Phoretic self-propulsion of Janus discs in the fast-reaction limit

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Abstract

Due to net interfacial consumption of solute, the two-dimensional problem of phoretic swimming is ill-posed in the standard description of diffusive transport, where the solute concentration satisfies Laplace's equation. It becomes well-posed when solute advection is accounted for. We consider here the case of weak advection, where solute transport is analyzed using matched asymptotic expansions in two separate asymptotic regions, a near-field region in the vicinity of the swimmer and a far-field region where solute advection enters the dominant balance. We carry out the analysis for a standard Janus configuration, where half of the particle boundary is active and the other half is inert. Our main focus lies in the limit of fast reaction, which leads to a mixed boundary-value problem in the near field. That problem is solved using conformal mapping techniques. Our asymptotic scheme furnishes the following implicit equation for the particle velocity s in the direction of the active portion of its boundary,

$$2s\left(8\ln\frac{8D}{|s|a}-\gamma\right)=\frac{bc_\infty}{a},$$

wherein a is the particle radius, D the solute diffusivity, c_∞ its far-field concentration, b the diffusio-osmotic slip coefficient, and γ the Euler–Mascheroni constant. The nonlinear dependence of s upon bc_∞ is a signature of the non-vanishing effect of solute advection.

I. INTRODUCTION

Self-propelled "swimmers" have been natural candidates for idealized two-dimensional investigations [1]: being force-free, no "Stokes paradox" arises when addressing the relevant limit of inertia-free flow. It is desirable [2] to employ similar two-dimensional models to analyze phoretic swimmers, which propel through a liquid solution by a chemical reaction on their boundary [3]. When considering phoretic swimmers, however, the net consumption of solute implies that the two-dimensional version of the standard continuum description is ill-posed. In that description, the (presumably diffusive) transport of solute is described by Laplace's equation; in two dimensions, this equation gives rise to a sink term that diverges logarithmically at large distances. Thus, while the flow problem does not introduce any conceptual difficulties, it is the underlying solute-transport problem that poses a non-trivial obstacle [4].

To circumvent this obstacle Crowdy [5] analyzed a two-dimensional Janus particle with two faces having different surface activities, absorbing and emitting solute in such a way that there is no net source production. While this situation is not generic, it turns out to be a theoretically important case study because the steady velocity of the particle in an unbounded solution can be found in closed form, as can the dynamical system governing its unsteady motion near a no-slip wall [5].

Naturally, it is of interest to study the general case where the particle is a net source of solute. Sondak *et al.* [4] noted that allowing for solute advection results in a well-posed problem even when there is a net production of solute. It follows that the closest two-dimensional well-posed analog of the three-dimensional diffusive transport is provided by the asymptotic limit of weak (but non-vanishing) advection. This singular limit was addressed by Yariv [6] using matched asymptotic expansions. Thus, the transport of solute was calculated in two different asymptotic regions: one on the scale of the particle, where solute is transported diffusively, and one on a remote scale, where advection enters the leading-order balance. A key difference between that asymptotic analysis and comparable classical analyses of transport phenomena [7] is that the velocity field is not externally imposed but is rather set by the interfacial solute gradients at the particle boundary. This results in a non-standard coupling (above and beyond that of asymptotic matching) between the solutions in the two asymptotic regions.

In Yariv's scheme [6], the solute concentration on the particle scale has been expanded using two-dimensional multipoles of Laplace's equation. The coefficients of this expansion are set by the appropriate model of interfacial solute production. Following Michelin & Lauga [8], Yariv [6] used first-order kinetics, where the relative magnitude of interfacial reaction is specified by the Damköhler number. The associated boundary condition then results in an indeterminate linear system governing the coefficients; asymptotic matching with the remote region eventually provides a determinate system, which may be solved for any value of the Damköhler number.

Following realistic applications, typical interest lies in the "canonical" Janus-particle configuration, where the particle boundary consists of two homogenous portions — one inert and one active. For this configuration, the methodology used by Yariv [6] is inappropriate in the fast reaction limit. In that limit, where the Damköhler number becomes large, the solute concentration satisfies a mixed boundary-value problem, governed by a Neumann condition on the inert portion of the boundary and a Dirichlet condition on the active portion. Such a mixed problem does not readily provide algebraic equations for the coefficients in a multipole expansion. This failure may be attributed to the singular nature of the fast reaction limit: indeed, mixed boundary-value problems are known [9] to exhibit square-root-type singularities at the points of transition between the different types of boundary conditions.

On the other hand, it turns out that the mixed boundary-value problem which emerges in that very limit may be naturally handled using conformal mapping techniques, similar to those which have been applied to analyze longitudinal flows about superhydrophobic surfaces [10]. The goal of the present paper is to revisit the two-dimensional autophoresis problem with a view towards the fast-reaction limit, which is known to be relevant to realistic experiments [11]. In contrast to the generic analysis of Yariv [6], which allows for arbitrary distributions of interfacial kinetics, we focus here upon the Janus configuration from the outset.

II. PHYSICAL PROBLEM

A chemically reactive circular particle (radius a) is freely suspended in an unbounded solution (solute diffusivity D). The reference solute concentration, at large distances from the particle, is denoted by c_∞. We assume a Janus configurations, where half of the particle boundary is chemically active while the other half is chemically inert. On the active portion, solute transfer is modeled using a first-order chemical reaction [8, 11],

$$\text{solute absorption (per unit area)} = k \times \text{local value of solute concentration}, \tag{2.1}$$

where the (presumably uniform) rate constant k is positive.

On the macroscale, the short-range interaction between the solute molecules and the particle is manifested by diffusio-osmotic slip [12],

$$\text{slip velocity} = b \times \text{surface gradient of solute concentration}. \tag{2.2}$$

Following the common practice [4, 8], we assume that b is uniform. Note that b is a signed quantity, positive for repulsive interactions and negative for attractive ones. The velocity scale associated with (2.2) is $\mathcal{U} = |b|c_\infty/a$. Defining the intrinsic Péclet number Pe as $a\mathcal{U}/D$ thus gives

$$\mathrm{Pe} = \frac{|b|c_\infty}{D}. \tag{2.3}$$

It follows from the problem symmetry that the force-free particle reacts by moving along its symmetry diameter with a constant velocity, say s (defined positive when the particle propagates in the direction of its active cap). By not rotating, the torque-free condition is trivially satisfied. Our goal is the determination of s.

III. DIMENSIONLESS FORMULATION

We employ a dimensionless notation where all length variables are normalized by a. The analysis is carried out in a particle-fixed reference system with origin at the particle center. In that system we use the (x, y) Cartesian coordinates, defined such that the x-axis is aligned along the symmetry diameter of the particle, pointing in the direction of the active cap. We additionally utilize the (r, θ) polar coordinates, with θ measured in the counterclockwise direction from the x-axis. In what follows we consider the coupled transport–flow problem governing the solute concentration c, normalized by c_∞, and fluid velocity $\mathbf{u}$, normalized by $\mathcal{U}$. Our interest is in the velocity $U(= s/\mathcal{U})$ of the particle relative to the otherwise quiescent liquid; in the particle-fixed reference frame this velocity is manifested as the uniform streaming $-U\hat{\boldsymbol{\imath}}$ at infinity, $\hat{\boldsymbol{\imath}}$ being a unit vector in the x-direction.

The dimensionless solute transport problem is governed by: (i) the advection–diffusion equation,

$$\nabla^2 c = \mathrm{Pe}\, \mathbf{u} \cdot \boldsymbol{\nabla} c \quad \text{for} \quad r > 1; \tag{3.1}$$

(ii) the kinetic condition at the particle boundary,

$$\frac{\partial c}{\partial r} = \begin{cases} \mathrm{Da}\, c, & 0 < |\theta| < \pi/2 \\ 0, & \pi/2 < |\theta| < \pi \end{cases} \quad \text{at} \quad r = 1, \tag{3.2}$$

where

$$\mathrm{Da} = \frac{ak}{D} \tag{3.3}$$

is the Damköhler number, representing the ratio of diffusive (a^2/D) to reactive (a/k) time scales; and (iii) the approach to the reference concentration at large distances,

$$\lim_{r \to \infty} c = 1. \tag{3.4}$$

The flow is governed by: (i) the continuity and Stokes equations [the former tacitly employed in (3.1)]; (ii) diffusio-osmotic slip [cf. (2.2)]

$$\mathbf{u} = \hat{\mathbf{e}}_\theta M \frac{\partial c}{\partial \theta} \quad \text{at} \quad r = 1, \tag{3.5}$$

where $M = b/|b|$; (iii) far-field approach to a uniform stream,

$$\lim_{r \to \infty} \mathbf{u} = -U \hat{\boldsymbol{\imath}}; \tag{3.6}$$

and (iv) the requirement that the particle is force-free. The latter, in conjunction with (3.5)–(3.6), provides the particle velocity as a quadrature [13],

$$U = \frac{M}{2\pi} \int_{-\pi}^{\pi} \left. \frac{\partial c}{\partial \theta} \right|_{r=1} \sin\theta \, d\theta. \tag{3.7}$$

The coupled flow–transport problem is described in Fig. 1. In principle, this nonlinear problem provides U as a function of Pe, Da and $M(= \pm 1)$. While (3.7) may appear to suggest that there is no need to solve for the flow, this is not the case due to the advective term in (3.1).

Relying upon the three-dimensional problem in the absence of advection, one may naívely assume that

$$c \to c, \; \mathbf{u} \to -\mathbf{u} \quad \text{under the transformation} \quad M \to -M, \tag{3.8}$$

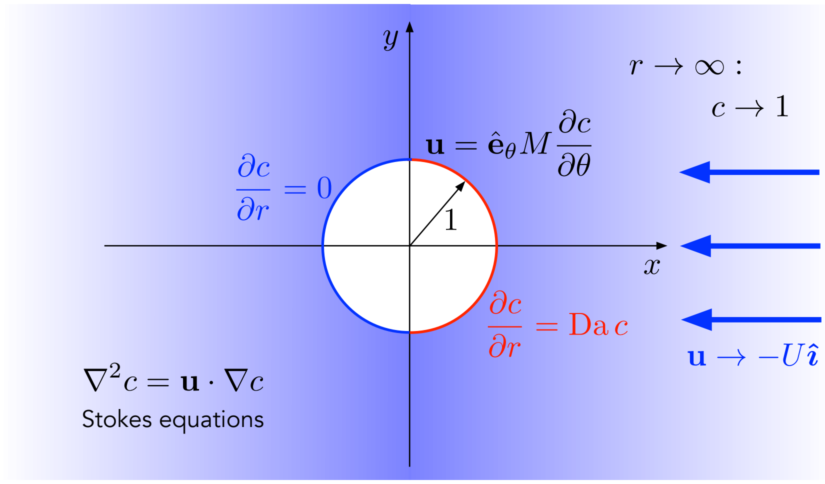

FIG. 1. The nonlinearly coupled boundary-value problem.

and, in particular,

$$U \to -U \quad \text{under the transformation} \quad M \to -M. \tag{3.9}$$

However, symmetry (3.8) is incompatible with (3.1) for all positive Pe. In general, the nonlinear dependence upon the flow carries out a non-trivial dependence upon M.

Regardless of the validity of (3.9), it is plausible that the sign of U coincides with that of M. Indeed, with condition (3.2) implying larger fluxes on the "right" face of the particle ($|\theta| < \pi/2$), and with the concentration being fixed at infinity by (3.4), it is anticipated that c is smaller on that face. The gradients of c are then expected to point to the "left" face of the particle, where c is larger. For positive M this is also the direction of slip [see (3.5)], so a force-free particle reacts by moving to to the right. [This is also evident from (3.7).] This suggests that:

$$U = M|U|. \tag{3.10}$$

IV. SMALL PÉCLET NUMBERS

For small Péclet numbers, the advection–diffusion equation (3.1) degenerates at leading order to Laplace's equation:

$$\nabla^2 c = 0 \quad \text{for} \quad r > 1, \tag{4.1}$$

whereby coupling with the flow may seem to have disappeared. However, condition (3.2) implies that

$$\oint_{r=1} \frac{\partial c}{\partial r}\, d\theta > 0 \quad (= F, \text{ say}), \tag{4.2}$$

for all positive Da values. The associated net flux into the particle necessitates the asymptotic behavior

$$c \sim \frac{F}{2\pi} \ln r \quad \text{for} \quad r \gg 1, \tag{4.3}$$

which is incompatible with the decay condition (3.4). The limit of small Péclet numbers is a singular one.

It is of course well known [14] that small-Péclet-number problems are generically singular, becoming nonuniform at large distances. The non-uniformity in the present problem may be traced back to (4.3), which implies that $\boldsymbol{\nabla} c$ decays as $1/r$ at large r. With that decay rate we find that the left- and right-hand sides of (3.1) are $O(r^{-2})$ and $O(\mathrm{Pe}\, r^{-1})$, respectively; regardless of how small is Pe, advection always enters the leading-order balance at $r = O(\mathrm{Pe}^{-1})$. Laplace's equation (4.1) thus constitutes a leading-order approximation only on the particle-scale region, where $r = O(1)$. It needs to be supplemented by an additional "remote" expansion, valid at $r = O(\mathrm{Pe}^{-1})$. With that approach, the logarithmic divergence in (4.3) is acceptable, as it is the remote expansion that needs to satisfy (3.4).

It is important to note that the flow is coupled to the solute concentration only through the slip condition (3.5). The flow problem is accordingly "unaware" of the scale separation in the solute-transport problem, and remains formulated on a single length scale. It follows that expression (3.7) for the particle velocity remains intact.

Since interest ultimately lies in that velocity, it may appear that it is sufficient to solve the particle-region transport, and then make use of (3.7). In the general case, however, such an independent analysis cannot be realized. Indeed, consider the refinement of the asymptotic behavior (4.3),

$$c \sim \frac{F}{2\pi} \ln r + G + O(r^{-1}) \quad \text{for} \quad r \gg 1, \tag{4.4}$$

where G represents the "background" solute concentration, relative to that at infinity, and the $O(r^{-1})$ error represents decaying harmonics. Since condition (3.4) cannot be utilized in the particle-region analysis, it does not aid in determining F and G. It is then evident that condition (3.2) does not suffice to determine both F and G. Without asymptotic matching with the remote region, the particle-scale problem is indeterminate.

It is therefore necessary to analyze both the particle region and the remote region. The procedure we adopt is threefold: first, we solve the remote-region transport; second, we exploit that solution in conjunction with (4.4) to obtain a relation governing F, G and U from the requirement of asymptotic matching between the two regions; third, we solve the particle-region transport, treating F as given. The resulting expressions for G and U (as functions of F), in conjunction with the above relation, eventually furnish the particle velocity.

V. REMOTE-REGION ANALYSIS

The remote region is naturally analyzed using the stretched Cartesian coordinates (x', y'), defined by

$$(x', y') = \mathrm{Pe}\,(x, y). \tag{5.1}$$

Similarly, we define $r' = \mathrm{Pe}\,r$. In the remote region we additionally employ the fields c' and $\mathbf{u}'$, functions of (x', y'), which are defined as

$$c' = c - 1, \quad \mathbf{u}' = \mathbf{u}. \tag{5.2}$$

We therefore obtain at leading order from (3.1)

$$\nabla'^2 c' = \mathbf{u}' \cdot \boldsymbol{\nabla}' c', \tag{5.3}$$

where $\boldsymbol{\nabla}' = \hat{\boldsymbol{\imath}}\,\partial/\partial x' + \hat{\boldsymbol{\jmath}}\,\partial/\partial y'$ is the stretched gradient operator. This equation is subject to large-r' decay, which follows from (3.4).

Given the approach (3.6) to a uniform streaming velocity, it is evident that $\mathbf{u}' \equiv -\hat{\boldsymbol{\imath}} U$ at leading order. Equation (5.3) is therefore simplified to $\nabla'^2 c' = -U \partial c'/\partial x'$. Substituting $c' = e^{-\frac{1}{2}Ux'} H$ we find that H satisfies the modified Helmholtz equation, $\nabla'^2 H = \frac{1}{4} U^2 H$. The solution of that equation that decays at infinity and is least singular at the origin is a radially symmetric screened source of magnitude D, $(D/2\pi) K_0(|U| r'/2)$, in which K_0 is the modified Bessel function of the second kind. We therefore obtain

$$c' = \frac{D}{2\pi} e^{-\frac{1}{2}Ux'} K_0\left(\frac{|U| r'}{2}\right). \tag{5.4}$$

Using the small-argument behavior of K_0 [15] we find, with an algebraically small error,

$$c' \sim -\frac{D}{2\pi}\left(\ln \frac{|U| r'}{4} + \gamma\right) \quad \text{as} \quad r' \to 0, \tag{5.5}$$

wherein γ is the Euler–Mascheroni constant. Asymptotic matching with (4.4) gives

$$F = -D, \quad G = 1 + \frac{D}{2\pi}\left(\ln\frac{4}{|U|\mathrm{Pe}} - \gamma\right), \tag{5.6}$$

where, following the conventional approach [16], logarithmic terms are considered on par with $O(1)$ terms as $\mathrm{Pe} \to 0$. Combining (5.6) to eliminate D we obtain

$$G = 1 - \frac{F}{2\pi}\left(\ln\frac{4}{|U|\mathrm{Pe}} - \gamma\right) \tag{5.7}$$

which provides the requisite extra condition for uniquely determining the particle-scale solution. While the equations governing c are linear, condition (5.7) is nonlinear, representing the non-vanishing signature of solute advection. Since Pe is small, it follows from (5.7) that the signs of F and G are opposite; with F being positive [see (4.2)], the background solute concentration G is negative, as could have been expected for an absorption process.

We now claim that at leading order, the symmetry (3.9) does hold. The proof consists of three elements: (i) the particle-scale calculation of c (and in particular the relation it imposes between F and G) is unaffected by the flow and hence by the sign of M; (ii) The calculation of U via (3.7) is compatible with (3.9); and (iii) the closure condition (5.7), which serves to uniquely determine the inner problem, is also compatible with (3.9).

VI. THE PARTICLE-SCALE PROBLEM IN THE FAST REACTION LIMIT

The particle-scale solute-transport problem is governed by Laplace's equation (4.1) and the kinetic condition (3.2). The far-field condition (3.4) does not apply on that scale. Instead, we impose the asymptotic condition (4.3), treating $F > 0$ as given. This results in a well-posed problem (and excludes the trivial solution $c \equiv 0$)

Our interest lies in the limit $\mathrm{Da} \to \infty$, where condition (3.2) is degenerated to the following mixed Dirichlet–Neumann condition at $r = 1$:

$$c = 0 \quad \text{for} \quad 0 < |\theta| < \pi/2, \tag{6.1a}$$

$$\frac{\partial c}{\partial r} = 0 \quad \text{for} \quad \pi/2 < |\theta| < \pi. \tag{6.1b}$$

The problem governing c on the particle scale, associated with that condition, is described in Fig. 2. This problem depends only upon (the yet unknown) flux F. In solving the particle-scale problem, our goal is to obtain — in terms of F — both the constant G appearing in (4.4) and the particle velocity U, as given by the quadrature (3.7).

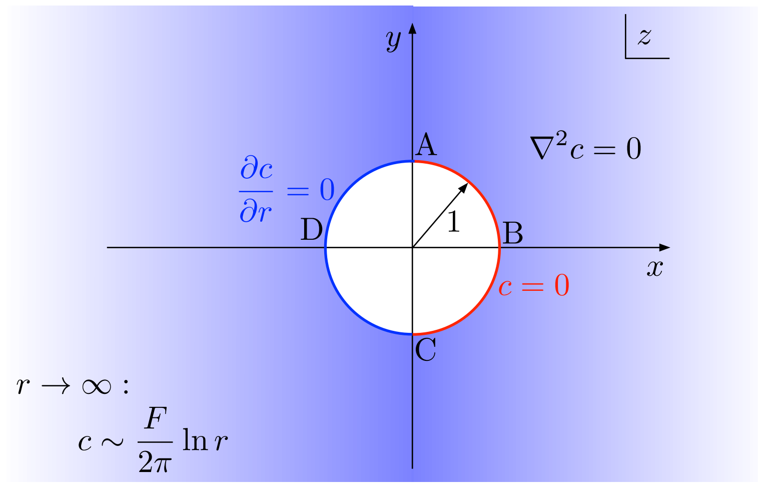

FIG. 2. The particle-scale transport problem in the fast-reaction limit.

Denoting the harmonic conjugate of c by ψ, we embed the concentration c in the complex potential Φ,

$$\Phi = c + i\psi, \tag{6.2}$$

an analytic function of $z = x + iy$. Making use of the Cauchy–Riemann conditions we find that, in terms of Φ, the mixed condition (6.1) reads (refer to Fig. 2)

$$\mathrm{Re}\{\Phi\} = 0 \text{ on ADC}, \quad \mathrm{Im}\{\Phi\} = 0 \text{ on ABC}. \tag{6.3}$$

Also, in terms of Φ, the far-field asymptote (4.4) becomes

$$\Phi(z) \sim \frac{F}{2\pi} \log z + G + O(|z|^{-1}) \quad \text{for} \quad |z| \gg 1. \tag{6.4}$$

Following Crowdy [10, 17, 18], we employ two conformal mappings (see Fig. 3) in terms of the parametric variable ζ. The first is

$$f(\zeta) = \frac{(\zeta - \bar{\alpha})(\zeta - 1/\bar{\alpha})}{(\zeta - \alpha)(\zeta - 1/\alpha)}, \tag{6.5}$$

where the point α is on the imaginary axis in the upper-half unit disc in the complex ζ-plane. This mapping transplants this semi-disc to the exterior of the unit circle with $\zeta = \alpha$ being mapped to infinity. Choosing

$$\alpha = i \tan(\pi/8), \tag{6.6}$$

which is derived by insisting that $f(1) = i$, the real diameter ABC $(-1 < \zeta < 1)$ is mapped onto the right side of the unit circle while the upper-half unit circle CDA $(|\zeta| = 1, \mathrm{Im}\{\zeta\} > 0)$ is mapped onto the respective left side.

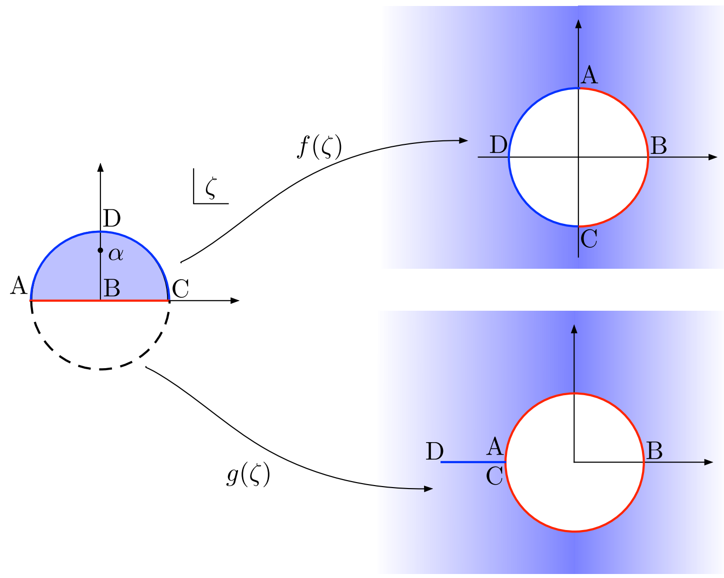

FIG. 3. The conformal map $f(\zeta)$ transplants the upper unit ζ disc to the unbounded region exterior to a unit disc. The conformal map $g(\zeta)$ transplants the upper unit ζ disc to the unbounded region exterior to a unit disc with a slit.

The second is the radial-slit mapping [10, 17, 18],

$$g(\zeta) = \frac{(\zeta - \bar{\alpha})(\zeta - 1/\alpha)}{(\zeta - \alpha)(\zeta - 1/\bar{\alpha})}, \tag{6.7}$$

which also sends the upper-half unit disc to the exterior of the unit circle. Now, however, the real diameter ABC is mapped onto the entire unit circle. The upper-half unit circle CDA is now mapped onto a finite slit which extends between -1 (points A and C) and $-(\sqrt{2}+1)/(\sqrt{2}-1)$ (point D).

We claim that the required solution for $\Phi(z)$ can be written down immediately in the parametric form

$$z = f(\zeta), \quad \Phi(z) = h(\zeta), \tag{6.8}$$

where

$$h(\zeta) = \frac{F}{2\pi} \log g(\zeta). \tag{6.9}$$

To prove the above claim we note, using the geometrical properties of the radial slit mapping $g(\zeta)$, that the real and imaginary parts of

$$h(\zeta) = \frac{F}{2\pi} \left\{ \ln |g(\zeta)| + i \arg g(\zeta) \right\} \tag{6.10}$$

respectively vanish on the unit circle ABC and the slit CDA in the transformed plane. Since homogeneous Dirichlet conditions are conformally invariant [19], it follows that conditions (6.3) are satisfied. Moreover, by considering f and g near the point α (where $z \to \infty$) we find that $g(\zeta) \sim 2z$. It then follows that

$$\Phi(z) \sim \frac{F}{2\pi} \log(2z), \tag{6.11}$$

consistent with the far-field requirement (4.3). Comparing with (4.4) we obtain

$$G = \frac{F}{2\pi} \ln 2. \tag{6.12}$$

It is possible to invert the mapping $z = f(\zeta)$ to find ζ, and hence $\Phi(z)$, explicitly as a function of z. The resulting expressions [17] make evident the presence of square root singularities at $z = \pm i$. To extend the solution form (6.8)–(6.9) to more general Janus particles with different coverage ratios requires merely altering the choice (6.6), as done in other studies [17].

Consider now the quadrature (3.7). Given condition (6.1a) on the right face of the particle, it now reads

$$U = \frac{M}{2\pi} \int_{\text{ADC}} \frac{\partial c}{\partial \theta} \sin\theta \, d\theta. \tag{6.13}$$

Using the Cauchy–Riemann conditions it is readily verified that

$$z\Phi'(z) = \frac{\partial c}{\partial r} - i\frac{\partial c}{\partial \theta}. \tag{6.14}$$

Since $\partial c/\partial r = 0$ at the left face ADC, we have there $\partial c/\partial \theta = iz\Phi'(z)$. Moreover, on the unit circle, where $z = e^{i\theta}$, $\sin\theta \, d\theta = -\operatorname{Re}\{dz\}$. We conclude that

$$U = \frac{M}{2\pi} \operatorname{Im} \int_{\text{ADC}} z\Phi'(z) \, dz. \tag{6.15}$$

Changing the integration variable to ζ using (6.8) we therefore obtain

$$U = \frac{M}{2\pi} \operatorname{Im} \int_{\text{ADC}} f(\zeta) h'(\zeta) \, d\zeta. \tag{6.16}$$

It is a simple matter to confirm directly from (6.5), (6.7) and (6.9) that

$$f(1/\zeta) = f(\zeta), \quad h(1/\zeta) = -h(\zeta), \tag{6.17}$$

allowing to express U as an integral over the entire unit circle in the ζ-plane:

$$U = \frac{M}{4\pi} \operatorname{Im} \int_{|\zeta|=1} f(\zeta) h'(\zeta) \, d\zeta, \tag{6.18}$$

where the integration is carried out in the clockwise direction. Substitution of (6.5), (6.7) and (6.9) reveals that the integrand is a rational function of ζ with a single (second-order) pole inside the unit circle, at $\zeta = \alpha$, with residue $-F/2\pi$. The residue theorem therefore gives

$$U = \frac{MF}{4\pi}. \tag{6.19}$$

VII. COMBINING THE RESULTS

Having derived relations (6.12) and (6.19), we have all that we need from the particle-scale analysis. Plugging these into the closure condition (5.7) and further assuming that (3.10) indeed holds, we eventually obtain the equation

$$2MU\left(\ln\frac{8}{MU\mathrm{Pe}} - \gamma\right) = 1, \tag{7.1}$$

which implictly provides MU as a function of Pe. Since it results in a positive MU for $\mathrm{Pe} \ll 1$, we have justified (3.10) *a posteriori*.

Note that the product MU constitutes the ratio of the dimensional velocity s to the signed velocity scale bc_∞/a, while the product $MU\mathrm{Pe}$ is equal to the Péclet number $|s|a/D$ associated with particle motion (as opposed to the intrinsic number Pe) . The dimensional counterpart of (7.1) is

$$2s\left(8\ln\frac{8D}{|s|a} - \gamma\right) = \frac{bc_\infty}{a}. \tag{7.2}$$

This equation illustrates how the inverse scaling with particle size, pertinent in the three-dimensional version of the fast-reaction limit [11], breaks down in two dimensions.

VIII. CONCLUDING REMARKS

The present contribution, which is based upon a combination of singular perturbation analysis with conformal mapping techniques, complements the original two-dimensional analysis of Yariv [6], which is inadequate to handle the mixed boundary-value problem that emerges in the fast reaction limit.

We briefly comment upon the non-dimensionalization process. In his two-dimensional analysis, Yariv [6] followed Michelin & Lauga [8] in choosing the solute-concentration scale as one that estimates the perturbation from the reference concentration at infinity, rather than that very concentration. Unsurprisingly, then, the resulting velocity scale (Da$\mathcal{U}$ in the present notation) properly represents the small-Da limit, where the deviation from the reference concentration is indeed a small perturbation. While that choice is also adequate to analyze moderate Da numbers, it results in non-representative concentration and velocity scales in the analysis of the large-Da limit. As a consequence, it has the unfortunate artifact of a dimensionless particle velocity that diminishes as Da $\to \infty$. That is indeed evident from Fig. 1 of Yariv [6].

In the present analysis, where we use the reference concentration and the associated velocity scale $\mathcal{U}$, the problem formulation differs from that of Yariv [6]. In the large-Da limit, the dimensionless particle velocity now attains a finite limit. The calculation of that velocity has been the ultimate goal of the present analysis.

It is worth emphasizing that the present analysis made use of two limits, namely small Pe and large Da. These two limits differ fundamentally, as the first is singular and the second is regular; given this difference, there is no restriction upon the relative values of Pe and 1/Da. In particular, note that the discussion in Sec. IV and the subsequent remote-region analysis in Sec. V are valid for all values of Da.

A natural followup of the present work is the analysis of the comparable three-dimensional problem. As that problem is clearly well posed for zero Péclet numbers, no need arises for the incorporation of weak advection. The major challenge in that followup is the mixed boundary-value problem that arises at large Damköhler numbers. The particle speed attained in that limit was obtained by Ebbens *et al.* [11] by extrapolating from speed values obtained numerically at finite values of Da. To the best of our knowledge, the mixed boundary-value problem appropriate to the limit Da $\to \infty$ has never been solved directly.

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