the maximum magnitude of the slip velocity for a range of surface coverage: $\theta_c \in (0, \pi)$. (c) Concentration profile and its gradient along the polar angle (on particle surface) for $\theta_c = \pi/2$. (d) Shows the profile for $c_{\theta\theta}$. First 700 modes were used to describe the concentration field and its gradients.

These coefficients are found by taking an inner product of (2.25) with the Legendre polynomials (Michelin & Lauga 2014), and are obtained as

$$\mathcal{K}_0 = \frac{(1 - \cos \theta_c)}{2} \quad \text{and} \quad \mathcal{K}_n = \frac{-1}{2} (P_{n+1}(\cos \theta_c) - P_{n-1}(\cos \theta_c)) \quad \text{for } n \geq 1. \quad (2.26)$$

Using the expression (2.24) for disturbance concentration, we obtain the total tangential slip velocity ($M_0 c_\theta(1, \theta) + De \delta M_1 c_\theta(1, \theta) c_{\theta\theta}(1, \theta)$)

$$\mathbf{u}|_{r=1} = M_0 \sum_{n=1}^{\infty} \frac{-\mathcal{K}_n}{n+1} \frac{\partial P_n}{\partial \theta} \mathbf{e}_\theta + De \delta M_1 \left(\sum_{n=1}^{\infty} \frac{-\mathcal{K}_n}{(n+1)} \frac{\partial P_n}{\partial \theta} \right) \left(\sum_{n=1}^{\infty} \frac{-\mathcal{K}_n}{(n+1)} \frac{\partial^2 P_n}{\partial \theta^2} \right) \mathbf{e}_\theta. \quad (2.27)$$

Fig. 5(a) shows the Newtonian slip velocity varying across the polar angle for three different surface coverages ($\theta_c = \pi/4, \pi/2, 3\pi/4$). The negative value depicts that the diffusio-osmotic flow is towards the active region, and is highest in magnitude at the point of discontinuity of activity ($\theta = \theta_c$) †. For a second-order fluid, figure 5(b) shows a localized reversal of slip velocity across θ_c for all three surface coverages i.e. the slip velocity exhibits sharp non-linear gradients across the point of discontinuity of activity. Natale *et al.* (2017) also reported such behavior in viscoelastic stresses at θ_c in their FEM simulations. They reported a viscoelasticity triggered extensional flow, across the point of discontinuity in surface activity.

† These results are validated with those reported by Michelin & Lauga (2014) and are described in the supplementary material.

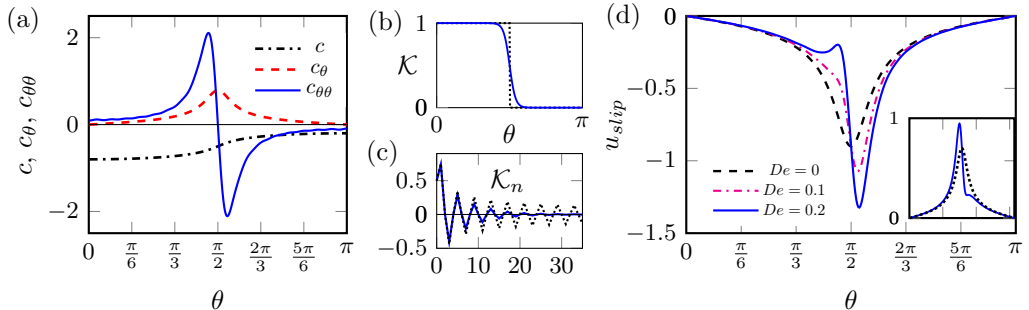


FIGURE 6. (a) Profile of concentration and its first and second gradient along the polar angle on particle surface for $\theta_c = \pi/2$. (b) Variation of surface activity as a step (dotted line) and smooth function (solid line). (c) Convergence of spectral modes: solid (blue) line shows fast convergence for the sigmoidal activity; dotted (black) line depicts slow convergence for step activity. (d) Different curves represent the total slip velocity along the polar angle for $\hat{\psi}(\xi) = -1e^{-\xi}$, $\zeta = 16$, $\delta = -0.5$ and different Deborah numbers. The inset shows the slip velocities for Newtonian and second-order fluid for the case of repulsive interactions; $\Phi_0 = +1$, $De = 0$ (.....), and $De = 0.2$ (—).

To analyze this slip reversal or the extensional flow across the point of transition in activity, we examine the behavior of concentration field around the particle, as the slip (2.19) depends on the first and second polar gradient of the concentration field. In fig. 5(c,d), c , c_θ and $c_{\theta\theta}$ profiles are shown for $\theta_c = \pi/2$. A step change in surface activity causes the second tangential gradient to undergo a sharp reversal at $\theta = \theta_c$, which is also reflected in the slip velocity (in fig. 5b). The profile of $c_{\theta\theta}$, despite accounting for first 700 modes, exhibits oscillations near the point of discontinuity of surface activity. This seemingly divergent behavior is because of the step function representation of surface activity (2.23), which is widely employed in the literature (Golestanian *et al.* 2007; Michelin & Lauga 2014; Natale *et al.* 2017). This oscillatory behavior indicates a violation of scaling in the inner region: in section §2.1.2 the length scale in the tangential direction was assumed to be a^* , which is not the true characteristic of the system as variations in the tangential direction are rapid for a step discontinuity in surface activity. Since such step discontinuities are unlikely to be realized in experiments, we now consider a smooth variation in the activity and demonstrate that the oscillatory behavior of $c_{\theta\theta}$ for step activity is a mathematical artefact.

2.3. Diffusio-osmotic slip on an active particle: Sigmoidal function approximation

The step change in surface activity is approximated as a logistic function:

$$\mathcal{K}(\theta) = 1 / (1 + \exp[-\zeta(\theta_c - \theta)]). \quad (2.28)$$

Here ζ (transition parameter) determines the sharpness of the transition of non-dimensional activity from 1 to 0. We have chosen $\zeta = 16$ such that the transition length is large enough to maintain the consistency of the analysis with respect to the scaling used in §2.1.2 (i.e. $l_{transition}^* \sim a^*$)†. The expression for concentration and slip velocity is identical to (2.24) and (2.27), respectively. The spectral modes can be found by substituting (2.28) into (2.25) and taking the inner product with respect to P_n on both

† For a particle of $a^* = 5\mu\text{m}$, $\zeta = 16$ corresponds to a circumferential length of $\sim 2\mu\text{m}$, which is much larger than the typical interaction layer thickness ($\sim 10\text{nm}$).

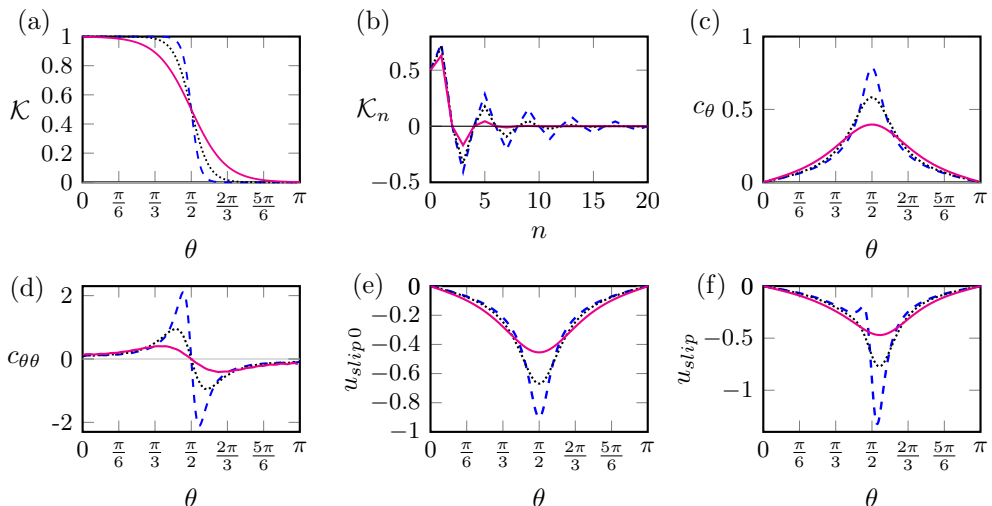


FIGURE 7. Analysis of activity, spectral modes, concentration gradients and slip velocity for three different transition parameters: $\zeta = 4$ (—), $\zeta = 8$ (⋯), $\zeta = 16$ (- - -). (a) Variation of activity with polar angle θ . (b) Convergence of spectral modes. (c) Tangential concentration gradient. (d) Second tangential derivative of concentration field. (e) Slip velocity of a Newtonian fluid. (f) Slip velocity of the second-order fluid. Parameters: $De = 0.1$, $\delta = -0.5$, $\Phi_0 = -1$.

sides

$$\mathcal{K}_n = \frac{2n+1}{2} \int_0^\pi \mathcal{K}(\theta) P_n(\cos\theta) d\theta. \quad (2.29)$$

For activity represented by (2.28), the concentration field and its tangential gradients (at the surface) are depicted in fig.6(a). The variation of concentration field and its gradients along θ is qualitatively similar to that reported for the step change; the magnitudes, however, are significantly reduced. Fig. 6(b) & (c) show the variation of activity and its faster convergence in comparison to the step function. Fig.6(d) compares the slip velocity for a second-order fluid with that of a Newtonian fluid.

We observe a reduction and an increase in slip velocity (relative to the Newtonian slip) across θ_c . The magnitude of modification in the slip is significantly reduced (as compared to that of step change in activity) which is a consequence of smooth transition of activity. The above observations are for attractive interactions between solute and particle surface; for repulsive interactions (as shown in the inset of fig. 6d), the effect of viscoelasticity is qualitatively reversed.

Fig. 7 demonstrates the effect of the transition parameter (ζ) on the spectral coefficients, concentration gradients and slip velocity. Fig. 7(a) shows that as ζ decreases, the transition from active to passive surface becomes smoother. Fig. 7(b) depicts that the convergence is faster for lower ζ . As the transition becomes smoother the magnitude of tangential gradients of concentration field reduces (fig.7c,d). As a result, the slip velocity for Newtonian and second-order fluid reduces (shown in fig.7e-f, respectively).

The effect of viscoelasticity on the tangential velocity field inside the interaction layer is shown in fig. 8 (for $\theta_c = \pi/2$). Fig. 8(a) depicts the symmetry of velocity field across θ_c for Newtonian fluid. Addition of viscoelasticity breaks this symmetry: the contours of the SOF in 8(b) depict that the magnitude of velocity field is reduced before the transition of activity ($\theta < \theta_c$) and increased after it ($\theta > \theta_c$). These results suggest that the elastic effects (characterized by De) tend to locally reverse the slip velocity. At

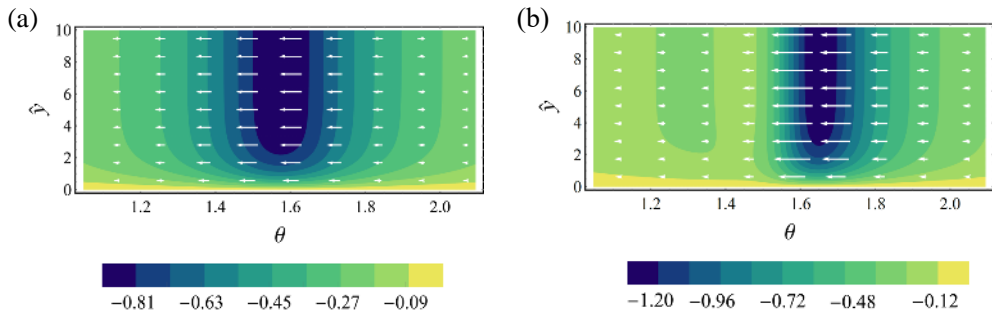


FIGURE 8. (a) Newtonian tangential velocity contours inside the interaction layer for $\theta_c = \pi/2$. (b) Tangential velocity contours corresponding to the second-order fluid for $\hat{\psi}(\xi) = -e^{-\xi}$, $\zeta = 16$, $\delta = -0.5$, and $De = 0.2$. Arrows depict the velocity vectors ($\hat{\mathbf{u}}_1^{(0)}$).

higher Deborah numbers, a reversal in slip velocity can generate a local extensional flow near the transition of activity (as shown by Natale *et al.* (2017)), resembling a puller or pusher-type flow field. Interestingly, there have been similar observations in the context of electrophoresis. Recently, Li & Koch (2020) reported that, in weakly viscoelastic fluids, polymer elasticity in the inner region changes the electrophoretic particle into a puller-type squirmer: the strong shear flow in the double layer causes the polymers to be stretched tangentially, resulting in a pulling-flow at the front & rear end, and a pushing-flow from the sides. Since electrophoretic and diffusiophoretic mechanisms share qualitative similarities (such as strong shear inside the thin layer), it is plausible that the effects of elasticity, observed in electrophoresis, emerge in diffusiophoresis as well.

2.4. Swimming velocity

The motion of the Janus sphere is found by using the reciprocal theorem. Following Stone & Samuel (1996) and Ho & Leal (1976), we obtain the swimming velocity as

$$U = \frac{-1}{4\pi} \int_{S_p} (\mathbf{u}_0 + De \mathbf{u}_1)|_{r=1} \cdot \mathbf{e}_z dS - \frac{1}{6\pi} De \int_{V_f} \mathbf{S}_0 : \nabla \mathbf{u}^t dV. \quad (2.30)$$

Here, \mathbf{u}_0 is the Newtonian slip velocity ($M_0 c_\theta(1, \theta) \mathbf{e}_\theta$); $De \mathbf{u}_1$ is the modification to slip due to viscoelasticity ($De M_1 \delta c_\theta(1, \theta) c_{\theta\theta}(1, \theta) \mathbf{e}_\theta$); \mathbf{u}^t is the test flow field which governs the motion of a rigid sphere in z -direction with unit velocity in a quiescent Newtonian medium (Michelin & Lauga 2014; Datt *et al.* 2015), and \mathbf{S}_0 is the polymeric stress. The first integral (U_λ) denotes the contribution to swimming velocity arising from the slip, which contains a Newtonian (U_{λ_0}) and a non-Newtonian (U_{λ_1}) component:

$$U_{\lambda_0} = \frac{M_0}{2} \int_0^\pi c_\theta(1, \theta) \sin^2 \theta d\theta; \quad U_{\lambda_1} = \frac{De \delta M_1}{2} \int_0^\pi c_\theta(1, \theta) c_{\theta\theta}(1, \theta) \sin^2 \theta d\theta. \quad (2.31)$$

We evaluate the above components and compare them in fig. 9 for different surface coverages. For attractive interactions between the solute molecules and particle, the Newtonian swimming velocity is in the negative z -direction (see fig. 9(a)). Figure(s). 9(b,c) show that the modification to the swimming velocity is relatively small and changes sign with the surface coverage: the swimming velocity increases for $\theta_c < \pi/2$ and reduces for $\theta_c > \pi/2$.

For a repulsive interaction, U_{λ_0} changes sign (depicting swimming in the opposite direction) as also reported by earlier studies (Sharifi-Mood *et al.* 2013; Michelin &

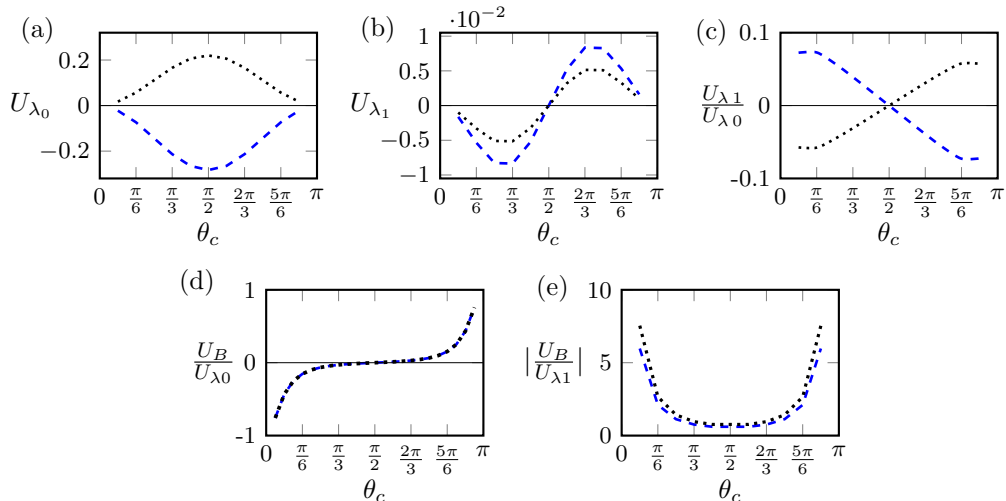


FIGURE 9. (a) Variation of swimming velocity due to Newtonian surface slip (U_{λ_0}) for various surface coverages. (b) Perturbation to swimming velocity due to modification in the slip (U_{λ_1}). (c) Ratio of the swimming velocity components i.e. (b) and (a). (d) Variation of modification to the swimming velocity due to bulk polymeric stresses (U_B). (e) Ratio of (d) and (c): comparison of two components arising due to viscoelasticity. Parameters: $De = 0.1$, $\delta = -0.5$, $\zeta = 16$, $\Phi_0 = -1$ (attractive; ---), and $\Phi_0 = +1$ (repulsive; ····).

Lauga 2014; Natale *et al.* 2017). However, U_{λ_1} for a repulsive interaction is in the same direction as that for attractive interaction. In this case, the contribution of U_{λ_1} is such that the swimming velocity is reduced for $\theta_c < \pi/2$ and enhanced for $\theta_c > \pi/2$. This outcome can be understood by comparing the flow field inside the interaction layer for attractive and repulsive cases i.e. fig.2 and fig.3(a), respectively. In comparison to the attractive interaction, Newtonian velocity field was reversed for repulsive interactions, whereas the modification (arising from viscoelasticity) was found to be in the same direction. It is also interesting to note that, contrary to U_{λ_0} , U_{λ_1} is an odd function (antisymmetric about $\pi/2$) which is due to the proportionality to double tangential derivative of surface concentration, whereas the former is solely proportional to the single tangential derivative.

The second integral (U_B) accounts for the contribution from polymeric stresses in the bulk. In their evaluation of swimming velocity, Datt *et al.* (2017) accounted for the bulk viscoelastic effects (U_B), and provided the following analytical expression (re-expressed here using the scales in eq.2.5):

$$\frac{U_B}{U_{\lambda_0}} = -De(1 + \delta) \sum_{n=1}^{\infty} \frac{6n}{(n+1)^2(n+2)} \frac{\alpha_n \alpha_{n+1}}{\alpha_1^2}, \quad (2.32)$$

where $\alpha_n = n\mathcal{K}_n/(2n+1)$ and \mathcal{K}_n is determined using (2.28) and (2.29). The modification due to the bulk stresses (U_B/U_{λ_0}) is plotted in fig.9(d) for smooth activity. The figure shows that U_B can significantly enhance (or impede) the active swimming, depending upon the surface coverage (θ_c) being more (or less) than $\pi/2$. This was also reported by Datt *et al.* (2017) for step activity. A comparison of the two effects of viscoelasticity, U_B and U_{λ_1} (shown in fig.9(e)), reveals that the contribution to the swimming velocity from the modification in slip (U_{λ_1}) is comparable to that from U_B for $5\pi/6 < \theta_c < \pi/6$.

Fig. 10 shows the effect of transition parameter (ζ) on the components of swimming

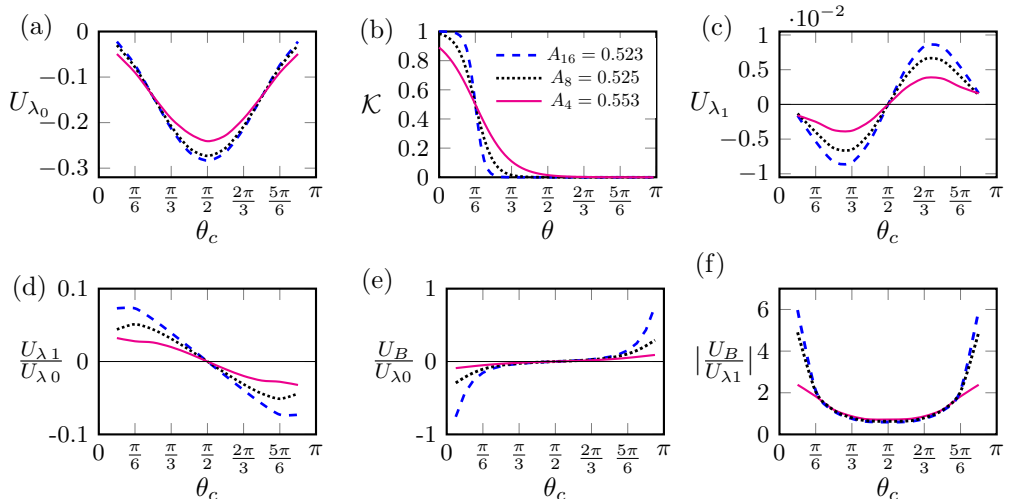


FIGURE 10. Analysis of different components of swimming velocity for three different transition parameters: $\zeta = 4$ (—), $\zeta = 8$ (⋯), $\zeta = 16$ (- - -). (a) Variation of U_{λ_0} with different surface coverages. (b) Variation of activity \mathcal{K} with polar angle θ . (c) Variation of U_{λ_1} for different surface coverages. (d) Contribution from the slip modification to the swimming velocity for different surface coverages. (e) Bulk stress contribution to the swimming velocity for different surface coverages. (f) Comparison of (e) with (d) (i.e. U_B with U_{λ_1}) for different surface coverages. Parameters: $De = 0.1$, $\delta = -0.5$, $\Phi_0 = -1$.

velocity for various surface coverages. As ζ decreases the Newtonian swimming velocity reduces (fig.10a) because the slip velocity decreases (shown in fig.7). However, for $\theta_c < \pi/4$ & $\theta_c > 3\pi/4$, U_{λ_0} is greatest for $\zeta = 4$. This is because, for such coverages, the total area of catalytic activity is more for $\zeta = 4$ than steeper activity transitions ($\zeta = 8, 16$). Fig.10(b) shows the area under the $\mathcal{K} - \theta$ curve for $\zeta = 4, 8, 16$, representing the total area of activity for $\theta_c = \pi/6$. For $\pi/4 < \theta_c < 3\pi/4$, the area of catalytic activity is independent of ζ and thus a direct comparison can be made. Fig.10(c),(d) show that the contribution from U_{λ_1} reduces as ζ decreases due to significant reduction in slip velocity. Similarly, the contribution from bulk polymeric stresses reduces as ζ decreases, as shown in fig.10(e),(f).

3. Active particle in a shear-thinning fluid

The equations governing a weak shear-thinning flow are described by

$$\nabla^* \cdot \mathbf{u}^* = 0, \quad -\nabla^* p^* + \nabla^* \cdot (\mu^* \mathbf{A}^*) - C^* \nabla^* \mathcal{P}^* = \mathbf{0}. \quad (3.1)$$

The viscosity follows a general non-linear relation with respect to Newtonian shear-rate (γ_0) (Khair *et al.* 2012): $\mu^*(\gamma_0) = \mu_0^*(1 + \chi\mu_1(\gamma_0))$. Here, χ is a small parameter which represents the viscosity ratio: $(\mu_0^* - \mu_\infty^*)/\mu_\infty^*$; μ_0^* is the zero shear rate viscosity and μ_∞^* is the infinite shear rate viscosity. The variables are non-dimensionalized as in §2. Since the concentration field is decoupled from the hydrodynamics (provided $\epsilon^2 Pe \ll 1$), the solution to concentration field is unchanged and is given by (2.14a).

3.1. Evaluation of the diffusio-osmotic slip

Similar to §2.1, we use matched asymptotic expansion (assuming $Pe \ll \epsilon^{-2}$ and $\epsilon \ll 1$) and perturbation expansion (in ϵ) to obtain the leading order slip for a generalized weakly shear-thinning fluid.

3.1.1. Outer region

At the leading order, the equations in the outer region are:

$$\nabla \cdot \mathbf{u}^{(0)} = 0, \quad -\nabla p^{(0)} + \mu(\gamma_0) \nabla^2 \mathbf{u}^{(0)} + \nabla \mu(\gamma_0) \cdot \mathbf{A} = \mathbf{0}, \quad (3.2a)$$

subject to the following boundary conditions

$$\mathbf{u}^{(0)} \rightarrow \mathbf{0}, \quad p^{(0)} \rightarrow 0 \quad \text{as } y \rightarrow \infty. \quad (3.3)$$

Here $\mu(\gamma_0) = \mu^*/\mu_0^* = 1 + \chi\mu_1(\gamma_0)$.

3.1.2. Inner region

We rescale the variables (similar to §2.1.2) and obtain the governing equations in the inner region (at the leading order) as:

$$\frac{\partial \hat{u}^{(0)}}{\partial \hat{x}} + \frac{\partial \hat{v}^{(0)}}{\partial \hat{y}} = 0, \quad (3.4a)$$

$$-\frac{\partial \hat{p}^{(0)}}{\partial \hat{x}} + \mu(\gamma_0) \frac{\partial^2 \hat{u}^{(0)}}{\partial \hat{y}^2} + \frac{\partial \mu(\gamma_0)}{\partial \hat{y}} \frac{\partial \hat{u}^{(0)}}{\partial \hat{y}} = (\hat{c}^{(0)} + C_\infty) \frac{\partial \hat{\psi}^{(0)}}{\partial \hat{x}}, \quad (3.4b)$$

$$-\frac{\partial \hat{p}^{(0)}}{\partial \hat{y}} = (\hat{c}^{(0)} + C_\infty) \frac{\partial \hat{\psi}^{(0)}}{\partial \hat{y}}, \quad (3.4c)$$

subject to the pressure decay and no-slip boundary condition

$$\hat{p} \rightarrow 0 \text{ as } \hat{y} \rightarrow 0; \quad \hat{u}^{(0)} \Big|_{\hat{y}=0} = 0. \quad (3.5)$$

Assuming that the difference between zero shear and infinite shear viscosity is small (i.e. $\chi \ll 1$), we perform a regular perturbation expansion in χ and represent the velocity and pressure fields as

$$f^{(i)}(x, y) = f_0^{(i)}(x, y) + \chi f_1^{(i)}(x, y) + \dots \quad (3.6)$$

Substituting the perturbed field variables in (3.4-3.5) gives the equations governing the system at $O(1)$ and $O(\chi)$.

At $O(1)$, the equations and their solutions are identical to (2.14). The hydrodynamics at $O(\chi)$ is governed by

$$\frac{\partial \hat{u}_1^{(0)}}{\partial \hat{x}} + \frac{\partial \hat{v}_1^{(0)}}{\partial \hat{y}} = 0, \quad (3.7a)$$

$$-\frac{\partial \hat{p}_1^{(0)}}{\partial \hat{x}} + \frac{\partial^2 \hat{u}_1^{(0)}}{\partial \hat{y}^2} + \mu_1(\gamma_0) \frac{\partial^2 \hat{u}_0^{(0)}}{\partial \hat{y}^2} + \frac{\partial \mu_1(\gamma_0)}{\partial \hat{y}} \frac{\partial \hat{u}_0^{(0)}}{\partial \hat{y}} = 0, \quad (3.7b)$$

$$-\frac{\partial \hat{p}_1^{(0)}}{\partial \hat{y}} = 0. \quad (3.7c)$$

Integrating (3.7c) and using the pressure decay condition, we get $\hat{p}_1^{(0)} = 0$. (3.7b) thus yields

$$\frac{\partial^2 \hat{u}_1^{(0)}}{\partial \hat{y}^2} = -\frac{\partial}{\partial \hat{y}} \left(\mu_1(\gamma_0) \frac{\partial \hat{u}_0^{(0)}}{\partial \hat{y}} \right) \quad (3.8)$$

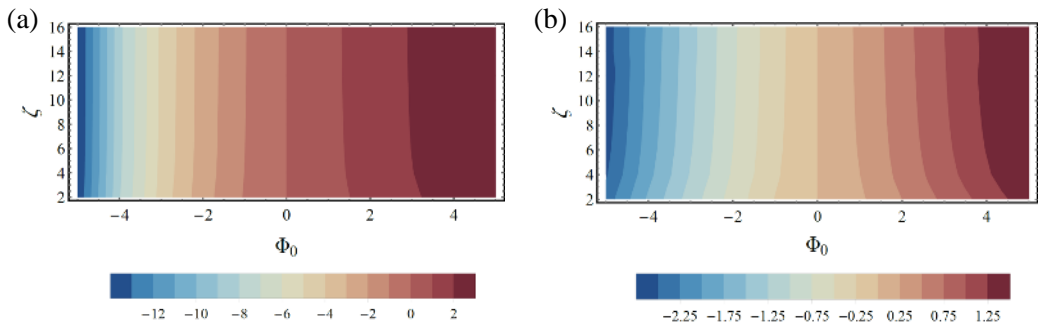


FIGURE 11. Non-Newtonian mobility (M_1) at $\theta = \pi/2$ for two interaction potentials (a) exponential-type and (b) van der Waals-type. Other parameters: $n = 0.25$, $Cu_\lambda = 10^2$, $\theta_c = \pi/2$.

Integrating the above equation twice gives

$$\hat{u}_1^{(0)} = \mathcal{I}'(x) \int_0^{\hat{y}} \mu_1(\gamma_0) \int_t^\infty \mathcal{F}(s) ds dt - \mathcal{J}_0(\hat{x}) \int_0^{\hat{y}} \mu_1(\gamma_0) ds + \mathcal{J}_1(\hat{x}) \hat{y}, \quad (3.9)$$

where $\mathcal{F}(s) = -1 + e^{-\hat{\psi}(s)}$.

3.1.3. Matching

As in §2.1.3, upon matching the inner and outer solutions, we find that $\mathcal{J}_0 = \mathcal{J}_1 = 0$ and obtain:

$$u^{(0)} \Big|_{y=0} = c_x^{(0)} \Big|_{y=0} (M_0 + \chi M_1), \quad (3.10)$$

where M_0 is the mobility coefficient representing the Newtonian contribution and is expressed in (2.20). The expression for M_1 is

$$M_1 = \int_0^\infty \mu_1(\gamma_0^{(0)}) \int_t^\infty \mathcal{F}(s) ds dt. \quad (3.11)$$

Since the power-law model diverges for asymptotically small shear-rates (Bird *et al.* 1987), we choose the Carreau model to capture viscosity variation: $\mu_1 = (1 + \tau^{*2} |\dot{\gamma}_0^*|^2)^{\frac{n-1}{2}} - 1$ (Bird *et al.* 1987). Here τ^* is the fluid relaxation time scale and n characterizes the degree of shear-thinning ($n < 1$). For Carreau fluid, the non-Newtonian mobility coefficient (M_1) is

$$M_1 = \int_0^\infty \left\{ \left[1 + \left(Cu_\lambda c_x^{(0)} \Big|_{y=0} \int_t^\infty \mathcal{F}(s) ds \right)^2 \right]^{\frac{n-1}{2}} - 1 \right\} \int_t^\infty \mathcal{F}(s) ds dt, \quad (3.12)$$

where Cu_λ (based on shear in the interaction layer) is the ratio of timescales associated with relaxation (τ^*) to that of shear in the flow (λ_I^*/U_{ch}): $Cu_\lambda = \tau^*/(\lambda_I^*/U_{ch})$. This is different than the Carreau number corresponding to shear in the bulk fluid (i.e. outer region): $Cu_B = \tau^*/(a^*/U_{ch})$. The ratio of the two Carreau numbers ($Cu_\lambda/Cu_B = a^*/\lambda_I^*$) provides the comparison of shear rate inside to that outside the interaction layer i.e. $\sim O(\epsilon^{-1})$.

3.2. Diffusio-osmotic slip on an active particle

As in §2.3, we now apply the above results to an axi-symmetric Janus sphere. Since the shear in the thin interaction layer is generally high (i.e. $U_{ch}/\lambda_I^* \sim 10^2 s^{-1}$) and

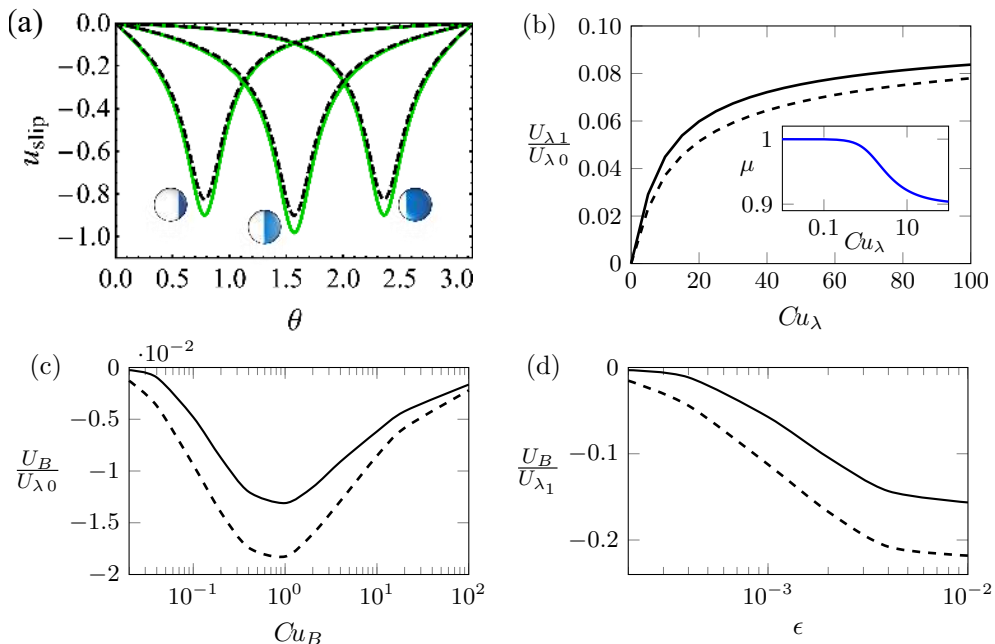


FIGURE 12. (a) The dashed black line represents the Newtonian slip velocity. The solid green line represents the variation of total slip velocity for a shear thinning fluid along the polar angle for $\hat{\psi}(\xi) = -e^{-\xi}$ and $Cu_\lambda = 100$. (b) Variation of non-Newtonian contribution to the swimming velocity (arising from the surface slip) with Cu_λ , for three different surface coverage (θ_c) and $\hat{\psi}(\xi) = -e^{-\xi}$. Solid (—) and dashed lines (---) correspond to $\theta_c = \pi/2$ and $\theta_c = \pi/4$ or $3\pi/4$, respectively. Inset showing the variation in non-dimensional viscosity μ with Cu_λ at $y = 0$ and $\theta = \theta_c = \pi/2$. (c) Variation of non-Newtonian contribution to the swimming velocity (arising from the bulk stresses) with Cu_B . The convergence was obtained by accounting for first 15 modes. (d) Comparison between the two non-Newtonian components of swimming velocity for $Cu_\lambda = 10^2$. Other parameters: $\chi = 0.1$, $n = 0.25$, $\zeta = 16$.

relaxation time scales of the biological fluids are $\sim 1s$ (Zare *et al.* 2019), large Carreau numbers are possible ($Cu_\lambda \sim 10^2$). Contrary to the second-order fluid, M_1 for Carreau fluid depends both on the magnitude of interaction (Φ_0) as well as the concentration gradient (determined by the activity transition parameter ζ). Since the concentration field varies in tangential direction, M_1 varies tangentially. Thus, we analyze the effect of Φ_0 and ζ on M_1 for $\theta = \theta_c = \pi/2$ in fig.11. For both short-range exponential and long-range van der Waals interactions, M_1 is a weak function of ζ . M_1 for Carreau fluid is of different signs for attractive ($\Phi_0 < 0$) and repulsive interactions ($\Phi_0 > 0$), as opposed to the case of second-order fluid (fig.4b).

Using (2.24), (2.28), (3.10), and (3.12), we numerically evaluate the slip for shear-thinning fluid ($n = 0.25$) and depict its variation along the surface of Janus particle in fig. 12 (a). For large Carreau numbers ($Cu_\lambda \sim 10^2$) and attractive interactions, we find a marginal increase in the total slip velocity. For repulsive interactions, the enhancement in slip velocity is qualitatively similar (see Appendix A). The modification in slip (in both attractive and repulsive cases) is maximum at the transition of activity, depicting a local symmetry about θ_c .

3.3. Swimming velocity

We now evaluate the swimming velocity using the reciprocal theorem (Stone & Samuel 1996; Khair *et al.* 2012):

$$U = \frac{-1}{4\pi} \int_{S_p} (\mathbf{u}_0 + \chi \mathbf{u}_1)|_{r=1} \cdot \mathbf{e}_z \, dS - \frac{1}{6\pi} \chi \int_{V_f} \mu_1(\gamma_0) \mathbf{A}_0 : \nabla \mathbf{u}^t \, dV. \quad (3.13)$$

As in §2.4, the first integral contains the Newtonian component U_{λ_0} (given by eq.2.31) and non-Newtonian component U_{λ_1} :

$$U_{\lambda_1} = \frac{\chi}{2} \int_0^\pi M_1(\theta) c_\theta(1, \theta) \sin^2 \theta \, d\theta. \quad (3.14)$$

Substituting (3.12) in (3.13) and integrating numerically, we find that the contribution from U_{λ_1} modestly affects the swimming of a Janus particle. This is shown in figure 12(b) for three different surface coverages. The positive sign of the ratio shows that U_{λ_1} adds to the contribution arising from the Newtonian slip (U_{λ_0}). As Cu_λ increases, the viscosity in the interaction layer decreases, which results in a faster diffusio-osmotic flow. However, this enhancement reaches a plateau as the viscosity reduction stagnates at high Cu_λ (see inset in fig.12b). For repulsive interactions, this enhancement in swimming velocity (U_{λ_1}) is qualitatively similar (see Appendix A); shear thinning enhances the swimming velocity, irrespective of the interaction between solute molecules and the particle.

The second integral (U_B) accounts for the bulk stresses arising from viscosity variations. The modification due to bulk non-Newtonian stresses is quantified in fig.12(c) for three different surface coverages†. The negative sign of U_B/U_{λ_0} denotes that the bulk stresses always reduce the swimming velocity, for all surface coverages (also reported by Datt *et al.* (2017) for step activity). The non-monotonic behavior is consistent with the fact that a viscosity, following Carreau model, reduces to a Newtonian fluid of a lower viscosity (μ_∞^*) as $Cu_B \rightarrow \infty$. As a result, the bulk non-Newtonian stresses are maximum at intermediate Cu_B and vanish at high Cu_B (Khair *et al.* 2012; Datt *et al.* 2015, 2017). Fig.12(d) compares the contribution from the two non-Newtonian components to swimming velocity i.e. U_B and U_{λ_1} . We take $Cu_\lambda = 10^2$, as shear rate in the interaction layer is generally high. This yields $Cu_B = (\epsilon)10^2$. Thus, in fig.12(d), we compare the two components, keeping ϵ as an independent variable. Since the current analysis is valid for asymptotically thin interaction layers, we vary ϵ from 10^{-4} to 10^{-2} . Physically, $Cu_\lambda/Cu_B = \epsilon^{-1}$ corresponds to the disparity in shear rates in the inner and outer regions. For $\epsilon \sim 10^{-4}$, the shear rate inside the interaction layer is very high and thus the contribution from U_{λ_1} dominates that from U_B . As ϵ increases (i.e. thickness of interaction layer), the shear in the interaction layer decreases, which increases the magnitude of U_B/U_{λ_1} . This continues until U_B reaches a plateau.‡

The contribution from both non-Newtonian components is modest in magnitude. In context of electrophoresis, Khair *et al.* (2012) also reported a similar enhancement and trends in a shear-thinning medium. Analogous to their study, our result (3.10) also reveals that the shear-thinning modification of the slip does not add a size dependency to the motion induced due to self-diffusiophoresis. On the other hand, the modification due to bulk non-Newtonian stresses adds a size dependency (it enters through the bulk Carreau number).

† We follow the approach of Blake (1971); Datt *et al.* (2017) to find U_B for smooth activity. The details of implementation and reproduced results (for step activity) are provided in the supplementary material.

‡ The above results are for $\zeta = 16$. Appendix A shows the effect ζ on the swimming velocity.

4. Conclusions

The results of the current study reveal the effects of fluid rheology on the diffusio-osmotic slip over an active surface and its consequence on the mobility of a Janus particle. Using matched asymptotic expansions in conjunction with perturbation theory, we derived the modification to slip velocity for a second-order fluid. Our result (eq.2.19), for an axisymmetrically active Janus particle, shows that the polymeric stress significantly alters the slip velocity, and is valid for low to moderate advective effects. The proportionality to second tangential gradients results in a sharp reversal of the surface slip, triggered by large gradient in polymeric stress across the discontinuity of surface activity. This explains the generation of extensional flow across a step change in activity observed by Natale *et al.* (2017). An examination of characteristic scales reveals that the localized reversal of slip velocity is a result of mathematical inconsistency: a consequence of employing the activity as a step function, which can be overcome by choosing a smooth (sigmoidal) function. The results reveal that the polymer elasticity tends to generate local elongational flows across the transition of surface activity. We also explored the effects of attractive and repulsive interaction between the solute molecules and Janus particle. Using the reciprocal theorem, we found that the modification in the slip has an effect on the swimming velocity which is comparable to that arising from bulk viscoelastic stresses (Datt *et al.* 2017).

We further applied the framework to a generalized weakly shear-thinning fluid and obtained the modification to slip velocity (eq.3.10). Employing a Carreau-fluid model, we showed that the shear-thinning effects marginally increase the slip velocity, provided the time scale associated with the shear in the interaction layer is asymptotically smaller than the fluid relaxation time. Using the reciprocal theorem, we showed an enhancement in the swimming velocity due to modification in slip. For an asymptotically thin interaction layer, this enhancement dominates the retardation caused by the bulk non-Newtonian stress.

Another key implication of our results is that the modification in slip provides an estimate of the hydrodynamic disturbance around an active particle for weak non-Newtonian effects. This can be a significant contribution towards accurate modeling of the interaction of two or more active particles in polymeric fluids (Rallabandi *et al.* 2019; Stark 2018). Recently, Michelin *et al.* (2015); Michelin & Lauga (2019) have designed ‘phoretic pumps’ which can transport fluids without the need of applying pressure difference across the channel. This can be helpful in transportation of biological fluids through narrow channels. The flow in such systems occurs due to local concentration gradients, arising either from geometric variation (Lisicki *et al.* 2016; Michelin *et al.* 2015) or variation in surface activity (Michelin & Lauga 2019). Since our results are applicable to a general diffusio-osmotic slip, they should be useful to model the flow of complex fluids through such pumps.

The current work and previous investigations corresponding to the active motion in complex fluids (Datt *et al.* 2017; Natale *et al.* 2017) have assumed the diffusivity of solute molecules to be constant. This might be reasonable in the limit of weak non-Newtonian effects, but may lead to imprecise conclusions for fluid mediums exhibiting strong non-Newtonian behavior, as the Stokes-Einstein equation fails in describing diffusion in complex media. Recently, Makuch *et al.* (2020) devised a relationship between translational and rotational diffusion coefficients which depends on the size of solute. Such theoretical formulation can provide a database for precise description of diffusion in various complex fluids. Such data can be employed in a theoretical framework similar to that devised by Vrentas & Vrentas (2003) and Tiefenbruck & Leal (1980) who studied

the effect of diffusion in non-Newtonian flows. Further research in this direction can help in a better understanding of self-propulsion through the mediums which exhibit significant deviations from the Newtonian behavior.

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Declaration of Interests

The authors report no conflict of interest.

Appendix A. Additional results for the shear-thinning fluid

Fig.13(a) shows the velocity field inside the thin interaction layer for Carreau fluid and a sigmoidal activity transition (2.28). The contours bear close resemblance to that for Newtonian fluids 8(a). Thus, we plot the difference between the two in fig.13(b). The local viscosity reduction due to shear-thinning increases the velocity inside the inner region and consequently the slip (as observed in fig.12(a)). This enhancement is symmetric about θ_c .

Fig.14 shows the results for repulsive exponential interactions between the solute molecules and active surface. Fig.14(a) depicts the slip velocity, which is qualitatively reversed in comparison to that depicted for the attractive interactions (see fig. 12(a)). The magnitude is lower in this case because the adsorption coefficient is reduced in repulsive interactions (Anderson *et al.* 1982). Figure 14(b) shows that the enhancement caused by U_{λ_I} is qualitatively similar to that observed in the case of attractive interactions. As the ratio U_B/U_{λ_0} is independent of mobility, its profile is identical to fig.12(c).

As in §2.4, we now analyze the effect of transition parameter (ζ) on the non-Newtonian components of the swimming velocity. ζ determines the magnitude of shear inside the interaction layer and slip velocity. Fig.15(a) shows that the transition parameter has a small effect on the contribution due to slip modification. This is because, M_1 has a weak dependency on ζ (see fig.11), which endows U_{λ_1} to have a ζ dependency similar to that of U_{λ_0} (c.f eq.3.14 & eq.2.31). Thus, the ratio ($U_{\lambda_1}/U_{\lambda_0}$) does not change substantially with ζ . Fig.15(b) depicts that, at low to moderate Cu_B , the contribution due to bulk stresses decreases significantly with decrease in ζ (as the shear rate $|\dot{\gamma}_0|$ reduces). As Cu_B increases, the bulk non-Newtonian stresses reduce because the flow approaches a

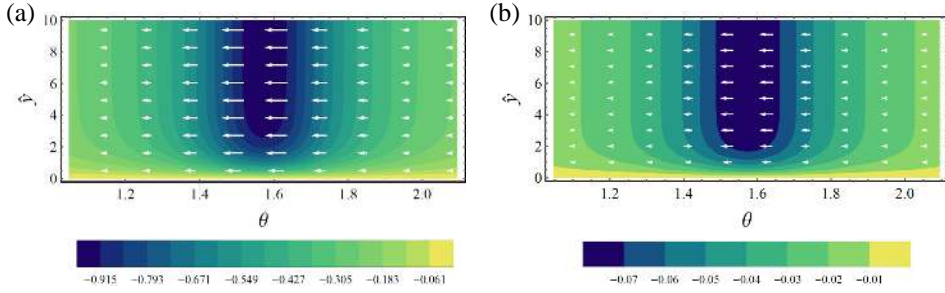


FIGURE 13. Tangential velocity contours inside the interaction layer for: (a) Newtonian fluid; (b) a weakly shear-thinning fluid. The arrows depict the velocity field vector $\hat{\mathbf{u}}_1^{(0)}$. Other parameters: $\chi = 0.1$, $n = 1/4$, $\zeta = 16$, $\hat{\psi}(\xi) = -e^{-\xi}$.

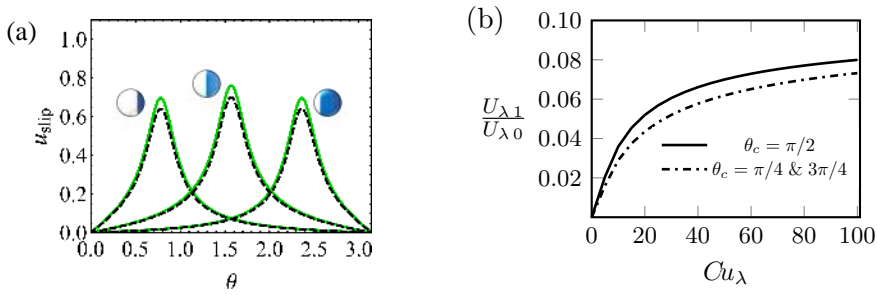


FIGURE 14. (a) The dashed line represents the Newtonian slip velocity. The solid line represents the variation of total slip velocity along the polar angle for $\hat{\psi}(\xi) = +e^{-\xi}$ and $Cu_\lambda = 100$. (b) Variation of swimming velocity (arising from the surface slip) with Cu_λ , for three different surface coverage (θ_c). Other parameters: $\chi = 0.1$, $n = 1/4$, $\zeta = 16$.

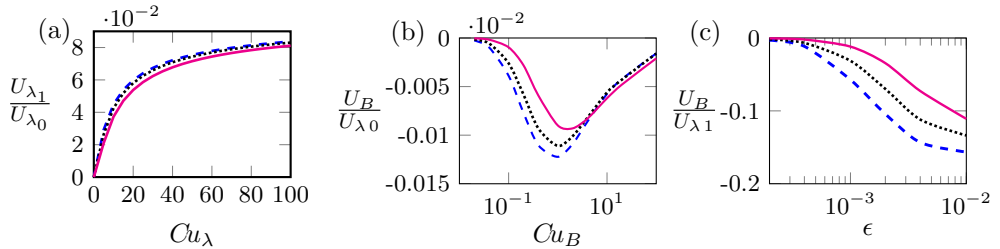


FIGURE 15. Analysis of different components of swimming velocity for three different transition parameters: $\zeta = 4$ (—), $\zeta = 8$ (⋯⋯), $\zeta = 16$ (- - -). (a) Contribution from the slip modification to the swimming velocity for different surface coverages and $\hat{\psi}(\xi) = -e^{-\xi}$. (b) Bulk stress contribution to the swimming velocity for different surface coverages. (c) Comparison of two non-Newtonian contributions to the swimming for $Cu_\lambda = 10^2$. Other parameters: $\chi = 0.1$, $n = 0.25$, $\theta_c = \pi/2$.

Newtonian state i.e. $\mu^* \rightarrow \mu_\infty^*$. For a lower ζ , the shear ($|\gamma_0|$) is reduced and thus, the reduction in non-Newtonian stresses occurs at a higher Cu_B . As a result, the trend reverses for $Cu_B > 10$ in fig.15(b): $\zeta = 4$ is greater than $\zeta = 8, 16$. Fig.15(c) compares the two components over the range of interaction layer thickness.

Supplementary Material

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In this Supplementary Material we provide details of the derivation of slip velocity in a second-order fluid.

S.1 Evaluation of $O(De)$ solution in the inner region

The flow field at $\mathcal{O}(De)$ is governed by

$$\frac{\partial \hat{u}_1}{\partial \hat{x}} + \frac{\partial \hat{v}_1}{\partial \hat{y}} = 0, \quad (\text{S1a})$$

$$-\frac{\partial \hat{p}_1}{\partial \hat{x}} + \frac{\partial^2 \hat{u}_1}{\partial \hat{y}^2} + \frac{\partial \hat{S}_{xx0}}{\partial \hat{x}} + \frac{\partial \hat{S}_{xy0}}{\partial \hat{y}} = 0, \quad (\text{S1b})$$

$$-\frac{\partial \hat{p}_1}{\partial \hat{y}} + \frac{\partial \hat{S}_{yy0}}{\partial \hat{y}} = 0. \quad (\text{S1c})$$

Here the subscripts on S denote the components of polymeric stress tensor. The components of the polymeric stress tensor (\mathbf{S}) are:

$$\hat{S}_{xx} = \left(\frac{\partial \hat{u}}{\partial \hat{y}} \right)^2, \quad \hat{S}_{yy} = (1 + 2\delta) \left(\frac{\partial \hat{u}}{\partial \hat{y}} \right)^2, \quad (\text{S2a})$$

$$\hat{S}_{xy} = \hat{S}_{yx} = 2\delta \left(\frac{\partial \hat{u}}{\partial \hat{y}} \frac{\partial \hat{u}}{\partial \hat{x}} + \frac{\hat{v} \frac{\partial^2 \hat{u}}{\partial \hat{y}^2} + \hat{u} \frac{\partial^2 \hat{u}}{\partial \hat{y} \partial \hat{x}}}{2} \right). \quad (\text{S2b})$$

Using the pressure decay condition: $\hat{p} \rightarrow 0$ as $\hat{y} \rightarrow \infty$ (Michelin & Lauga 2014, pg.579), the solution to (S1c) yields $\hat{p}_1 = \hat{S}_{yy0} - \mathcal{J}_0(\hat{x})^2(1 + 2\delta)$, where \mathcal{J}_0 is later shown to be zero through matching. Substituting \hat{p}_1 in (S1b), we obtain

$$\frac{\partial^2 \hat{u}_1}{\partial \hat{y}^2} + 2\mathcal{J}_0(\hat{x})\mathcal{J}'_0(\hat{x})(1 + 2\delta) = \frac{\partial \hat{S}_{yy0}}{\partial \hat{x}} - \left[\frac{\partial \hat{S}_{xx0}}{\partial \hat{x}} + \frac{\partial \hat{S}_{xy0}}{\partial \hat{y}} \right], \quad (\text{S3a})$$

$$= 2(1 + 2\delta)u_y u_{yx} - \left[2u_y u_{yx} + 2\delta \left(u_{yy} u_x + u_y u_{yx} + \frac{v_y u_{yy} + v u_{yyy} + u_y u_{yx} + u u_{yyx}}{2} \right) \right]. \quad (\text{S3b})$$

Here we have substituted (S2a) in (S3a) to obtain (S3b). The subscripts in above equation (and the next) denote the derivative of u and v with respect to x or y . We also have temporarily dropped the hat symbol and subscript 0 in the right hand side. The coloured terms (blue) get canceled out and using the continuity equation ($v_y = -u_x$), we simplify the equation as:

$$\frac{\partial^2 \hat{u}_1}{\partial \hat{y}^2} + 2\mathcal{J}_0(\hat{x})\mathcal{J}'_0(\hat{x})(1 + 2\delta) = 2\delta \left\{ 2u_y u_{yx} - \left[u_{yy} u_x + u_y u_{yx} + \frac{-u_x u_{yy} + v u_{yyy} + u_y u_{yx} + u u_{yyx}}{2} \right] \right\}, \quad (\text{S4a})$$

$$= 2\delta \left\{ 2u_y u_{yx} - \left[\frac{u_{yy} u_x}{2} + \frac{3u_y u_{yx}}{2} + \frac{v u_{yyy} + u u_{yyx}}{2} \right] \right\}, \quad (\text{S4b})$$

$$= 2\delta \left\{ \frac{u_y u_{yx}}{2} - \left[\frac{u_{yy} u_x}{2} + \frac{v u_{yyy} + u u_{yyx}}{2} \right] \right\}, \quad (\text{S4c})$$

$$= -\delta \left\{ -u_y u_{yx} + u_{yy} u_x + v u_{yyy} + u u_{yyx} \right\}. \quad (\text{S4d})$$

Dropping the subscript notation (for derivative), we obtain the differential equation for \hat{u}_1 as:

$$\frac{\partial^2 \hat{u}_1}{\partial \hat{y}^2} = -\delta \left\{ -\frac{\partial \hat{u}_0}{\partial \hat{y}} \frac{\partial^2 \hat{u}_0}{\partial \hat{y} \partial \hat{x}} + \frac{\partial \hat{u}_0}{\partial \hat{x}} \frac{\partial^2 \hat{u}_0}{\partial \hat{y}^2} + \hat{v}_0 \frac{\partial^3 \hat{u}_0}{\partial \hat{y}^3} + \hat{u}_0 \frac{\partial^3 \hat{u}_0}{\partial \hat{x} \partial \hat{y}^2} \right\} - 2\mathcal{J}_0(\hat{x})\mathcal{J}'_0(\hat{x})(1 + 2\delta). \quad (\text{S5})$$

We will now substitute the $O(De^0)$ solution in the above equation. \hat{u}_0 and its derivatives, and \hat{v}_0 (obtained from the continuity equation) are:

$$\hat{u}_0 = -\mathcal{I}'(\hat{x}) \int_0^{\hat{y}} \int_t^\infty \mathcal{F}(s) ds dt + \mathcal{J}_0(x)\hat{y}, \quad (\text{S6a})$$

$$\frac{\partial \hat{u}_0}{\partial \hat{y}} = -\mathcal{I}'(\hat{x}) \int_{\hat{y}}^\infty \mathcal{F}(s) ds + \mathcal{J}_0(x), \quad (\text{S6b})$$

$$\frac{\partial^2 \hat{u}_0}{\partial \hat{y}^2} = \mathcal{I}'(\hat{x})\mathcal{F}(\hat{y}), \quad (\text{S6c})$$

$$\hat{v}_0 = -\int_0^{\hat{y}} \frac{\partial \hat{u}_0}{\partial \hat{x}} dy + \mathcal{C}(\hat{x}) = +\mathcal{I}''(\hat{x}) \int_0^{\hat{y}} \int_0^r \int_t^\infty \mathcal{F}(s) ds dt dr - \frac{\mathcal{J}'_0(\hat{x})\hat{y}^2}{2} + \mathcal{C}(\hat{x}). \quad (\text{S6d})$$

Using the no-penetration condition at the surface, we find that $\mathcal{C}(\hat{x}) = 0$.

Substituting (S6) in the bracket terms of (S5), we obtain the four terms $\{A + B + C + D\}$ as:

$$A = -\left[\left(-\mathcal{I}'(\hat{x}) \int_{\hat{y}}^\infty \mathcal{F}(s) ds + \mathcal{J}_0(x) \right) \left(-\mathcal{I}''(\hat{x}) \int_{\hat{y}}^\infty \mathcal{F}(s) ds + \mathcal{J}'_0(\hat{x}) \right) \right], \quad (\text{S7a})$$

$$B = \left(-\mathcal{I}''(\hat{x}) \int_0^{\hat{y}} \int_t^\infty \mathcal{F}(s) ds dt + \mathcal{J}'_0(x)\hat{y} \right) (\mathcal{I}'(\hat{x})\mathcal{F}(\hat{y})), \quad (\text{S7b})$$

$$C = \left(\mathcal{I}''(\hat{x}) \int_0^{\hat{y}} \int_0^r \int_t^\infty \mathcal{F}(s) ds dt dr - \frac{\mathcal{J}'_0(\hat{x})\hat{y}^2}{2} \right) (\mathcal{I}'(\hat{x})\mathcal{F}'(\hat{y})), \quad (\text{S7c})$$

$$D = \left(-\mathcal{I}'(\hat{x}) \int_0^{\hat{y}} \int_t^\infty \mathcal{F}(s) ds dt + \mathcal{J}_0(\hat{x})\hat{y} \right) (\mathcal{I}''(\hat{x})\mathcal{F}(\hat{y})). \quad (\text{S7d})$$

These terms can be further simplified as

$$A = -\left[+\mathcal{I}'(\hat{x})\mathcal{I}''(\hat{x}) \left(\int_{\hat{y}}^\infty \mathcal{F}(s) ds \right)^2 - \mathcal{J}_0(\hat{x})\mathcal{I}''(\hat{x}) \left(\int_{\hat{y}}^\infty \mathcal{F}(s) ds \right) - \mathcal{I}'(\hat{x})\mathcal{J}'_0(\hat{x}) \left(\int_{\hat{y}}^\infty \mathcal{F}(s) ds \right) + \mathcal{J}_0(\hat{x})\mathcal{J}'_0(\hat{x}) \right], \quad (\text{S8a})$$

$$B = -\mathcal{I}'(\hat{x})\mathcal{I}''(\hat{x})\mathcal{F}(\hat{y}) \left(\int_0^{\hat{y}} \int_t^\infty \mathcal{F}(s) ds dt \right) + \mathcal{I}'(\hat{x})\mathcal{J}'_0(\hat{x})\hat{y}\mathcal{F}(\hat{y}), \quad (\text{S8b})$$

$$C = \mathcal{I}'(\hat{x})\mathcal{I}''(\hat{x})\mathcal{F}'(\hat{y}) \left(\int_0^{\hat{y}} \int_0^r \int_t^\infty \mathcal{F}(s) ds dt dr \right) - \mathcal{J}'_0(\hat{x})\mathcal{I}'(\hat{x})\mathcal{F}'(\hat{y})\frac{\hat{y}^2}{2}, \quad (\text{S8c})$$

$$D = -\mathcal{I}'(\hat{x})\mathcal{I}''(\hat{x})\mathcal{F}(\hat{y}) \left(\int_0^{\hat{y}} \int_t^\infty \mathcal{F}(s) ds dt \right) + \mathcal{J}_0(\hat{x})\mathcal{I}''(\hat{x})\hat{y}\mathcal{F}(\hat{y}). \quad (\text{S8d})$$

Substituting the above four terms in (S5) and simplifying we get:

$$\begin{aligned} \frac{\partial^2 \hat{u}_1}{\partial y^2} = & -\delta \mathcal{I}'(\hat{x}) \mathcal{I}''(\hat{x}) \left\{ - \left(\int_{\hat{y}}^{\infty} \mathcal{F}(s) ds \right)^2 - 2 \mathcal{F}(\hat{y}) \left(\int_0^{\hat{y}} \int_t^{\infty} \mathcal{F}(s) ds dt \right) \right. \\ & + \mathcal{F}'(\hat{y}) \left(\int_0^r \int_0^{\omega} \int_t^{\infty} \mathcal{F}(s) ds dt d\omega \right) + \\ & + \left(\frac{\mathcal{J}_0(\hat{x})}{\mathcal{I}'(\hat{x})} + \frac{\mathcal{J}'_0(\hat{x})}{\mathcal{I}''(\hat{x})} \right) \left(\hat{y} \mathcal{F}(\hat{y}) + \int_{\hat{y}}^{\infty} \mathcal{F}(s) ds \right) \\ & \left. - \frac{\mathcal{J}_0(\hat{x})}{\mathcal{I}''(\hat{x})} \mathcal{F}'(\hat{y}) \frac{\hat{y}^2}{2} - \frac{\mathcal{J}_0(\hat{x}) \mathcal{J}'_{(0)}(\hat{x})}{\mathcal{I}'(\hat{x}) \mathcal{I}''(\hat{x})} + \frac{2 \mathcal{J}_0(\hat{x}) \mathcal{J}'_0(\hat{x}) (1+2\delta)}{\delta \mathcal{I}'(\hat{x}) \mathcal{I}''(\hat{x})} \right\}. \quad (\text{S9}) \end{aligned}$$

Integrating the above equation once, we obtain

$$\begin{aligned} \frac{\partial \hat{u}_1}{\partial y} = & C_1 - \delta \mathcal{I}'(\hat{x}) \mathcal{I}''(\hat{x}) \int_0^{\hat{y}} \left\{ - \left(\int_r^{\infty} \mathcal{F}(s) ds \right)^2 - 2 \mathcal{F}(r) \left(\int_0^r \int_t^{\infty} \mathcal{F}(s) ds dt \right) \right. \\ & + \mathcal{F}'(r) \left(\int_0^r \int_0^{\omega} \int_t^{\infty} \mathcal{F}(s) ds dt d\omega \right) \\ & + \left(\frac{\mathcal{J}_0(\hat{x})}{\mathcal{I}'(\hat{x})} + \frac{\mathcal{J}'_0(\hat{x})}{\mathcal{I}''(\hat{x})} \right) \left(r \mathcal{F}(r) + \int_r^{\infty} \mathcal{F}(s) ds \right) \\ & \left. - \frac{\mathcal{J}_0(\hat{x})}{\mathcal{I}''(\hat{x})} \mathcal{F}'(r) \frac{r^2}{2} - \frac{\mathcal{J}_0(\hat{x}) \mathcal{J}'_{(0)}(\hat{x})}{\mathcal{I}'(\hat{x}) \mathcal{I}''(\hat{x})} + \frac{2 \mathcal{J}_0(\hat{x}) \mathcal{J}'_0(\hat{x}) (1+2\delta)}{\delta \mathcal{I}'(\hat{x}) \mathcal{I}''(\hat{x})} \right\} dr. \quad (\text{S10}) \end{aligned}$$

As $\hat{y} \rightarrow \infty$, $\frac{\partial \hat{u}_1}{\partial y} = \mathcal{J}_1(\hat{x})$ which is to be determined through matching. Simplifying the above equation (replacing $C_1 = \mathcal{J}_1 + \delta \mathcal{I}' \mathcal{I}'' \int_0^{\infty} \dots dr$), we get:

$$\begin{aligned} \frac{\partial \hat{u}_1}{\partial y} = & \mathcal{J}_1(\hat{x}) + \delta \mathcal{I}'(\hat{x}) \mathcal{I}''(\hat{x}) \int_{\hat{y}}^{\infty} \left\{ - \left(\int_r^{\infty} \mathcal{F}(s) ds \right)^2 - 2 \mathcal{F}(r) \left(\int_0^r \int_t^{\infty} \mathcal{F}(s) ds dt \right) \right. \\ & + \mathcal{F}'(r) \left(\int_0^r \int_0^{\omega} \int_t^{\infty} \mathcal{F}(s) ds dt d\omega \right) \\ & + \left(\frac{\mathcal{J}_0(\hat{x})}{\mathcal{I}'(\hat{x})} + \frac{\mathcal{J}'_0(\hat{x})}{\mathcal{I}''(\hat{x})} \right) \left(r \mathcal{F}(r) + \int_r^{\infty} \mathcal{F}(s) ds \right) - \frac{\mathcal{J}_0(\hat{x})}{\mathcal{I}''(\hat{x})} \mathcal{F}'(r) \frac{r^2}{2} \\ & \left. - \frac{\mathcal{J}_0(\hat{x}) \mathcal{J}'_{(0)}(\hat{x})}{\mathcal{I}'(\hat{x}) \mathcal{I}''(\hat{x})} + \frac{2 \mathcal{J}_0(\hat{x}) \mathcal{J}'_0(\hat{x}) (1+2\delta)}{\delta \mathcal{I}'(\hat{x}) \mathcal{I}''(\hat{x})} \right\} dr. \quad (\text{S11}) \end{aligned}$$

Integrating once more and using the no-slip condition (renders the constant C_2 zero), we get:

$$\begin{aligned} \hat{u}_1 = & \mathcal{J}_1(\hat{x}) \hat{y} - \delta \mathcal{I}'(\hat{x}) \mathcal{I}''(\hat{x}) \int_0^{\hat{y}} dp \int_p^{\infty} \left\{ \left(\int_r^{\infty} \mathcal{F}(s) ds \right)^2 + 2 \mathcal{F}(r) \left(\int_0^r \int_t^{\infty} \mathcal{F}(s) ds dt \right) \right. \\ & - \mathcal{F}'(r) \left(\int_0^r \int_0^{\omega} \int_t^{\infty} \mathcal{F}(s) ds dt d\omega \right) \\ & - \left(\frac{\mathcal{J}_0(\hat{x})}{\mathcal{I}'(\hat{x})} + \frac{\mathcal{J}'_0(\hat{x})}{\mathcal{I}''(\hat{x})} \right) \left(r \mathcal{F}(r) + \int_r^{\infty} \mathcal{F}(s) ds \right) \\ & \left. + \frac{\mathcal{J}_0(\hat{x})}{\mathcal{I}''(\hat{x})} \mathcal{F}'(r) \frac{r^2}{2} + \frac{\mathcal{J}_0(\hat{x}) \mathcal{J}'_{(0)}(\hat{x})}{\mathcal{I}'(\hat{x}) \mathcal{I}''(\hat{x})} - \frac{2 \mathcal{J}_0(\hat{x}) \mathcal{J}'_0(\hat{x}) (1+2\delta)}{\delta \mathcal{I}'(\hat{x}) \mathcal{I}''(\hat{x})} \right\} dr. \quad (\text{S12}) \end{aligned}$$

Here $\mathcal{F}(r) = -1 + e^{-\hat{\psi}(r)}$ and $\mathcal{F}'(r) = -\hat{\psi}'(r) e^{-\hat{\psi}(r)}$.

We now reduce the integrals (inside the bracket) by changing the order of integration:

$$2\mathcal{F}(r) \left(\int_0^r \int_t^\infty \mathcal{F}(s) ds dt \right) dr = 2\mathcal{F}(r) \left(\int_0^r s\mathcal{F}(s) ds + r \int_r^\infty \mathcal{F}(s) ds \right) dr \text{ and}$$

$$\int_0^r \int_0^\omega \int_t^\infty \mathcal{F}(s) ds dt d\omega = \int_0^r \left(r - \frac{s}{2} \right) s\mathcal{F}(s) ds + \frac{r^2}{2} \int_r^\infty \mathcal{F}(s) ds. \quad (\text{S13})$$

Substituting the above simplification in (S12), we obtain

$$\begin{aligned} \hat{u}_1 = \mathcal{J}_1(\hat{x})\hat{y} - \delta\mathcal{I}'(\hat{x})\mathcal{I}''(\hat{x}) \int_0^{\hat{y}} dp \int_p^\infty \left\{ \left(\int_r^\infty \mathcal{F}(s) ds \right)^2 + 2\mathcal{F}(r) \left(\int_0^r s\mathcal{F}(s) ds + r \int_r^\infty \mathcal{F}(s) ds \right) \right. \\ \left. + \hat{\psi}'(r)e^{-\hat{\psi}(r)} \left(\int_0^r \left(r - \frac{s}{2} \right) s\mathcal{F}(s) ds + \frac{r^2}{2} \int_r^\infty \mathcal{F}(s) ds \right) \right. \\ \left. - \left(\frac{\mathcal{J}_0(\hat{x})}{\mathcal{I}'(\hat{x})} + \frac{\mathcal{J}'_0(\hat{x})}{\mathcal{I}''(\hat{x})} \right) \left(r\mathcal{F}(r) + \int_r^\infty \mathcal{F}(s) ds \right) + \frac{\mathcal{J}_0(\hat{x})}{\mathcal{I}''(\hat{x})} \mathcal{F}'(r) \frac{r^2}{2} \right. \\ \left. + \frac{\mathcal{J}_0(\hat{x})\mathcal{J}'_0(\hat{x})}{\mathcal{I}'(\hat{x})\mathcal{I}''(\hat{x})} - \frac{2\mathcal{J}_0(\hat{x})\mathcal{J}'_0(\hat{x})(1+2\delta)}{\delta\mathcal{I}'(\hat{x})\mathcal{I}''(\hat{x})} \right\} dr. \quad (\text{S14}) \end{aligned}$$

S.2 Matching inner and outer solutions

For any field variable f (representing concentration and velocity), the matching condition at $\mathcal{O}(\epsilon^0)$ is

$$\lim_{y \rightarrow 0} (f_0^{(0)} + Def_1^{(0)} + \dots) = \lim_{\hat{y} \rightarrow \infty} (\hat{f}_0^{(0)} + De\hat{f}_1^{(0)} + \dots). \quad (\text{S15})$$

The matching condition for the concentration field (\hat{c}) yields: $\mathcal{I} = \lim_{y \rightarrow 0} c^{(0)}(x, y) + C_\infty$.

At $O(1)$, the matching condition for velocity yields:

$$u_0^{(0)} \Big|_{y=0} = \lim_{\hat{y} \rightarrow \infty} \hat{u}_0^{(0)}.$$

We substitute (S6a) in the above equation, which yields

$$u_0^{(0)} \Big|_{y=0} = - \left(\frac{\partial c}{\partial x} \Big|_{y=0} \right) \int_0^\infty \int_t^\infty (e^{-\hat{\psi}(s)} - 1) ds dt + \lim_{\hat{y} \rightarrow \infty} \mathcal{J}_0 \hat{y}. \quad (\text{S16})$$

For a bounded solution, $\mathcal{J}_0 = 0$. We thus obtain the solution reported previously in the literature Derjaguin *et al.* (1947); Anderson *et al.* (1982); Michelin & Lauga (2014). At $O(De)$, we use (S14) and obtain

$$u_1^{(0)} \Big|_{y=0} = -\delta \left(\frac{\partial c}{\partial x} \Big|_{y=0} \frac{\partial^2 c}{\partial x^2} \Big|_{y=0} \right) \int_0^\infty dp \int_p^\infty \mathcal{G}(r) dr + \lim_{\hat{y} \rightarrow \infty} \mathcal{J}_1 \hat{y}. \quad (\text{S17})$$

$$\begin{aligned} \text{Here, } \mathcal{G}(r) = \left\{ \left(\int_r^\infty \mathcal{F}(s) ds \right)^2 + 2\mathcal{F}(r) \left(\int_0^r s\mathcal{F}(s) ds + r \int_r^\infty \mathcal{F}(s) ds \right) \right. \\ \left. + \hat{\psi}'(r) e^{-\hat{\psi}(r)} \left(\int_0^r \left(r - \frac{s}{2} \right) s\mathcal{F}(s) ds + \frac{r^2}{2} \int_r^\infty \mathcal{F}(s) ds \right) \right\}. \quad (\text{S18}) \end{aligned}$$

Similar to the $O(1)$ solution, we obtain $\mathcal{J}_1 = 0$.

2.1 Intermediate matching

The above results for velocity can also be obtained using intermediate matching Hinch

(1991). In an arbitrary intermediate region ($y \sim \epsilon^\alpha$, where $0 < \alpha < 1$), the matching condition for both outer and inner region is

$$\lim_{\epsilon \rightarrow 0} (u_0^{(0)} + Deu_1^{(0)} + \dots) = \lim_{\epsilon \rightarrow 0} (\hat{u}_0^{(0)} + De\hat{u}_1^{(0)} + \dots). \quad (\text{S19})$$

Using the Taylor series in the LHS of (S19), at $O(1)$ the matching condition yields

$$\lim_{\epsilon \rightarrow 0} u_0^{(0)} \Big|_{y=0} + y \left(\frac{\partial u_0^{(0)}}{\partial y} \Big|_{y=0} \right) + \dots = \lim_{\epsilon \rightarrow 0} \hat{u}_0^{(0)}. \quad (\text{S20})$$

We substitute (S6a) in the RHS of the above equation and obtain

$$\lim_{\epsilon \rightarrow 0} \left[u_0^{(0)} \Big|_{y=0} + y \left(\frac{\partial u_0^{(0)}}{\partial y} \Big|_{y=0} \right) + \dots \right] = \lim_{\epsilon \rightarrow 0} \left[- \left(\frac{\partial c}{\partial x} \Big|_{y=0} \right) \int_0^{\hat{y}} \int_t^\infty \mathcal{F}(s) ds dt + \mathcal{J}_0 \hat{y} \right]. \quad (\text{S21})$$

Rescaling y and \hat{y} in terms of the intermediate coordinate (\bar{y}): $y = \bar{y} \epsilon^\alpha$ and $\hat{y} = \bar{y} \epsilon^{-\alpha}$.

$$\lim_{\epsilon \rightarrow 0} \left[u_0^{(0)} \Big|_{y=0} + \epsilon^\alpha \bar{y} \left(\frac{\partial u_0^{(0)}}{\partial y} \Big|_{y=0} \right) + \dots \right] = \lim_{\epsilon \rightarrow 0} \left[- \left(\frac{\partial c}{\partial x} \Big|_{y=0} \right) \int_0^{\bar{y} \epsilon^{-\alpha}} \int_t^\infty \mathcal{F}(s) ds dt + \mathcal{J}_0 \bar{y} \epsilon^{-\alpha} \right]. \quad (\text{S22})$$

Comparing the coefficients of \bar{y} , we obtain

$$\mathcal{J}_0 = \epsilon^{2\alpha} \left(\frac{\partial u_0^{(0)}}{\partial y} \Big|_{y=0} \right). \quad (\text{S23})$$

Since the velocity gradient at the surface ($y = 0$) is less than or equal to $O(1)$, \mathcal{J}_0 can be neglected at the leading order (as it is $O(\epsilon^{2\alpha})$, where $0 < \alpha < 1$). In the limit $\epsilon \rightarrow 0$, equation (S22) yields

$$\lim_{\epsilon \rightarrow 0} u_0^{(0)} \Big|_{y=0} = - \left(\frac{\partial c}{\partial x} \Big|_{y=0} \right) \int_0^\infty \int_t^\infty \mathcal{F}(s) ds dt. \quad (\text{S24})$$

S.3 Validation with literature

3.1 Concentration field and Newtonian slip velocity

The slip velocity and concentration field for different surface coverage is shown in fig. S1. For step change in activity, our results agree well with that of Michelin & Lauga (2014) (i.e. Newtonian fluid). To obtain these results, the Newtonian mobility coefficient M_0 is fixed to be -1 (as performed by Michelin & Lauga (2014)). It should be noted that M_0 in the main text is -1.1465 (corresponding to $\Phi_0 = -1$).

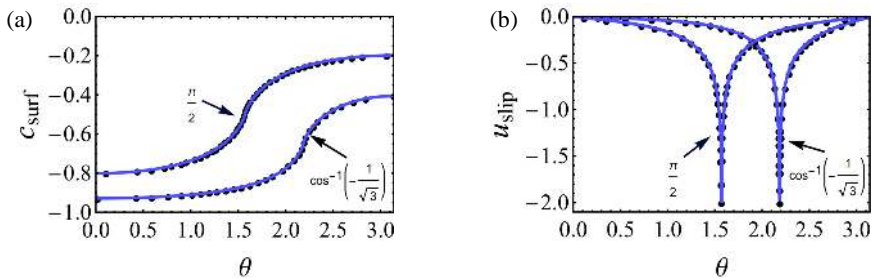


FIGURE S1. (a) Surface concentration profile and (b) slip velocity profile for $\cos \theta_c = 0$ and $-1/\sqrt{3}$. The filled circles represent the results of Michelin & Lauga (2014).

3.2 U_B for shear-thinning fluid

The velocity field around an axisymmetric squirmer was provided by Blake (1971), which was later used by Datt *et al.* (2017) to find the bulk non-Newtonian effects on the swimming of an axisymmetric Janus sphere. The radial component of the outer region disturbance field is

$$u_r = \alpha_1 \frac{P_1}{r^3} + \sum_{m=2}^{\infty} \left(\frac{1}{r^{m+2}} - \frac{1}{r^m} \right) \left(m + \frac{1}{2} \right) \alpha_n P_m, \quad (\text{S25})$$

and the tangential component is

$$u_\theta = \alpha_1 \frac{V_1}{2r^3} + \sum_{m=2}^{\infty} \left[\frac{m}{2r^{m+2}} - \left(\frac{n}{2} - 1 \right) \frac{1}{r^m} \right] \left(m + \frac{1}{2} \right) \alpha_m V_m. \quad (\text{S26})$$

Here, $V_m = [-2 \sin \theta / (m(m+1))] P_m^1(\cos \theta)$, P_m^1 is an associated Legendre polynomial of the first kind, $\alpha_m = m \mathcal{K}_m / (2m+1)$, and \mathcal{K}_m is the m^{th} spectral mode for the step activity which is given by (2.26) in the main text. We convert the above field in Cartesian coordinates and substitute it in the expression for U_B :

$$U_B = -\frac{1}{6\pi} \chi \int_{V_f} \mu_1(\gamma_0) \mathbf{A}_0 : \nabla \mathbf{u}^t dV. \quad (\text{S27})$$

Here $\mu_1 = (1 + Cu_B^2 |\gamma_0|^2)^{\frac{n-1}{2}} - 1$ and $|\gamma_0| = (\mathbf{A}_0 : \mathbf{A}_0 / 2)^{1/2}$. We use inbuilt Gauss-Kronrod rule in Mathematica 12 to numerically evaluate U_B . Fig.S2 shows an agreement with the results reported by Datt *et al.* (2017) for $\theta_c = \pi/2$.

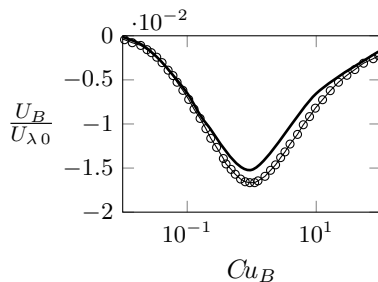


FIGURE S2. Comparison of numerical calculation of U_B (—) with Datt *et al.* (2017) (—○—) for $\chi = 0.1$, $\theta_c = \pi/2$, $n = 0.25$, $m = 15$.

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