Formation of self-propelling clusters starting from randomly dispersed Brownian particles

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Abstract. We present a simple chemical strategy for the formation of a self-propelling cluster via the process of capture and assembly of passive colloids on the surface of a chemically active colloid. The two species of colloids that are isotropic and Brownian otherwise interact to form propelling clusters. With the help of coarse-grained numerical simulations, we show that a chemically active colloid can induce diffusiophoretic motility to nearby chemically inert colloids towards itself. This propulsion and then self-assembly can then lead to the formation of active clusters. We observe the formation of propelling dimers, trimers, tetramers, etc. depending on the chemical activity and size of the colloids.

Keywords. Diffusiophoresis; colloids; self-propulsion; assembly; active cluster.

1. Introduction

Both biological and non-biological organisms have been reported to respond to chemical gradients around them, by moving either up or down the gradient. In biological frameworks, bacteria have been found to migrate towards the glucose-rich regions in search of their food and form active assemblies in this process [1]. Sperm cells are also well known to collectively swim towards chemo-attractants [2]. On the contrary, in the realm of synthetic nano/micro world, colloidal particles have been designed to behave like artificial micro-swimmers [3–5]. These micro-swimmers have enormous potential to understand many elementary questions in active matter and self-organization [4,6,7] and can also be employed as carriers to pick-up and deliver cargo [4,8–11].

In the view of having a variety of applications of such assembled structures in real and synthetic life, they have attracted remarkable attention in the past decade [1,4,6,12]. These studies have led to the creation of various functional materials as per the demand of present technologies. Dynamic self-assembly in a mixture of eccentric active particles and passive particles was shown to form a large dense, dynamic cluster [13]. Similarly, a colloidal suspension of magnetic particles was demonstrated to have dynamic assemblies depending on the applied magnetic field frequencies [14]. Recent experimental work on a suspension of light-absorbing and non-absorbing immotile microspheres has shown to form an assembly of active colloidal molecules in a highly controlled manner using light [15]. In the context of chemically active colloidal particles, self-assembly of a pair of colloids leading to spontaneous motor formation was also revealed in recent studies [4,16]. Similarly, chemically modified Janus has been illustrated to form on-the-fly assemblies that have potential application towards optimal cargo transport and delivery [8,17,18]. During the process of self-assembly, the role played by an individual entity is important. In this work, our interest lies in the process where a non-equilibrium interaction between the colloids leads to assemblies that can spontaneously acquire directional motility. The significant challenges in this regard are to fine-tune the factors that influence the assembly and motion. Some of the factors which ultimately decide the structure and then the motion of the assembled structure are the motility of participating entities [19] which can be influenced by the generated chemical gradient [4,20], thermal gradient [21], externally applied fields [12,14], etc. Also, the hydrodynamic interactions (HI) present in the system are crucial to determine the same [22–24].

Here, we study the dynamics of self-assembly followed by self-propulsion in a system having two species of colloids. Both of these isotropic colloids are diffusive individually, but upon interaction, they acquire motility. Out of these two species, one is a chemically active colloid that forms a uniform radial chemical gradient when immersed in a chemical fuel. The other class of colloid that we name as an inert or passive colloid senses chemical gradient produced by the active sphere and exhibits self-propulsion towards it, similar to a living bacterium move towards chemo-attractants. Such a combination of chemically active and passive spheres has been studied in the past in the context of propelling dimers [16,25,26]. With the help of mesoscopic coarse-grained simulation methods, we show that inert colloids in the neighbourhood of a chemically active colloid get attracted towards it through diffusiophoresis. Hence, get captured on the surface of the active colloid thus making an assembly.
Another interesting observation is the reconfiguration of the assembly, i.e., the spontaneous rearrangement of inert spheres on the surface of the active particle such that the cluster then starts to swim. The active assembly of the colloids in the form of a dimer, trimer, tetramer, etc. is observed in our study depending on the radius of the active and inert colloids. The propulsion speed, capture time and assembly time have been quantified here, and the factors affecting the performance of the motors have been identified.

The article is organized as follows. In section 2, we have described the simulation model and the system of active and passive colloids in the chemically active medium. Section 3 discusses the capture dynamics of inert particles on the surface of active colloids and also explains the reason for self-assembly and propulsion. In this section, we start the discussion with a pair of colloids and then move to multiple inert particle systems. Finally, section 4 concludes the study.

2. Diffusiophoretic theory and simulation model

2.1 Simulation

We employ a particle-based hybrid simulation model where the particle motion is governed by the molecular dynamics (MD) coupled with multi-particle collision dynamics (MPCD) to implement hydrodynamics and thermal effects. This is a well-known method and has been used previously in different contexts [16,25,26]. Initially, a reactive colloid (R) with radius \( R \) and an inert colloid (I) with radius \( R_i \) are immersed in a solvent with A-type point particles. The reactive colloid is a chemically active species and converts the fuel fluid particle A to the product particle B by an irreversible chemical reaction \( A + R \rightarrow B + R \) with an intrinsic rate. On the contrary, the chemically inert colloids do not possess this property. The chemical reaction on colloid hence generates a radially outward gradient of product molecules, which can be sensed by any inert colloid in its vicinity. These inert colloids hence exhibit the diffusiophoretic motion [27] towards \( R \) upon experiencing the chemical gradient.

The colloidal spheres interact with the fluid particles through the repulsive Lennard–Jones (LJ) potential given as

\[
V_{ij}(r) = 4\epsilon\left[(\sigma/r)^{12} - (\sigma/r)^{6}\right] + \epsilon \quad \text{for} \quad r \leq r_c \quad \text{and} \quad V_{ij}(r) = 0 \quad \text{for} \quad r > r_c.
\]

Here \( \sigma \) and \( \epsilon \) are the characteristic distance and energy parameters. To model the diffusiophoretic motion to the colloids, we choose the interaction energy parameter of A and B as \( \epsilon_A = \epsilon_B = \epsilon \) for R spheres whereas for I spheres we choose \( \epsilon_A > \epsilon_B = \epsilon \). In addition to this, excluded volume interaction has been employed between the two colloids. The combination of the chemical gradient produced by R around I (see figure 1) along with the above stated difference in interaction energies of fuel and product with I leads to the propulsion of I spheres towards the R sphere. To keep the system out-of-equilibrium, which is required for continuous propulsion of I, the product B particles are converted back to A at a distance far from spheres.

We use a coarse-grained mesoscopic dynamical model for fluid particles, popularly known as MPCD [28,29]. In this scheme, \( N_s \) point-like particles with mass \( m_s \) can move in a cubic box of length \( L \). The method comprised of two alternative steps: streaming and collision. With the help of forces based on the potentials used in the system, the fluid particles are evolved by Newton’s equation of motion in the streaming step. However, in the collision step, all the fluid particles are sorted into small cubic cells of size \( a_0 \) such that the mean free path \( \lambda \ll a_0 \). The multi-particle collision (MPC) is performed in each cell by a random rotation matrix \( \mathcal{R} \) that results in the rotation of the relative velocities of all the fluid particles. The velocity of fluid-particle \( i \) after each collision step \( t_c \) in cell \( \chi \) is given by

\[
V_i(t_c + t_s) = V_i(t) + \mathcal{R}(t_s) [V_i(t) - V_i(t_c)],
\]

where \( V_i \) is the velocity of the centre-of-mass of all fluid particles in cell \( \chi \). A random grid shift in each direction is applied to ensure the Galilean invariance for the system [30]. The described method here conserves mass, momentum and energy.

2.1a Simulation parameters: Throughout the paper, the parameters \( a_0, \epsilon \) and \( m_s \) are described in dimensionless units for length, energy and mass, respectively. We use the cubic simulation box of dimension \( L = 50 \). The temperature of the system is fixed at \( k_B T = 1.0 \). Unit cell length \( a_0 \) is taken for performing the MPCs. In each MPC cell, the rotation of velocity is carried out by an angle of \( \alpha = 120^\circ \) about a random axis at every collision time step \( t_c = 0.1 \). Average fluid number density in each MPC cell is \( \rho_i = 10 \), and the mass of fluid-particle is taken to be \( m_i = 1.0 \). Energy parameters in LJ potential \( \epsilon_A = 1.0 \), whereas \( \epsilon_B \) is varied among 0.01, 0.1 and 0.5 to achieve different propulsion rates of I colloids.
To study the effect of size, we have taken two different sizes of the reactive colloid $R_t = 2.0$ and $R_t = 4.0$, whereas, the size of the inert colloid is kept constant at $R_i = 2.0$. The masses of colloidal particles are adjusted according to the density matching with the surrounding fluid. MD time step is taken to be $\Delta t = 0.01$.

### 2.2 Diffusiophoretic theory for the inert colloid

The inert colloid when comes in the vicinity of R colloid can experience the gradient of B-type particles around its surface and respond to it by moving towards the R colloid following the diffusiophoresis mechanism [27]. The different interaction of A and B-type particles with the surface of inert colloid makes it move due to the fluid slip velocity $v_s$ generated around the boundary layer [31]:

$$ v_s = -\frac{k_B T}{\eta} (\nabla C_B) \Lambda, \quad (1) $$

where $\eta$ is the viscosity of the medium, $C_B$ is the concentration of product B particles on the outer edge of the boundary layer and $\Lambda$ determines the net strength of the interaction between the fluid particles and the surface of the colloid. The velocity of the inert colloid can be calculated by averaging the slip velocity over the entire surface of the colloidal particle [16,32]:

$$ V = -\langle v_s \rangle_{\text{surface}}. \quad (2) $$

To determine the concentration field of products $C_B$, we solve the diffusion equation, $D \nabla^2 C_B(r) = 0$ by using the radiation boundary condition $\hat{k}_0 C_A(R_t, t) = \hat{k}_D R_t \hat{\nabla} C_A(R_i, t)$. To maintain the steady state, there should be a continuous supply of fuel molecules at the boundaries. For this, we assume $C_A(r \to \infty) = C_0$ and $C_A + C_B = C_0$. Here, $D$ is the diffusion coefficient of the solvent particles, $k_D = 4\pi R_i D$ is the Smoluchowski rate constant and $k_0$ is the intrinsic reaction rate constant. By solving the diffusion equation, the estimated concentration field of product particles can be given as:

$$ C_B(r) = \frac{k_0 C_0}{k_0 + k_D} \frac{R_i}{r}. \quad (3) $$

At any time instant $t$, let $R(t)$ be the vector distance from I to R. By averaging the gradient of product particles over the outer edge of the sphere and projecting its velocity along the direction of $R(t)$, we obtain the velocity of the inert colloid as:

$$ V(t) = \frac{2k_B T C_0 \Lambda}{3\eta} \frac{k_0}{k_0 + k_D} \frac{R_t}{R(t)^2} \equiv \frac{\lambda}{R(t)^2}, \quad (4) $$

where

$$ \lambda = \int_0^\infty dr r (e^{-\beta V_B(r)} - e^{-\beta V_A(r)}). $$

Further, the time evolution of separation between the colloidal particles can be obtained by integrating equation (4) and the time taken by inert colloids to reach the encounter distance $R_t = R_t + R_i$ is given by:

$$ \langle t_{\text{cap}} \rangle = \frac{L_d^3 - R_t^3}{3\lambda}. \quad (5) $$

### 3. Results and discussion

The interaction between a pair of freely moving R and I colloids having different sizes has been investigated earlier both experimentally and theoretically [4,16,33]. Here, our interest is to explore what happens to the dynamics of R colloids in the presence of multiple I spheres. To be precise, we want to probe the capture and assembly dynamics of I spheres on R, which then leads to symmetry breaking and hence converts a diffusive sphere to a chemical motor. To begin with, let us re-examine the results from the case of R--I pair.

#### 3.1 Capture dynamics of a single inert colloid

When the chemical reaction on R sphere is switched on, a chemical gradient is produced around I, which then starts moving towards R by diffusiophoresis and eventually gets captured. In the entire process of capture, R sphere mostly exhibit diffusive motion, which then transforms to a weak drift towards I when they are very close to each other. A stable dimer is formed once the I and R spheres come very close to each other, and the assembly then exhibits a directional motion over a long distance and time similar to a motor. The motion of this bound pair is similar to the sphere-dimer motors [4]. Figure 2 shows the snapshots from simulation for capture dynamics and directional motion of dimers.

Chemotaxis and capture of inert colloids to the reactive target site is quite robust and seems to take place for various sizes of colloids. Of course, when the size of I becomes too small, thermal fluctuations disrupt the directed motion of I spheres. On the contrary, the radius of R spheres plays an important role.
role in both capture and propulsion. The concentration field of B species around I which is a key factor in determining the velocity and capture time, is influenced by \( R_t \). Therefore, we probe the effect of the size of \( R \) on the capture dynamics. The velocity of I spheres \( V_I \) is plotted in figure 3 as a function of \( L_d \), which is the separation distance between the centres of spheres with \( R_t = 2.0 \) and \( R_t = 4.0 \), respectively. As the distance between the spheres decreases, the concentration gradient of B around I increases leading to the increased velocity. Furthermore, the propulsion velocity \( V_I \) is substantially more for larger \( R_t \), which is in line with equation (4). Then the capture time also reduces accordingly. For all further studies, since the focus is on moving I, we choose \( R_t = 4.0 \).

### 3.2 Capture dynamics for a pair of inert colloids

Self-assembly of the active matter giving rise to complex architectures such as living clusters due to symmetry breaking has been widely observed \([15,34–37]\). The phoretic interaction followed by short-range van der Waals and critical Casimir attractions are argued to be responsible for such clustering in the case of the active particles \([15,34]\). To understand such self-assembly, here we probe the dynamics of two inert colloids \( I_1 \) and \( I_2 \) with radius \( R_t = 2.0 \) in the presence of a reactive colloid \( R \) with radius \( R_s = 4.0 \).

Figure 4a shows the schematic representation of the initial configuration of the system where both the inert spheres are situated at the same distance from \( R \) at \( t = 0 \). As discussed in the previous section, \( I_1 \) and \( I_2 \) spheres will initially exhibit phoretic motion towards the reactive colloid, due to the concentration gradient of product, B species around them as shown in figure 4b. In this section, we are interested in the motion of inert colloids after getting captured to the surface of \( R \).

Before starting the discussion on what happens after capture, we discuss the capture dynamics for the two inert colloid systems. A comparison of the velocity of the inert colloids at various \( L_d \) is shown in table 1 as it approaches the reactive target sphere for the initial configuration given in figure 4. It is clear that both the inert spheres move with a similar speed as they approach the target. This is because both of them experience the same diffusiophoretic pull by the \( R \) sphere, by virtue of being at a similar distance from \( R \).

Further, to check the effect of the initial configuration of the system on the capture dynamics of inert spheres, we investigated three different initial arrangements. A representation of the initial set-up is shown in figure 5, where \( C_1 \), \( C_2 \) and \( C_3 \) have a colloidal arrangement such that the vectors joining I and \( R \) are at an angle of 180, 90 and 30°, respectively. As the reactive sphere \( R \) sets up a radial gradient of product B, we expect a similar velocity and hence the capture time of inert colloids as initially they are at the same radial distance from \( R \). A comparison of the average directed velocity of inert colloids in these three configurations is shown in figure 6 and concludes that the directed velocity of the inert spheres for the capture to happen depends only on the radial distance between the target and walker.

| \( L_d \) | \( V_{I_1} \)  | \( V_{I_2} \)  |
|---------|-------------|-------------|
| 8.0     | 0.047 ± 0.007 | 0.04 ± 0.006 |
| 10.0    | 0.036 ± 0.010 | 0.032 ± 0.004 |
| 15.0    | 0.018 ± 0.005 | 0.029 ± 0.009 |
| 20.0    | 0.01 ± 0.002  | 0.014 ± 0.003 |

Figure 3. Propulsion velocity \( (V_I) \) of inert colloids varying with initial separation \( L_d \) for two sets of radii of reactive colloids, \( R_t = 4.0 \) (purple plot) and \( R_t = 2.0 \) (blue plot) and the average capture time \( (\tau_{cap}) \) taken by inert colloids is shown in the inset.

Figure 4. (a) Schematic diagram representing the initial configuration of the system with two inert spheres \( (I_1 \text{ and } I_2) \) and a reactive sphere \( R \). At \( t = 0 \), both the inert spheres are situated at the same distance from \( R \). (b) The colour plot for the concentration of B-type solvent particles.
3.3 Spontaneous self-assembly by symmetry breaking

Once the colloids get captured to the reactive target sphere, we expect them to make a diffusive cluster if the capture of $I_s$ is head-on. This is because the head-on capture will lead to a symmetric configuration, which cannot exhibit any directed motion. However, we observe a dramatic configurational adjustment leading to breaking of symmetry, which then induces directed propulsion to the cluster. During this adjustment, both the inert spheres self-assemble on one side of the reactive sphere, to obtain a configuration similar to the Janus dimer as in section 3.2. Figure 7 shows the process of capture and self-assembly of inert spheres on R. It is important to note that the self-assembly of $I_s$ on R which then leads to spontaneous motor formation is independent of initial configuration and $C_2$ and $C_3$ also exhibit a similar dynamics.

Another noteworthy observation, in this case, is about the speed of centre-of-mass ($V_{cm}$) of the propelling aggregate. We compared the $V_{cm}$ of the cluster for single $I$ and a pair of $I$ case and reported in table 2. It is evident that in the presence of more $I$ the speed of cluster increases as the force-generating entities have increased.

The obvious question that arises from the above observation is, why do the inert particles cluster on one side of the reactive sphere, and how does it chooses the direction to form such an assembly? To answer these queries, we calculate the time average force experienced by the inert spheres due to the solvent as a function of azimuthal angle (defined in figure 4a) between configurations shown in figure 7b and c, i.e., time span after the capture and before the assembly. For a particular ensemble shown in figure 7, the force calculation depicts higher force on the right sides of $I$ spheres; therefore, they must move towards the left hemisphere of R and indeed we observe them moving in the same direction (figure 8).

Of course, the next question to ask would be what generates such force asymmetry on inert spheres. The diffusiophoretic model tells us that any chemical gradient is sufficient to provide the force asymmetry. Therefore, we calculate the concentration of B-type fluid particles around the inert colloids. The normalized concentration of B fluid particles ($C_B/C_0$) as a function of $\theta$, where $\theta$ is defined in figure 4a, is plotted in figure 9. This clearly depicts the presence of chemical gradient around the inert particles, which then leads to force asymmetry and hence self-assembly of $I$ colloids on R.

Figure 9 (inset) shows the normalized concentration of B fluid particles ($C_B/C_0$) as a function of $\theta$ when the $I_s$ are not captured on R, i.e., they are sufficiently far from the target. Hence, when $I_s$ are sufficiently away from R, there is no bias on them to choose the direction of the assembly. The choice of the direction of the assembly shown by inert colloids after

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**Figure 5.** Initial configurations when two $I$ colloids approach the $R$ colloid at (a) $180^\circ$ ($C_1$), (b) $90^\circ$ ($C_2$) and (c) $30^\circ$ ($C_3$).

**Figure 6.** Average velocity ($V_I$) of $I$ colloids for different initial distances ($L_d$) from the reactive colloid for different initial configurations in the main plot and the average capture time ($\tau_{cap}$) is shown in the inset.

**Figure 7.** Configurations depicting trimer formation which is ready to move is shown in the top panel. The motion of trimer motor with representative trajectories is shown in the bottom panel, for the radii of reactive and inert colloids as $R_r = 4.0$ and $R_i = 2.0$, respectively.

**Table 2.** Speed of motor shows increment when we deal with two inert colloidal systems in the presence of one reactive colloid.

| $V_{I1\, cm}$ | $0.0390 \pm 0.0001$ |
| $V_{I2\, cm}$ | $0.0451 \pm 0.0001$ |
Figure 8. Force exerted by the fluid particles on the inert colloids during the self-assembly process as a function of azimuthal angle $\theta$. Here, $I_1$ assembles towards the left hemisphere of $R$.

Figure 9. Concentration of B fluid particles around $I_1$ and $I_2$, respectively when they have started assembling towards the left hemisphere of the $R$ colloid whereas the inset shows the concentration of B when $I_1$ and $I_2$ are far from $R$ at $t = 0$.

Figure 10. Average time taken by I colloids to self-assemble after their capture at $R$ which varies with $\epsilon_B$.

Figure 11. Formation of a pentamer with four I and one R systems which move at a constant speed. Here $R_1 = 2.0$ and $R_2 = 4.0$.

3.4 Factors affecting the self-assembly

Self-assembly of inert spheres on $R$ is influenced by factors such as the interaction of solvent with $I$, HI in the system, the interaction strength, etc. We probe some of them below, and the quantification is done on the basis of time taken by I spheres to self-assemble on $R(\tau_{assm})$ after getting captured on its surface.

For self-assembly point of view, we believe that the interaction of solvent with $I$ is a critical parameter. The ratio of $\epsilon_B/\epsilon_A$ decides the asymmetric force experienced by $I$, which in turn decides its motility. For $\epsilon_B/\epsilon_A = 1.0$ the inert particles are purely diffusive and do not show any propulsion and assembly. Therefore, to probe the effect of solvent interaction on self-assembly, we plot $\tau_{assm}$ vs. $\epsilon_B/\epsilon_A$ in figure 10. For very small values of $\epsilon_B/\epsilon_A$ i.e., for more asymmetry in forces, self-assembly is more rapid leading to smaller $\tau_{assm}$ supporting the fact that local fluid interaction with $I$ is one of the deciding factors for self-assembly on the target sphere.

The HI involved during the capture and self-assembly process has its unique role [22,23]. To confirm the role of the hydrodynamic effect further, we calculate $\tau_{assm}$ for the 2I system by switching off HI. The advantage of MPCD simulation lies in the fact that it is possible to switch off the HI effect whenever needed. The standard method to suppress HI in MPC simulations is to randomize the velocities of the solvents by re-sampling the velocities from the Maxwell–Boltzmann distribution at every collision step. In this case, we do observe a similar capture and assembly process; however, the speed of the capture and self-assembly slows down to a very large extent.

3.5 Assembly of multiple inert colloids leading to a large motile cluster

The work by Schmidt et al [15] on active self-assembly induced by light in a suspension colloid has demonstrated the dynamic formation of complex architectures such...
as living clusters. Therefore, it would definitely be interesting to ask whether such dynamic self-assembly into a plethora of complex structures is possible in a chemical system such as ours. To answer this question, we add multiple inert colloids to ask whether such dynamic self-assembly into a plethora of active colloids and its interaction with the fluid. The velocity of inert colloids, depends on its initial distance from the assembly. The capture time, as well as the propulsion of reactive one through diffusiophoresis. The assembly then converts to a self-propelled cluster. Further, the capture and conversion on a self-propelled cluster. An example of such a case is shown in figure 11 where a system comprises four I, and one R shows the formation of an assembled structure (figure 11b) which then can self-propel as a dynamical motor (figure 11c and d).

Therefore, the above observed chemically powered self-assembly leading to the formation of the chemical motor is quite robust and provides an easy route to obtain large chemical motors from a dispersion of spherically symmetric colloids.

4. Conclusions

In conclusion, we have presented a simple strategy for the formation of self-propelling chemical motors via the process of self-assembly. In this work, we studied a chemical system which comprises a chemically reactive colloid that converts the fuel to the product, which is then sensed by inert colloids in its vicinity. Two species of isotropic colloids that cannot self-propel by themselves interact to form a self-assembled cluster that can swim. Here, we identified and discussed the factors that are responsible for such self-assembly and self-propulsion. The process of assembly and propulsion is highly scalable, and we showed the formation of dimers to pentamers in our study.

We showed that the chemical field around the reactive colloid causes the binding of inert spheres on the surface of reactive one through diffusiophoresis. The assembly then forms a cluster which by virtue of symmetry breaking gets converted to a self-propelled cluster. Further, the capture and assembly process of passive colloids is independent of the initial configuration. The capture time, as well as the propulsion velocity of inert colloids, depends on its initial distance from active colloids and its interaction with the fluid.

The aggregation of colloidal particles is an outcome of dynamic self-organization, leading to the formation of a motile motor is an interesting study. Such self-motile motors are involved in many biological processes also; for example, the self-assembly process of kinesin motor proteins has an implication in the formation of the mitotic spindle [38].

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