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Symmetrical Catalytically Active Colloids Collectively Induce Convective Flow

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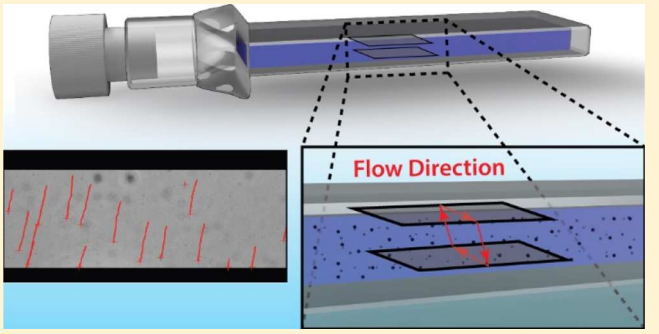
Supporting Information

ABSTRACT: Although much attention has focused on self-motile asymmetrical catalytically active "Janus" colloids as a route to enable new fluidic transport applications, the motion of symmetrical catalytically active colloids is less investigated. This is despite isotopically active colloids being more accessible and commonly used as supports for heterogeneous catalysis. Here, we addressed this by systematically investigating the motion of platinum-coated colloids capable of isotropically decomposing hydrogen peroxide. We observed the onset of collective convective flow as the colloidal volume fraction increased above a threshold. The ballistic velocities induced by the collective flow were quantified by particle tracking and were found to increase with the volume fraction. We also determined the associated increase in the Péclet number as an evidence of the potential to use convection as a simple method to enhance mass transport rates. By determining the persistence lengths, we were able to correlate the magnitude of convective flow with the overall catalytic activity per unit volume. This suggests that the mechanism for the collective flow is driven by chemical activity-induced local density differences. Finally, we discussed these results in the context of potential new fluidic applications and highlighted the role that activity-induced convection may play in experiments designed to investigate self-motile catalytic systems.



INTRODUCTION

Investigating the motion of catalytically active colloids capable of decomposing fuel molecules dissolved in a surrounding fluidic medium has become an area of significant interest over the last decade.[1] A significant amount of research in this area has focused on asymmetrical catalytically active "Janus" colloids that are able to produce enhanced motion by self-phoretic mechanisms or bubble release.[2−7] The capacity to utilize these catalytically propulsive colloids to enable a wide range of new fluidic applications in disciplines, including medical diagnostics, drug delivery, and environmental remediation, has been a key driver for this effort.[8] However, much less attention has been given to experimentally determining the motion phenomena for colloids performing catalytic reactions symmetrically over their entire surface. This is somewhat surprising, given the direct relevance to catalytic processes performed using isotropic distributions of the catalyst at a colloidal surface.[9] Recently, we partially addressed this issue by documenting the chaotic bubble propulsive motion of symmetrical catalytically active colloids.[10] However, bubble propulsion is not typical for uniformly active catalytic colloids, as it requires at least one gaseous reaction product, high surface reactivity,[11] and a large enough colloid radius to make the bubble nucleation energetically favorable.[12] Some theoretical studies have investigated the potential for enhanced motion for the more general case of a propulsive symmetrically catalytic active colloid without bubble propulsion. In one example, analysis of self-generated phoretic mechanisms in isolated individual symmetrical catalytically active colloids suggests that short-time scale deviations from conventional Brownian diffusion will be observed.[13] Additionally, the ability of individual isotropic catalytically active colloids above a certain size to undergo self-sustained phoretic motion has been theoretically predicted.[14] However, there are little existing experimental data to allow these predicted phoretic motion phenomena for isotropic catalytic colloids to be tested. Also, very recently, the possibility for a single symmetrical catalytically active colloid to undergo motion due to the self-generation of convection currents in a solid-walled container has been explored theoretically.[15] This latter phenomena of chemical activity-induced fluid convection is driven by heat evolution and/or the differences in the density of the reactant and product molecules involved in the catalytic reaction.[16] However, it is important to highlight that the phenomenology for convection is fundamentally different from self-phoresis: convection results in the entire bulk fluid moving, whereas self-phoresis causes individual colloids to move through the surrounding fluid, with a rapidly decaying local flow field. A related experimentally demonstrated example of chemical-induced convection, the ability of fixed solid surfaces decorated with areas of catalyst to produce convective flows,

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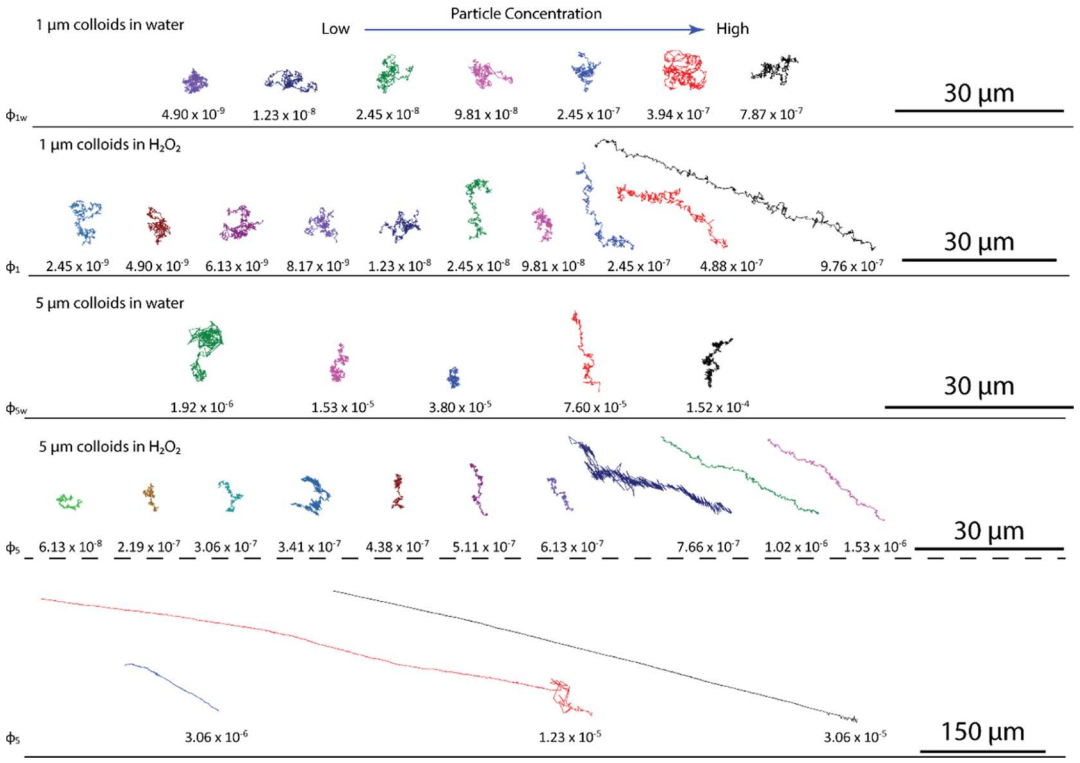

Figure 1. Representative trajectory data (30 s duration) for 1 and 5 μm colloids at varying particle concentrations in water and 10 w/v % hydrogen peroxide. Particle concentrations and equivalent volume fractions are shown below each trajectory.

has recently been proposed as a useful method of microfluidic pumping.[17,18] Whereas a similar collective fluid flow has been observed during previous experiments involving catalytic colloid systems,[19] systematic experiments to isolate and investigate the onset, character, and origin of this motion are lacking. This paucity of attention may partly stem from these drift phenomena being viewed as undesirable, as they hamper the study of self-phoretic or bubble release motility, which is the strong current focus of the active colloid research community. However, convective motion may in fact be desirable in a range of scenarios, for example, to enhance fluidic mixing and stirring and to potentially speed up diffusion-limited catalytic reactions carried out on colloidal supports. In addition, these phenomena are potentially more accessible as they do not rely on engineering a Janus structure at the colloid's surface.

Against this background, here we focus on exploring the motion of homogeneous catalytically active colloids coated with platinum decomposing dissolved hydrogen peroxide fuel. The aim of these experiments is to perform a quantitative analysis of convective, whole fluid drift and also to determine the presence of any phoretic propulsion effects of symmetrical colloids. The peroxide/platinum catalytic system is commonly used to power bubble-propulsive and self-phoretic devices[20−22] and so allows our results to directly inform this field. Furthermore, this catalyst system provides easily accessible rapid decomposition kinetics under ambient conditions. In addition, similar reactions have also been shown to cause fixed catalyst patch convective pumping, and so these prior data can provide mechanistic insights to aid the interpretation of our findings.[17] A recent theoretical analysis of convection by symmetrical active colloids was also based on the peroxide/platinum system.[15] Note that despite the decomposition of hydrogen peroxide evolving gaseous oxygen, the investigations could be performed without the complication of bubble propulsion by selecting appropriate colloidal sizes (smaller than our previous work[10]) and surface catalytic reaction rates.

We performed experiments over a range of well-defined volume fractions based on prior qualitative observations that a catalytic colloid drift is associated with high catalytic colloid concentrations. Low volume fraction experiments in the absence of a collective drift most easily allow the presence of any phoretic motion effects in our symmetrical catalytically active system to be established. Determining a link between the volume fraction and fluid flow will be useful not only to deliberately instigate these phenomena where desirable for future applications but also to determine limits where accurate measurements of phoretic propulsion without convection is possible. This latter goal is important, given the increasing interest in the collective behavior for self-phoretic colloids.[23,24] Our methodology is to use video microscopy combined with image analysis to arrive at quantitative trajectory data for a statistically significant number of colloids under each experimental condition. These trajectory data are then further processed to extract parameters that aid interpretation of colloidal diffusive, propulsion, and advection rate, including mean square displacements (MSD), Péclet number, and persistence length.

EXPERIMENTAL SECTION

Monodisperse polystyrene colloids 1 and 5 μm in diameter were chemically coated with platinum via in situ reduction of a platinum salt (Kisker Biotech custom synthesis). The coating obtained by this method consisted of a homogeneous distribution of 2−5 nm radius nanoparticles of platinum that were well-adhered to the surface of the colloids. To prepare samples for analysis at a range of volume fractions, a dilution series was made from a common aqueous stock colloidal suspension. To prepare a sample, aliquots were taken from an

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appropriate diluted solution and added directly into a cuvette. An additional equivalent volume of 20 w/v % H_2O_2 was added to obtain an overall H_2O_2 concentration of 10 w/v %. The remaining cuvette volume was then filled with 10% aqueous hydrogen peroxide. Prior to measurement, the sample was mixed via inverting the cuvette several times. For control experiments with 5 μm diameter colloids in water or at low volume fractions where there was no convective flow, the colloids would sediment to the bottom of the cuvette within a few minutes. In order for us to perform colloid measurements in bulk solution, the cuvette was inverted periodically as required to resuspend the colloids to allow free solution observations. Because of our observations being made in the horizontal observation plane, any vertical sedimentation velocity possessed by the colloids did not contribute to the subsequent trajectory analysis. The cuvette used was a 1 mm quartz cuvette (Hellma no. 110-1-40, path length 1 mm × 9.5 mm × 38 mm). Video microscopy of the contents of the cuvette was performed using an upright optical microscope (Nikon Eclipse LV100) equipped with an Andor CCD camera (Neo 5.5 sCMOS 5.5 megapixel resolution). To allow the colloidal motion to be analyzed, videos (frame rate of 33 fps) of at least 1000 frames were recorded at various positions within the cuvette. The center of mass for each colloid within the field of view was then determined as a function of time using custom threshold-based image analysis algorithms (LabVIEW). MSD and persistence lengths were calculated from the position/times series data, as described in detail previously.[10] High-magnification video inspection revealed that, in all experiments, there was no evidence of occurrence of bubble nucleation and detachment, confirming that the experiments were conducted under conditions that did not promote motion generation by bubble propulsion.

■ RESULTS AND DISCUSSION

Figure 1 displays typical 30 s duration trajectories for 1 and 5 μm diameter colloids uniformly coated with platinum in both water and hydrogen peroxide solutions at increasing volume fractions. For all conditions, the 1 μm colloids were observed in the bulk solution near the top or bottom of the cuvette, keeping a distance of approximately 100 μm away from the walls (to avoid diffusion hindrance due to the walls). It is clear that the trajectories of the colloids recorded in water appear similar at all volume fractions and qualitatively resemble Brownian diffusion. However, in the presence of hydrogen peroxide, at higher volume fractions, a directional drift in the trajectories for both 1 and 5 μm colloids was observed. The extent of the drift clearly increased with the colloid volume fraction. Strikingly, at the highest volume fraction observed for the 5 μm colloid, the drift entirely dominates stochastic motion, and colloids cover hundreds of microns during the period of observation. To assess the collective features of this drift phenomena, Figure 2B shows the trajectories of many neighboring 1 μm platinum-coated colloids in a high-volume fraction sample, for regions near the upper and lower walls of the cuvette (indicated by the image planes in Figure 2A), recorded consecutively within a time frame of 1−2 min. These observations allow the individual trajectories shown in Figure 1 to be understood in the context of the collective motion of colloids over a large region of the cuvette. It is consequently clear that there was an overall collective movement within each region, evidenced by the drift of a similar magnitude and direction for each individual colloid. In addition, it is clear that the collective direction of motion was reversed between the top and bottom of the cuvette. Observations at the vertical walls of the cuvette confirmed that the colloids were sinking and rising in directions consistent with a continuous cyclical motion illustrated in the inset in Figure 2A. In addition, observations in the middle of the cell revealed that colloids were mostly undergoing Brownian

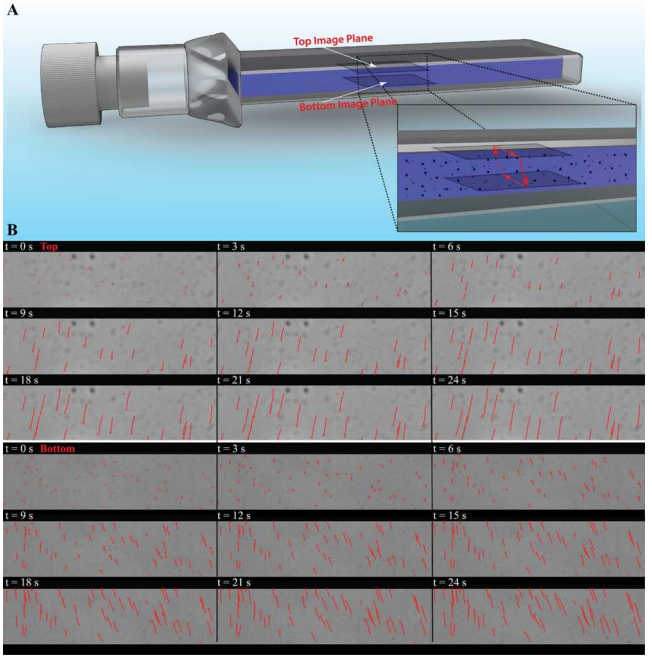

Figure 2. (A) Schematic of the cuvette used in all motion studies. The inset shows the image planes at which the image series were captured, and the red arrows indicate the direction of particle circulation within the cell, determined using video microscopy. (B) Representative time-stamped image sequences recorded at the top and bottom image planes for 1 μm platinum-coated colloids in 10 w/v % H_2O_2 (crosshairs indicate the current position of each colloid in each frame, and red lines show the past trajectory). See also Video S1.

motion, without any notable flow affecting the particles (see Video S1). Similar circulations were also observed for high volume fraction samples of 5 μm colloids. This observation that each colloid moves in a similar direction within a given region of the cell and the overall continuity of motion within the cuvette strongly suggest that at high volume fractions, the entire fluid undergoes a convective flow driven by the catalytic decomposition of hydrogen peroxide. We note that the orientation of the circulations of the colloids relative to the cuvette geometry would vary between different experiments, indicating that the onset of the collective motion phenomena may be to some extent chaotic. To illustrate that the entire fluid was moving, 1 μm fluorescent tracer particles were mixed with 5 μm platinum-coated colloids in water and hydrogen peroxide, as shown in Videos S2 and S3, respectively.

Having understood the qualitative features of our observations, we now examined the quantitative details that could be extracted from the colloidal trajectories. To do so, we fitted MSD curves (ΔL^2) (averaged over observations made for many colloids at each condition) as a function of the time step (Δt) to the expression $\Delta L^2 = 4D\Delta t + v^2\Delta t^2$, allowing both the magnitude of any ballistic velocity, v, and the Brownian diffusion coefficient, D, to be determined.[5] To start with, we considered trajectories recorded at low volume fractions, where there is no evidence of the collective convection phenomena. The MSD versus time data in the absence of convection for both 1 and 5 μm with or without the presence of hydrogen peroxide fuel colloids was linear (see Figure S1/S2), consistent with Brownian diffusion. Furthermore, at low volume fractions, there was no consistent difference in the diffusion coefficient attributable to the decomposition of peroxide fuel compared to

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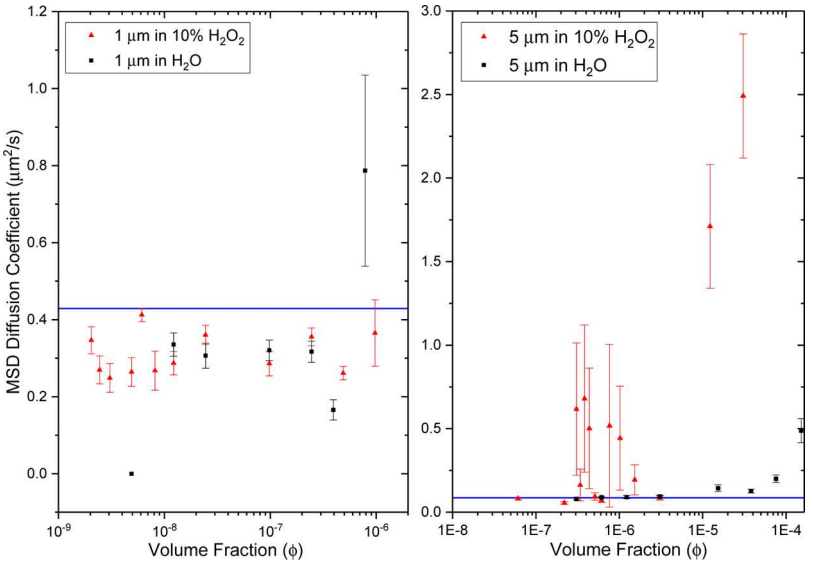

Figure 3. Average diffusion coefficient for 1 and 5 μm uniformly platinum-coated colloids in water and 10% H_2O_2 determined using MSD fitting to the trajectory data. The blue lines indicate the theoretical diffusion coefficients for particles of this size, being 0.429 $\mu m^2/s$ for 1 μm and 0.086 $\mu m^2/s$ for 5 μm colloids.

the values measured for catalytically inactive colloids in water (Figure 3. The values of D determined by MSD fitting also showed good agreement with the values calculated using the Stokes−Einstein equation. These data consequently does not provide any evidence that the symmetrical catalytic colloids studied here individually display enhanced or modified motion phenomena such as self-phoresis, subject to the time resolution limits available in this experiment. This finding simplifies the interpretation of the subsequent collective motion phenomena and provides a further justification for using symmetrically active colloids to investigate convection, rather than self-motile Janus colloids.

At the higher volume fractions in the presence of hydrogen peroxide, that is, under the conditions observed to produce a collective drift in the trajectories shown in Figure 1, corresponding MSD curves are parabolic, consistent with colloids that undergo ballistic motion (see Figure S2). Figure 4 displays the ballistic velocities determined by fitting the MSD data as a function of the volume fraction for both catalytically active and inactive colloids (in water). This analysis reveals that at the highest volume fractions, the average velocity was 12 μm s^{-1} for active 5 μm colloids and 1.6 μm s^{-1} for active 1 μm colloids. For the larger particle size, this velocity is significant and of an equivalent magnitude to that produced by self-phoretic Janus motors, suggesting that useful catalytic transport and pumping effects can be accessed at higher volume fractions even for colloids that are not capable of individually generating enhanced motion by phoresis or bubble propulsion.[5] It is also clear that significant ballistic velocities are only introduced above a certain volume fraction and that the velocities continue to increase with the volume fraction over the experimental conditions we accessed. In addition, it can be seen that the onset of significant ballistic motion for the 1 μm colloids occurs at a lower volume fraction than for the 5 μm colloids.

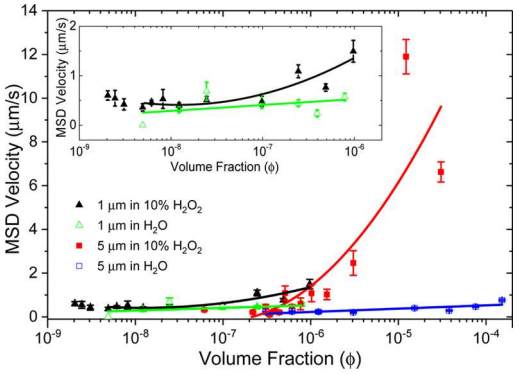

Figure 4. Mean platinum-coated colloid velocity (determined from a quadratic fit to MSD data) for 1 and 5 μm colloids in water and 10 w/v % H_2O_2 as a function of the volume fraction (inset is an expansion for the 1 μm data). Fitted lines are shown to guide the eye.

Control data for the same colloids measured in water reveal a very slight increase in the ballistic velocity with increasing volume fraction, but all velocities remained less than 0.5 μm s^{-1}. Although in theory, purely diffusive colloids should give a ballistic velocity of zero, in many previous studies by ourselves and others, a similar nonzero ballistic velocity has been reported for inactive colloids.[5] This reflects the difficulties in experimentally establishing true Brownian conditions and is the justification for recording these control data, to provide a baseline from which convection or propulsion can be separated. It is likely that thermally induced flow due to microscope illumination may be one factor contributing to this nonzero velocity. As for the increase in this nonzero velocity with the volume fraction, there is no obvious physical mechanism that would account for this, and the trend is in any case somewhat weak. Figure 3 also shows an apparent increase in the diffusion coefficient corresponding to conditions producing appreciable ballistic propulsion, which is likely to be due to cross-talk in the numerical fits, rather than a physical phenomenon. Although our assignment of the nonzero ballistic velocities here to convection is supported by observations of the collective fluid motion within the whole cuvette, we also consider the extent to which MSD analysis alone can be used to distinguish self-propulsion from convection. This is relevant in scenarios where it is difficult or impossible to assess overall colloidal/fluid motion. For the smaller particle size, the MSD curves alone can

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clearly distinguish the behavior we observe here from the case of an equivalently sized colloid producing a self-propulsion vector that corotates with the colloid body, such as a self-phoretic Janus colloid (see Figures S1 and S2). It is well-established analytically and experimentally that in this case, the rapid Brownian rotation rate (Brownian diffusion time 1 μm colloids $\tau_R^{-1} = 0.78\ s^{-1}$) means that pure parabolic MSD versus time step curves are only expected at very short time scales ($\Delta\tau < 1$ s) and that beyond this, the MSD will become linear, with a gradient increase reflecting the degree of enhanced diffusion.[25] In this respect, the MSD curves for 1 μm colloids at higher volume fractions are parabolic MSD over the entire accessible time scale (30 s) and so not consistent with corotating self-propulsion (see Figures S3 and S4). Instead, convective drift, in the regions of the cuvette we performed analysis in, is well-approximated by Brownian colloids subject to a ballistic propulsion vector with a constant velocity and magnitude. However, for the 5 μm colloids, the Brownian rotation rate is much slower (Brownian diffusion time 5 μm colloids $\tau_R^{-1} = 97\ s^{-1}$), and the MSD for convection with constant direction or self-generated corotating propulsion is consequently hard to distinguish without observing the colloid for a long period. We also highlight that in this experiment, we analyzed colloidal motion far from the edges of the cuvette. However, to ensure fluid continuity, the convective flows' direction does in fact change in different positions within the cuvette (see Figure 2), so we can expect complicated MSD curves to be observed near the boundaries of cuvettes or for fluids constrained in containers with a complex geometry. In these cases, the convective propulsion vectors' direction and magnitude in the plane of analysis will vary.

As a high volume fraction catalytic convection might be exploited for mass transport applications, it is also useful to evaluate a Péclet number for our experiments, reflecting the ratio of advection to the natural Brownian colloidal diffusion rate. In analogy to a similar analysis for self-phoretic colloids,[26] we define the relevant Péclet number, $Pe = vd/D$, where v is the ballistic velocity, d is the particle diameter, and D is the Stoke–Einstein calculated diffusion coefficient for the colloid. Figure 5 displays Péclet numbers as a function of the volume fraction and highlights the order of magnitude increase in this measure of mass transport that can be achieved for 5 μm catalytically active colloids simply by increasing their volume fraction.

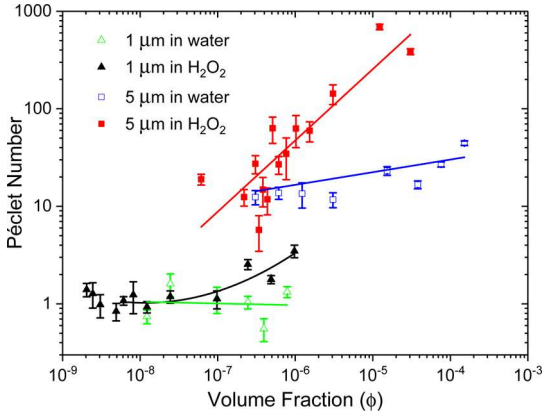

Figure 5. Péclet number for 1 and 5 μm platinum-coated colloids in water and 10% H_2O_2 as a function of the volume fraction. Lines are shown to guide the eye.

Finally, to verify the mechanism for the observed catalytically induced convection, we have attempted to normalize the data sets obtained for the two differently sized colloids based on the overall amount of catalytic activity in each experimental condition. This normalization is based on previous theoretical and experimental analysis for pumps driven by fixed surface bound patches of enzymes, which found that the origin of fluid flow is primarily due to the density differences between the reagents and the products resulting in convection.[17] Consequently, the increased overall hydrogen peroxide decomposition rate is expected to correlate with an increased degree of convection: a link that has been verified for the surface-localized enzymatic pumps.[17] In our system, we have previously quantified surface reaction rates for the platinum-coated colloids: expressed as molecules of hydrogen peroxide decomposed per unit surface area, $k_{1\mu m} = 3.7 \times 10^9 \pm 4.2 \times 10^8\ s^{-1}\ \mu m^{-2}$ and $k_{5\mu m} = 9.6 \times 10^9 \pm 4.2 \times 10^8\ s^{-1}\ \mu m^{-2}$, that is, the platinum coating of the 5 μm colloid is almost 3 times more intrinsically reactive than the coating on the smaller 1 μm colloids. Note that this difference in reactivity is likely to be due to differences in the uncontrolled properties of the different-sized chemically coated colloids (e.g., roughness and coating thickness), rather than a general finding. These parameters allowed us to estimate the overall decomposition rate per mL of solution in our experiments. For this analysis, we chose to use persistence lengths as our measure of convection, reflecting the average length over which a given colloid moves in a constant direction, and were thus expected to provide a good indicator of the degree of convection for the regions of the cuvette in which we performed the analysis. Figure 6 reveals a plot of

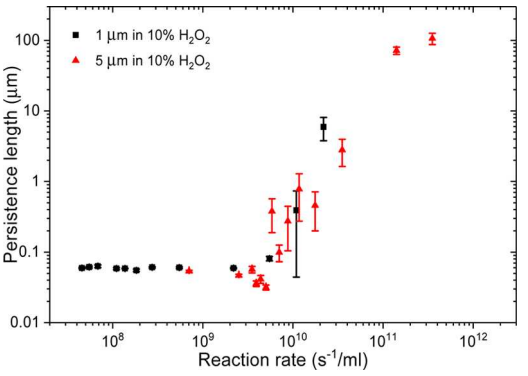

Figure 6. Persistence length as a function of the overall colloid reaction rate per unit volume.

persistence length[10] against the overall colloidal activity per mL in the cuvette. The overall catalytic activity was determined by evaluating the product of the number density of colloids per unit volume, surface area of an individual colloid, and the reaction rate per unit area (i.e., reaction rate per unit volume = $6\phi k/d$, where d is the diameter of the colloid). This procedure produces a good collapse of the data and suggests that a criterion of a reaction rate in excess of $5 \times 10^9\ s^{-1}\ mL^{-1}$ is needed to initiate a convective drift via catalytic activity within our particular cuvette geometry.

DISCUSSION

The observation that catalytic activity at the surface of colloids can induce bulk convective fluid motion is related to the previous use of surface-bound catalytic patches to "pump"

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fluids.[17,18] However, in contrast to fixed catalytic patches, the catalytic colloids that induce the fluid pumping in this study are themselves motile, which opens up new possibilities. For example, our results indicate that the onset of catalytic pumping is instigated only after the accumulation of a certain volume fraction of colloids in a given region, which could be exploited to "switch on" convective mixing at a desired stage of a lab-on-a-chip operation. This convective pumping phenomena hence appear attractive for exploitation in combination with other existing and emerging methods for controlling local colloidal densities. Examples include external colloidal manipulations using magnetic fields and the various self-phoretic and bubble propulsive transport phenomena possessed by catalytic colloids, which can spontaneously result in high volume fraction cluster formation.[27] In addition to local mixing and fluid transport applications, collective convective motion in colloids used as catalytic supports for heterogeneous catalysis could speed up diffusion-limited reactions. Indeed, this effect may be present, but not necessarily optimized, in existing catalyst systems.

It is likely that the convective flow observed here arises from the chemical activity generated by density differences around each active colloid. The products of hydrogen peroxide decomposition are less dense than the reagents, which is known to generate a rising convective flow above patches of catalase, performing hydrogen peroxide decomposition.[17] Recently, the way in which this phenomena can produce motion for a single symmetrical catalytically active colloid decomposing hydrogen peroxide in a walled container has been theoretically investigated.[15] Modeling this scenario showed how the density differences generated by the catalytic reaction at the colloid surface lead to fluid flows. When the colloid was positioned away from the center of the container, it experienced asymmetrical flows, resulting in motion toward the container walls, and eventually became trapped at the corner of the container. Removing sharp corners from the containers provided a route to achieve a sustained circulating flow in the presence of a second, passive particle. Although these simulations were performed with the colloids localized at a planar interface and only considered a single active particle, they appear to capture the essence of the phenomena we observe. In our experiment, it is the combination of each individual local density-driven flow and boundary conditions imposed by the cuvette that lead to the observed cyclical motion. An additional potential driving force for motion in our system is thermal-induced convection because of local temperature changes associated with the decomposition reaction. However, this has previously been assessed to be a minor effect in the related case of catalase decomposing hydrogen peroxide.[17]

A further result from our work is that we did not find any evidence for modified diffusion phenomena for either 1 or 5 μm diameter symmetrical catalytically active colloids at low volume fractions. However, we note that our experiment does not have the temporal resolution to assess the anomalous diffusion phenomena suggested by Golestanian,[13] and our system may not meet the criteria required for the suggested Autophoretic mechanism.[14] On this basis, verifying self-phoretic phenomena for symmetrical colloids will require further study.

Finally, in addition to the potential to positively exploit the collective convection reported here, convection remains a significant challenge when investigating self-phoresis and other catalytically induced motility phenomena. Here, we have identified a threshold for the catalytic collective convection onset based on the total colloidal chemical activity per unit fluid volume. Although this onset parameter may be specific to our cuvette geometry, it is likely that the flows we observed are a general phenomenon for experiments where catalytic colloids, symmetrical or otherwise, are confined for observation and perform reactions that can generate density variations. It consequently appears that when conducting experiments beyond a certain volume fraction/catalytic activity level, convective motion will be inevitable. Here, we have also shown that for individual colloids, the resulting trajectories can be hard to distinguish from self-phoretic effects even with the use of MSD analysis. These findings emphasize the requirement for researchers quantifying the motion of catalytic colloids to use methods that can identify convective motion. These include looking for cyclical patterns of collective motion and deploying inactive tracer particles as required to reveal bulk fluid flow.

CONCLUSIONS

We have studied the motion of 1 and 5 μm diameter colloids symmetrically coated with platinum while catalytically decomposing dissolved hydrogen peroxide. At low volume fractions, individual catalytically active colloids' motion was indistinguishable from Brownian diffusion. However, as the volume fraction increased, the onset of collective cyclical convective flow was observed. Particle tracking and MSD trajectory analysis were used to quantify the ballistic propulsion velocity due to the convective flow, which was found to progressively increase with the volume fraction. The magnitude of convective drift, quantified using persistence lengths, was found to be well-correlated with the reaction rate per unit volume of solution for both colloid sizes. On this basis, it is likely that the mechanism driving the flow is the solution density variations produced by the catalytic reaction. The convective flow significantly increases the colloidal Péclet number, suggesting applications for mass transport. Deliberately exploiting this volume fraction-dependent phenomena provides a potential new route to instigate fluid pumping in lab-on-a-chip systems. Additionally, there is potential to control the convective flow to enhance the reaction rate during the diffusion-limited heterogeneous catalysis. We also highlight that collective convective motion is likely to occur in many experiments designed to investigate self-motile catalytic systems, rendering it important that appropriate protocols are used to distinguish convection from self-propulsion. In this respect, we hope this study will be of relevance to other experimentalists working in the area of active colloid research.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.langmuir.8b00310.

Partial and full MSD curves for 1 and 5 μm platinum-coated polystyrene colloids in water and 10% v/wt hydrogen peroxide at low and high volume fractions (PDF)

Platinum-coated colloids (1 μm) in 10% v/w hydrogen peroxide undergoing induced convective flow in the cuvette at the top, middle, and bottom image planes over a time frame of 30 seconds (ZIP)

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Platinum-coated colloids (5 μm) in water with 1 μm fluorescent polystyrene tracer particles undergoing Brownian motion (ZIP)

Platinum-coated colloids (5 μm) in 5% v/w hydrogen peroxide with 1 μm fluorescent polystyrene tracer particles undergoing convective flow (ZIP)

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Notes

The authors declare no competing financial interest.

ABBREVIATIONS

MSD mean squared displacement

CCD charge-coupled device

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