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Diffusion-limited association of nanoparticles in fluid: Beyond the no-slip boundary conditions

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ABSTRACT

In the treatments of diffusion-limited association of suspended nanoparticles (NPs), their diffusion coefficients are usually considered to be constant or depend on the interparticle distance as determined by fluid dynamics with the no-slip boundary condition. In the latter case, due to the corresponding slowdown of diffusion at short distances, the association rate constant is smaller than that calculated by ignoring this slowdown and using the diffusion coefficients corresponding to single NPs. The no-slip boundary condition can, however, be violated, and now there is evidence that it may happen more often than one could expect. In such situations, the partial-slip boundary condition is more suitable. Employing the latter boundary condition, I derive herein general integral expressions for the rate constant of association of spherically shaped NPs without and with the NP-NP interaction. Simple analytical results have been obtained in various situations.

1. Introduction

Association of molecules or NPs suspended in liquid is a very basic process occurring in different areas of physics, chemistry, and biology. Often, the rate of this process is controlled by reactant diffusion. This control may take place both globally on the macroscopic length scale of the concentration gradients and locally on the length scale comparable with the size of reactants. The latter determines the association rate constant, k_a. The seminal theoretical studies were done in this field by Smoluchowski [1] in relation with colloids. According to him and later important works by Collins and Kimball [2] and Berg [3] (for more recent literature, see e.g. articles [4,5] and references therein), this rate constant can be safely introduced and used except a short initial phase of the process. In their calculations, NPs were considered to be spherical (this case is discussed below), the coefficients of diffusion of NPs were assumed to be constant, and the outcome was

$$k_a = 4\pi\mathcal{R}\mathcal{D}, \tag{1}$$

where $\mathcal{R}$ is the contact radius or, more specifically, the sum of the NP radii, and $\mathcal{D}$ is the sum of the NP diffusion coefficients. Since its derivation by Smoluchowski [1], Eq. (1) was widely employed in different contexts. Nowadays, with rapid expanding of nanoscience and biophysics with focus on biological macromolecules and nanoparticls circulating in liquid, e.g., blood and lymphatic vessels *in vivo* (reviewed, e.g., in [6–8]), the role of Eq. (1) and its extensions only increases.

In numerous applications of Eq. (1), the NP diffusion coefficient, D, is usually identified with that predicted by fluid dynamics for single particles far from interfaces, i.e.,

$$D_\circ = k_B T/6\pi\eta R, \tag{2}$$

where R is the NP radius, and η is the viscosity coefficient. This widely employed expression for $D_\circ$ is derived by using the no-slip boundary condition near the NP-solution interface. This boundary condition is needed in order to calculate the mobility of a NP, which is then converted into the diffusion coefficient by employing the detailed balance principle.

Eq. (1) does not take the particle-particle interaction into account. If D is constant, this interaction can be included into the model as described in [9].

In reality, according to fluid dynamics, the diffusion of NPs near a solution-solid interface and/or during their contacts is slowed down. In particular, the coefficients of diffusion perpendicular and parallel to the solution-solid interface become to be not equal and both are smaller than that given by (2). Customarily, this effect is described by using the no-slip boundary condition near all the interfaces. For diffusion of spherically shaped NPs perpendicular to a flat interface, this approach

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yields [10–12]

$$D = \frac{6h^2 + 2hR}{6h^2 + 9hR + 2R^2} D_\circ, \tag{3}$$

where h is the minimal distance between the particle surface and the solid surface. [Note that Eq. (3) is mathematically identical to Eq. (41) in review [12], although the first impression might be that they are different. The apparent difference is due to the difference in the variables used.]

According to (3), the coefficient of NP diffusion perpendicular to an interface decreases with decreasing h and vanishes at $h \to 0$. The coefficient of diffusion parallel to an interface decreases with decreasing h as well but does not vanishes at $h \to 0$ [10–12]. In general, both these features are important for calculations of the rate constants of diffusion-limited processes. For example, I may refer to attachment of NPs to membrane-immobilized receptors [13]. In the context of association of spherically shaped NPs [14] or some of the kinetic processes occurring at or near interfaces [15–19], the diffusion parallel to an interface is in fact irrelevant, and its specifics is here not discussed.

To illustrate the effect of slowdown of diffusion [as in (3)] on the rate of diffusion-limited association of suspended NPs, $A + B \to AB$, of radii R and ρ, it is instructive to focus on the case with $R \gg \rho$ [14]. In this limit, the diffusion coefficient of A is much smaller than that of B, and the location of A can be considered to be fixed. The B diffusion coefficient, D, appreciably deviates from its asymptotic value [as in (2)] only when A is located near B. In this region, the surface of A can be considered to be flat, and accordingly to describe the dependence of D on the distance, r, between the centers of A and B one can just replace in Eq. (3) R by ρ and h by $r - \mathcal{R}$, where $\mathcal{R} = R + \rho$, i.e.,

$$D = \frac{6(r-\mathcal{R})^2 + 2(r-\mathcal{R})\rho}{6(r-\mathcal{R})^2 + 9(r-\mathcal{R})\rho + 2\rho^2} D_\circ \tag{4}$$

with $D_\circ = k_B T/6\pi\eta\rho$ [as in (2)]. To calculate k_a analytically, Eq. (4) can be simplified as [14]

$$D(r) = \begin{cases} (r-\mathcal{R})\lambda^{-1} D_\circ & \text{at} \quad \mathcal{R} \le r \le \mathcal{R} + \lambda, \\ D_\circ & \text{at} \quad r \ge \mathcal{R} + \lambda, \end{cases} \tag{5}$$

where λ is the parameter comparable with ρ. At $r \to \mathcal{R}$, Eq. (4) obviously yields $\lambda = \rho$. With this value of λ, Eq. (5) is accurate, however, in a very narrow range of r values near $r = \mathcal{R}$. With $\lambda = 2\rho$ (or e.g 4ρ), the range of the applicability of Eq. (5) is somewhat wider [this can be easily shown by comparing graphically (4) and (5)]. Thus, depending on goals, one can use $\lambda = \rho$, 2ρ, or 4ρ. Below, I use $\lambda = 2\rho$. In addition, one should take into account that just near the interface or, more specifically, at $r - \mathcal{R} \le \delta$ [δ is the properly defined length scale ($\delta \ll \rho$)], there are deviations from the conventional fluid dynamics (e.g., due to roughness of the interfaces or presence of ligand and receptors). With this specification and in the absence of the particle-particle interaction, the analysis yields [14]

$$k_a = \frac{4\pi D_\circ \mathcal{R}}{1 + (\lambda/\mathcal{R}) ln(\lambda/\delta)}. \tag{6}$$

This expression shows that although, as expected, the slowdown of diffusion near the interfaces results in reduction of the association rate constant, this effect is usually not dramatic because the corresponding correction is logarithmic.

2. Beyond the no-slip boundary condition

As already noticed, the derivation of Eqs. (1)-(6) is based on the no-slip boundary condition or, more specifically, on the corresponding expression (3) for the coefficient of diffusion near an interface (for the other examples of the use of this expression, see Refs. [15–19]). In fact, this boundary condition implies two conditions concerning the normal and tangential velocities. Both of them should vanish. During the past two decades, there were experimental and theoretical attempts to scrutinize experimentally whether the no-slip boundary condition really holds (reviewed in [12]; for some additional references, see e.g. [20]). The outcome is that it can be violated more often than one could expect. For example, it may happen even in the biologically relevant systems (including, e.g., lipid bilayers [21]) with hydrophilic surfaces. In such cases, according to the classical Navier-Maxwell phenomenology (e.g., [22]) and experiments (briefly reviewed in [20]), the condition for the velocity along the interface should be reformulated in terms of partial slip as

$$v(0) = b \left. \frac{\partial v}{\partial z} \right|_{z=0}, \tag{7}$$

where z is the normal coordinate ($z = 0$ corresponds to the flat interface), $v(0)$ is the velocity at the interface, $b > 0$ is the length corresponding to extrapolation of the velocity, $v(z)$, to the region with $z < 0$ down to $v = 0$ (Fig. 1).

Formally, condition (7) means that the liquid velocity is finite at the interface. This velocity is, however, used on both sides of condition (7), and accordingly it can be arbitrary small. Physically, as already noticed, the situation just near the interface is different compared to that at larger distances. Thus, in fact, condition (7) corresponds not literally to $z = 0$ but rather to $z \simeq \delta$, where δ ($\delta \ll \rho$) is the length scale characterizing here a tiny layer just near the interface where the conventional phenomenology becomes inapplicable (this aspect is discussed, e.g., in Ref. [23]). Physically, this δ is not fully identical to that used in (6), but their scales are comparable. For the microscopic models aimed at the interpretation of b, one can read recent articles [24–27]. The generalizations of condition (7) proposed in [28,29] are identical to condition (7) in the limit of low velocities or shear rate (this limit is relevant in the context of diffusion). There is also generalization of condition (7) for two-phase solutions (see [30] and references therein). The latter subject is beyond my goals.

Concerning the applicability of condition (7) and corresponding expressions for the diffusion coefficient (see below), I can articulate that the underlying phenomenology does not take explicitly into account the effect of the charges located at the surface of NPs and those associated with the molecules forming solution. In reality, these charges are often essential in the context of the NP-NP interaction which can appreciably influence the association rate (Section 3). In principle, these charges can induce the convective motion of the molecules forming solution (in the region between NPs) and it in turn can influence diffusion of NPs. Such electrohydrodynamic effects are far from straightforward as discussed in the literature (see, e.g., recent articles [31–33] and references therein).

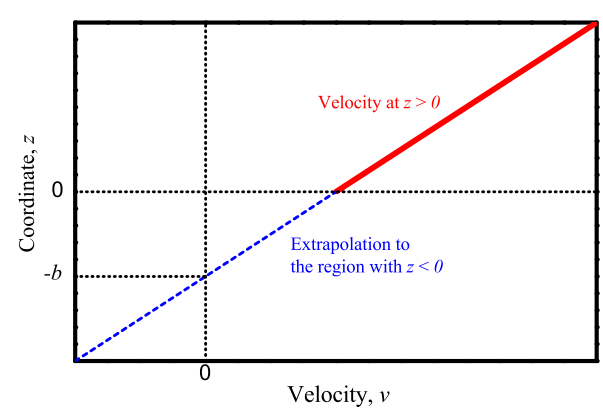

Fig. 1. Velocity of liquid (solid line) along the liquid-solid interface as a function of the coordinate normal to the interface (adopted from [20]). The dashed line represents extrapolation of the velocity to the region below the interface so that $v = 0$ at $z = -b$ (b is positive or zero).

Usually, the focus is there on periodic oscillatory convection [31,32]. In our context, diffusion of NPs is slow and the electrohydrodynamic effects are not expected to be central (this is obviously the case e.g. provided the ionic mobility is low). In the numerous theoretical studies of the kinetic processes occurring with participation of NPs, the NP-NP interaction is often taken into account but the electrohydrodynamic effects are neglected (the latter is the case since the seminal theoretical studies by Smoluchowski). With these reservations, I neglect the electrohydrodynamic effects as well. In fact, the analysis I present below is of interest even in the situations when the electrohydrodynamic effects are important because it can be used as the starting point for extensions and/or modifications.

For single NPs with the partial-slip boundary condition at the NP-solution interface, the mobility and diffusion coefficient can be calculated analytically (Sec. 4.20 in [22] and [20]), and the effect of this condition is reduced to the reduction of R or ρ by b in the expressions for $D_\circ$ provided $b \ll R$ and $b \ll \rho$, respectively (see Eq. (18) in [20]). This result as well as condition (7) itself (Fig. 1) indicate that the related corrections can formally be obtained by (i) assuming that the region with $-b \le z \le 0$ is filled by liquid and (ii) shifting the no-slip boundary condition to $z = -b$. For the tangential velocity, this prescription directly follows from (7). For the normal velocity, this is a reasonable approximation supported by the analysis in [20,22].

Focusing on the situations with $\delta \ll b$ and $b \le \lambda = 2\rho \ll R$ (these conditions are expected to hold e.g. in biologically related systems where b is not too small and simultaneously not too large [20,21]), I will consider that the partial-slip boundary condition takes place only at the surface of the larger NP and will adopt the prescription above in order to calculate the diffusion of the smaller NP. In this approximation, it can be done by neglecting δ and replacing $\mathcal{R}$ by $\mathcal{R} - b$ in (4), i.e.,

$$D = \frac{6(r-\mathcal{R}+b)^2 + 2(r-\mathcal{R}+b)\rho}{6(r-\mathcal{R}+b)^2 + 9(r-\mathcal{R}+b)\rho + 2\rho^2} D_\circ. \tag{8}$$

By analogy with (5), this expression can be replaced by

$$D(r) = \begin{cases} (r-\mathcal{R}+b)\lambda^{-1} D_\circ & \text{at} \quad \mathcal{R} \le r \le \mathcal{R} - b + \lambda, \\ D_\circ & \text{at} \quad r \ge \mathcal{R} - b + \lambda. \end{cases} \tag{9}$$

According to the latter expression, the scale of D at r close to $\mathcal{R}$ is $\simeq (b/\lambda) D_\circ \equiv (b/2\rho) D_\circ$ provided b is not too small. For comparison, it is instructive to mention one of the expressions obtained in [34] (Eq. (37b); see also Eq. (44) in review [12]) for the coefficient of diffusion of a spherical NP far the wall in the perpendicular direction,

$$D(h) = \left[1 - \frac{9\rho}{8(h+\rho)}\left(1 - \frac{b}{h+\rho}\right)\right] D_\circ, \tag{10}$$

where h is the minimal distance between the particle surface [as in (3)]. This expression can be employed for a rough estimate of $D(h)$ in the case of interest when $h + \rho$ is close to ρ and b is not too small. This yields $9\rho/[8(h+\rho)] \simeq 1$, $b/(h+\rho) \simeq b/\rho$, and accordingly $D \simeq (b/\rho) D_\circ$. The scale of D given by this estimate is comparable to that obtained above. This can be considered as an additional indication that the analysis is reasonable.

With the specification of the diffusion coefficient above [Eq. (9)] and in the absence of the particle-particle interaction, k_a can be calculated by using the steps similar to those employed in [14] in order to derive (6). This yields

$$k_a = \frac{4\pi D_\circ \mathcal{R}}{1 + (\lambda/\mathcal{R}) ln(\lambda/b)}. \tag{11}$$

This expression is valid provided $\ln(\lambda/b) > 0$, and accordingly k_a can be represented as

$$k_a = 4\pi D_\circ \mathcal{R} \varphi, \tag{12}$$

where φ is the correction factor defined by

$$\varphi = \begin{cases} 1/[1 + (2\rho/\mathcal{R}) ln(2\rho/b)] & \text{at} \quad \delta \ll b \le 2\rho, \\ 1 & \text{at} \quad b \ge 2\rho. \end{cases} \tag{13}$$

Expression (11) has been derived provided $\delta \ll b$, and accordingly it predicts that k_a is larger than that determined by (6). Another condition of its applicability is $b < \lambda = 2\rho$, and in this case it predicts that k_a is smaller than the conventional association rate constant, $4\pi D_\circ \mathcal{R}$ [cf. (1)]. For $b = \lambda = 2\rho$, expression (11) is reduced to the conventional one. If $b > \lambda = 2\rho$, the conventional expression becomes valid, because the corrections are negligible. In addition, the ratio $\lambda/\mathcal{R}$ ($\equiv 2\rho/\mathcal{R}$) was considered to be small ($2\rho/\mathcal{R} \ll 1$). In this limit, the correction factor defined by (13) reduces k_a only slightly as shown in Fig. 2 for $2\rho/\mathcal{R} = 0.1$. If the condition $2\rho/\mathcal{R} \ll 1$ does not hold, the correction factor (13) can be used for rough estimates, and the effect of the partial-slip boundary condition on k_a can be appreciable.

3. Inclusion of the particle-particle interaction

The association of NPs can be influenced by the NP-NP interaction, $U(r)$. This interaction, described customarily by the DLVO-type models, includes the double-layer part and the attractive long-range van der Waals and repulsive short-range hydration parts (see, e.g., [35,36]). In this section, k_a is calculated taking $U(r)$ and the no-slip or partial-slip boundary condition for diffusion into account.

In the presence of $U(r)$, the diffusion flux of B particles per A particle is given by

$$J = -4\pi r^2 D(r)\left(\frac{\partial c(r,t)}{\partial r} + \frac{c(r,t)}{k_B T}\frac{\partial U(r)}{\partial r}\right), \tag{14}$$

where $c(r, t)$ is the concentration of B particles. To get k_a, J can be calculated in the steady-state approximation (as in the sections above). It can be done by expressing $c(r)$ via another function, $\chi(r)$, as

$$c(r) = \chi(r) exp[-U(r)/k_B T]. \tag{15}$$

Substituting (15) into (14) and using the steady-state approximation yields

$$r^2 D(r) exp[-U(r)/k_B T]\frac{d\chi(r)}{dr} = const. \tag{16}$$

This equation is the first-order equation whose solution should at first sight be uniquely determined by a single boundary condition, but it contains *const*, which is arbitrary before fixing boundary conditions. For

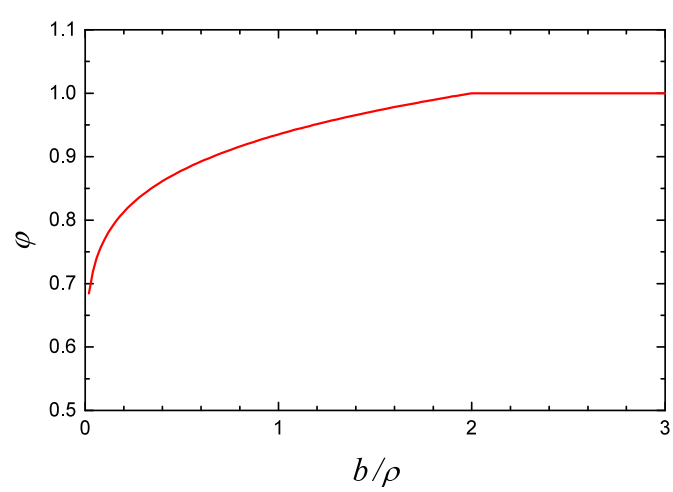

Fig. 2. Correction factor for the association rate constant [Eq. (13)] as a function of the ration of b and ρ for $2\rho/\mathcal{R} = 0.1$.

this reason, this equation can be solved by using two relevant boundary conditions, and this procedure allows to fix *const*. In the case under consideration, the suitable boundary conditions are $\chi(\mathcal{R}+\delta)=0$ and $\chi(r)=c_\circ$ at $r\to\infty$, and with these conditions Eq. (16) can be easily integrated so that

$$c(r)=c_\circ[1-F(r)/F(\mathcal{R}+\delta)]exp[-U(r)/k_BT], \tag{17}$$

where

$$F(r)=\int_r^\infty \frac{exp[U(z)/k_BT]}{z^2D(z)}dz. \tag{18}$$

Using this expression for $c(r)$, the association rate constant can then as usual be obtained by dividing the diffusion flux at $r=\mathcal{R}$ by $c_\circ$,

$$k_a=\frac{4\pi}{F(\mathcal{R}+\delta)}\equiv 4\pi/\int_{\mathcal{R}+\delta}^\infty \frac{exp[U(r)/k_BT]}{r^2D(r)}dr. \tag{19}$$

If D is constant, $D(r)=D_\circ$ [as in (2)], and δ is negligible (so that one can set $\delta=0$), Eq. (19) is reduced to that derived in [9],

$$k_a=4\pi D_\circ/\int_{\mathcal{R}}^\infty \frac{exp[U(r)/k_BT]}{r^2}dr. \tag{20}$$

With the dependence of D and U on r, the integration in (19) should be done numerically. Analytical estimates of k_a are possible in one of the practically interesting cases when the repulsive part of $U(r)$ at $r\simeq\mathcal{R}$ is appreciable ($\gg k_BT$) so that its contribution to the integral in (19) dominates. In this case, Eq. (19) can be represented as

$$k_a\simeq\frac{4\pi\mathcal{R}^2}{\gamma}\widetilde{D}exp[-U(\mathcal{R}+\delta)/k_BT], \tag{21}$$

where γ is the length scale (a few Å; comparable with or somewhat smaller than δ) characterizing the drop of $U(r)$ at $r\simeq\mathcal{R}$, and $\widetilde{D}$ is the scale of D in this region.

In the no-slip case (as in Section 1), Eq. (5) yields $\widetilde{D}\simeq\delta D_\circ/\lambda\simeq\delta D_\circ/2\rho$. Substituting this expression into (21) results in

$$k_a\simeq\frac{2\pi\mathcal{R}^2\delta}{\gamma\rho}D_\circ exp[-U(\mathcal{R}+\delta)/k_BT]. \tag{22}$$

In the partial-slip case (as in Section 2), Eq. (5) yields $\widetilde{D}\simeq bD_\circ/\lambda\simeq bD_\circ/2\rho$ (provided $b>\delta$ as it can be expected). With this expression, Eq. (21) results in

$$k_a\simeq\frac{2\pi\mathcal{R}^2b}{\gamma\rho}D_\circ exp[-U(\mathcal{R}+\delta)/k_BT]. \tag{23}$$

4. Conclusion

Finally, I notice that the full-scale analysis of mutual diffusion of NPs with partial-slip boundary condition and calculation of the corresponding association rate constant, k_a, are far from simple and still lacking. I have presented the first results concerning this rate constant. To simplify the analysis, the focus was on the situations when the size of spherically shaped NPs of one type is appreciably larger than that of NPs of the other type. The calculations have been performed without and with the NP-NP interaction.

The general conclusion drawn from the calculations is that with the partial-slip boundary condition k_a becomes larger than that calculated by taking the slowdown of diffusion into account and closer to that calculated by ignoring the slowdown of diffusion. Qualitatively, this is what one could expect. Quantitatively, in the limit under consideration, the effect of the boundary conditions on k_a is relatively weak provided the NP-NP interaction is negligible. The corresponding corrections are logarithmic [Eqs. (6) and (11)] irrespective of the type of the boundary conditions. With the NP-NP interaction, the effect of the boundary conditions on k_a can be more appreciable [Eqs. (22) and (23)]. In particular, k_a can be proportional to the slip length b [Eq. (23)] in the partial-slip case.

Concerning the comparison of the obtained results with experiments, I can notice that in the context under consideration the accurate measurements of k_a for NPs is still challenging. One of the problems here is that the first what one should quantify is the NP size. The conventional measurements of this size are based on the DLS and NTA methods which in turn imply the no-slip boundary condition, i.e., these methods in their standard realization are inconsistent with the partial-slip boundary conditions [20]. Then, one is expected to quantify b. This is challenging as well [21]. Another problem is that k_a is often influenced by the NP-NP interaction (Section 3). In this case, k_a depends not only on b but also on the length, γ, characterizing the NP-NP interaction. The experimental quantification of the latter length is also challenging. For these reasons, the comparison with experiments is now hardly possible. The current progress in nanoscience is, however, rapid, and I expect that the subject under consideration will attract more attention on both sides (experiment and theory) in the nearest future.

Author statement

The whole work was done by the author.

Declaration of Competing Interest

The author declares that he has no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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