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Modeling the biomimetic self-organization of active objects in fluids

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ABSTRACT

Living cells self-organize to perform various complex tasks that a single cell alone could not accomplish. Few synthetic nano- or micro-scale objects can autonomously carry out comparable coordinated behavior. We develop computational models to design fluid-filled microchambers containing both nano- and micro-particles that convert inputs of thermal, optical or chemical energy into mechanical motion to exhibit collective biomimetic activity. For example, the heat generated by illuminating the solution of nanoparticles within the microchamber drives the dispersed microparticles to self-organize into colloidal crystals that follow a moving light source, and thus exhibit life-like photo-tactic behavior. For microchambers containing catalyst-coated flexible sheets, chemo-responsive microparticles and the appropriate reactants, the sheets self-organize into autonomously moving "predators" that effectively collaborate to trap the "prey-like" particles. These computational studies provide valuable guidelines for developing microfluidic devices and soft-robots displaying autonomous bio-inspired properties that greatly expand the functionality of these systems.

The collective behavior of "active" nano- and micro-scopic particles in solution can resemble forms of self-organization found in nature, including the migration of photo-tactic organisms toward light or the dynamics of predator-prey interactions. Synthetic active particles are propelled by energy inputs from an external source (e.g., heat or light) or from chemical reactions occurring on a particle's surface. By harnessing thermal, optical or chemical energy, active particles undergo self-sustained motion in the surrounding fluid and hence, perform mechanical work. Commonly referred to as "motors", these particles can autonomously transport or direct cargo in fluid-filled microchambers and thereby increase the portability of microfluidic devices. Additionally, clusters of motors can perform complex, collaborative tasks and facilitate the development of small-scale robots that operate within fluidic environments (e.g., such as blood vessels).

The phenomena involved in the propulsion of active particles encompass the reaction and diffusion of chemicals, conduction of heat, generation of chemical or thermal gradients, and convection of fluids in microchambers. A significant challenge in developing active particles for specific applications is establishing design rules that take into account the different spatial, temporal, and energy domains involved in these complex, dynamic systems. Below, we highlight recent computational modeling studies aimed at addressing this challenge and facilitating the creation of fluidic systems exhibiting useful, life-like functionality. These simulations reveal how exposing dissolved nanoparticles to light converts optical and thermal energy into the mechanical motion of the host fluid and dispersed microparticles, which subsequently move toward the light. The studies provide guidelines for harnessing irradiated nanoparticles to reversibly construct and move colloidal crystals in microchambers. Additional simulations show that by simply tilting this illuminated microchamber, polydisperse mixtures of microparticles can be readily separated by size (or density). Finally, we describe simulations where the interplay of chemical and mechanical energy enables active sheets to collaborate and thus, entrap small-scale "prey".

When gold nanoparticles within a fluid-filled microchamber are irradiated with UV light, this energy is dissipated into heating the solution, which consequently undergoes net motion [1,2]. In Fig. 1A, the light beneath the transparent bottom wall produces temperature gradients and the heated, less dense fluid rises upward. Since the fluid is confined in the microchamber, the flow circulates to the bottom wall, forming an "inward" flow. If the solution contains sedimented microparticles, then the thermally generated inward flow imposes drag forces that drive the aggregation of the microparticles and the formation of colloidal crystals [1], which are useful for creating novel sensors, coatings, and optoelectronic devices [3].

In Fig. 1A–D, the crystal formation is nucleated at points where the light beam enters the microchamber. Moving the light source to a new position causes the cluster formation to occur around this

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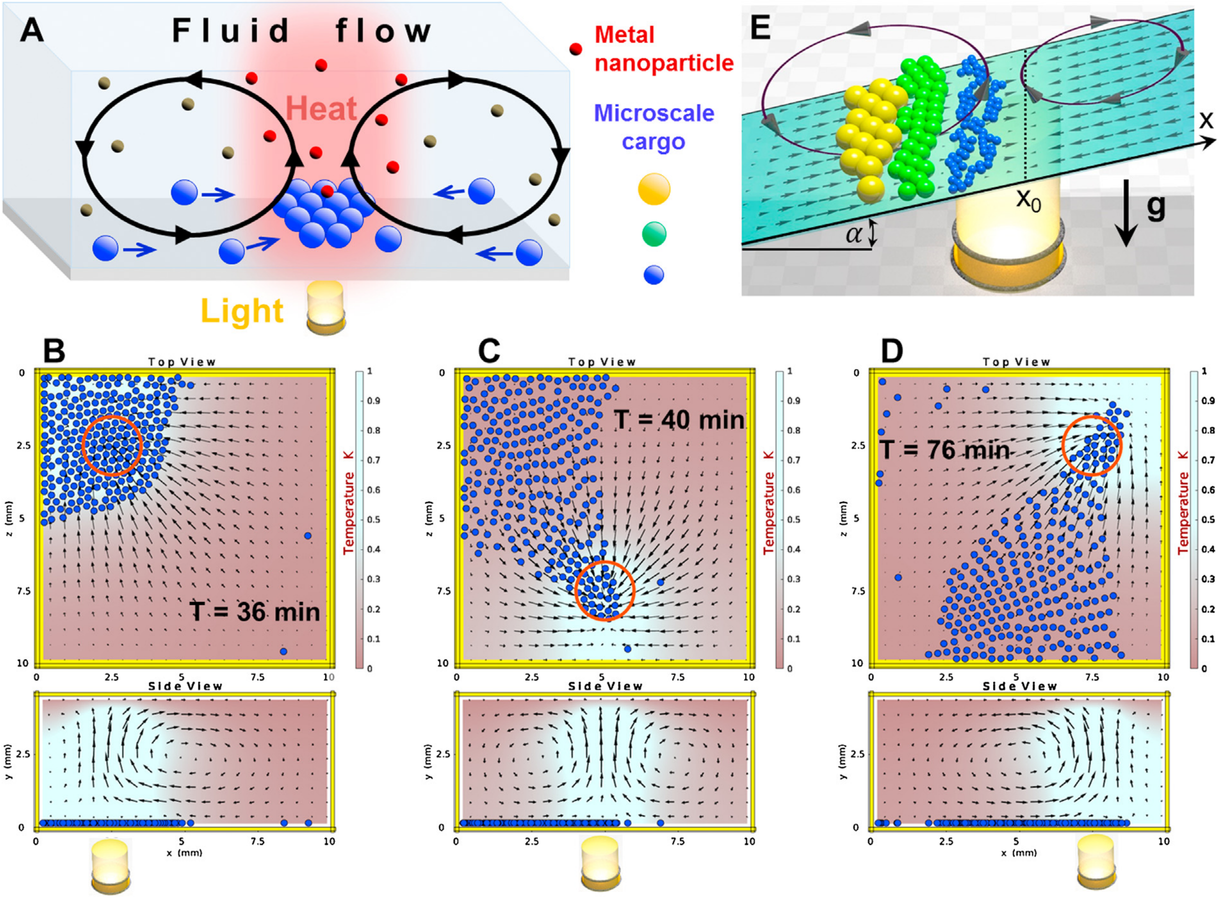

Fig. 1. Hydrodynamic "tweezers". (A). The bottom of the chamber is illuminated by an external light source. The fluid flow induced by the heat released from the irradiated nanoparticles aggregates microparticles into a colloidal cluster. The aggregated cluster (B) is driven to new positions (C and D) by the moving light source. (E) Segregation of different size particles by convective flow on the bottom of the inclined chamber.

new location (Fig. 1B–D). For example, particles of radius 150 μm aggregate into a single cluster in approximately 30 min. If the light source is moved sufficiently slowly, then the entire self-organized cluster follows the moving light without losing its integrity. Hence, the illuminated, dispersed nanoparticles function as "tweezers" that collect and relocate microscopic cargo to a desired position within the chamber. The temperature increase resulting from heating the solution is approximately one Kelvin, which is compatible with living specimens, and thus, these tweezers could gather bacteria or other biological cells and concentrate these species around a sensor for a biochemical analysis.

When this illuminated, nanoparticle-containing chamber is inclined with respect to the horizontal direction, the competition between gravity and the fluid drag acting on the microparticles provides a method for sorting cargo with different properties [4]. The dissipated heat again generates an inward flow (Fig. 1E); at a point x_0 between the resultant counter-rotating vortexes, where the flow velocity parallel to the wall is zero. As indicated by the arrows in Fig. 1E, the flow field about x_0 changes sign, with the drag flow driving the particles up the incline at $x < x_0$. On the other hand, gravity drives the sedimented particles to the bottom of the channel. Hence at $x < x_0$, the two effects drive particles in opposite directions. Scaling arguments indicate that particles of different sizes and/or densities attain a balance between the competing forces at different locations along the inclined wall. This size-dependent separation is clearly evident in Fig. 1E. The separation distance between the particles depends on the inclination angle and relative differences in the particle sizes or densities. For particles with radii of 40 and 120 μm, the separation distance between the respective clusters is approximately 0.5 mm at an inclination angle of 15°. This system is of significant technological importance since it provides a simple, low-cost approach for particle sorting, which forms a vital step in numerous industrial processes.

Fig. 2 illustrates another scenario where nanoscopic objects can drive microscopic flow. In this case, an isolated crystal of catalase is nano-scopic in one of its dimensions. In the dissolved state, catalase decomposes hydrogen peroxide into water and oxygen; through this catalytic process, even a single molecule of catalase anchored to a surface can act as a "chemical pump", enhancing diffusion of solute in a microchamber [5]. The micron-sized flexible sheets in Fig. 2 are partially coated with catalase (area marked in dark green) [6]. In the simulations, the sheets are modeled as a single layer of interconnected microparticles that form a four-lobed structure; the lobes' black tips are taken to be heavier than the catalase-coated region.

Within this system, two distinct mechanisms drive the transduction of chemical energy into mechanical motion: solutal buoyancy [7] and diffusiophoresis [8]. The solutal buoyancy mechanism becomes operative when the catalase on the sheet decomposes H_2O_2 into the less dense products (H_2O and O_2) and thereby creates density variations in the fluid. Analogous to the thermal buoyancy effects described above, the product-rich fluid rises upward from the surface to produce inward flow (black arrows in Fig. 2A). The centers of the sheets in these simulations (Fig. 2A–B) are restricted from moving in the lateral dimension, but the chemically-generated inward flow below the sheet drives the

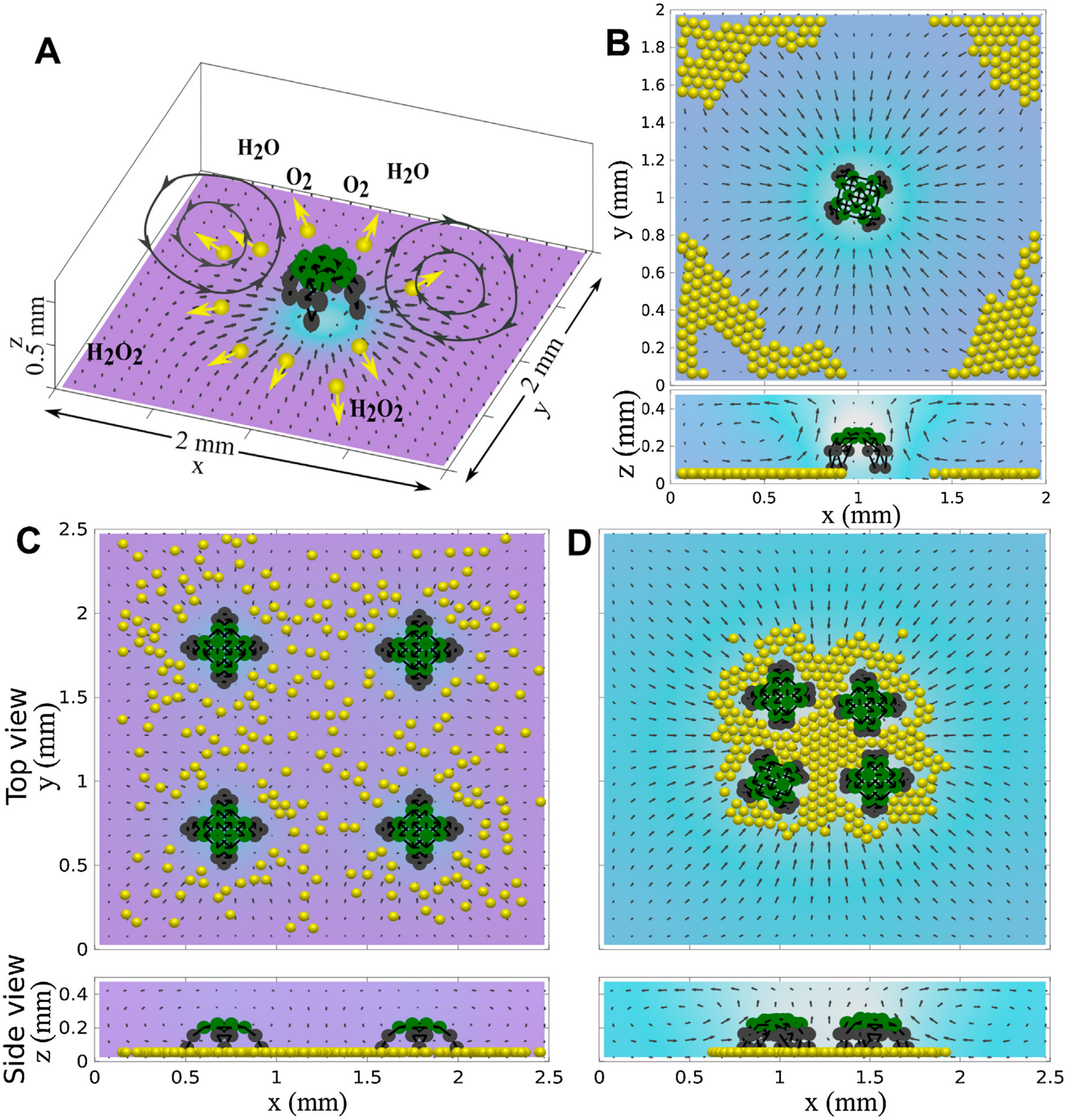

Fig. 2. Competition between chemically active objects. (A). A single chemically active sheet hydrodynamically attracts escaping diffusiophoretic particles. (B) Particles are driven away from the isolated sheet. (C)–(D) The combined fluid flow generated by four mobile sheets traps the particles and thus mimics collaborative behavior.

central portion to “pop up”, with the heavier black beads remaining localized on the surface. In other words, the active sheet generates fluid flows that sculpt the sheet into a crab-like shape [9].

The conversion of reactants to products in this system produces variations in the concentration of the chemicals in the solution. The non-uniform distribution of solute molecules along the surface of the yellow microparticles in Fig. 2 drives the motion of the solute to the less concentrated regions. Due to conservation of momentum, the microparticles spontaneously move in the direction opposite to the solute molecules, giving rise to diffusiophoretic motion (yellow arrows in Fig. 2A).

The components in Fig. 2 represent a highly interdependent system: the reactions at the surface of an active, catalase-coated sheet generate the forces that drive both the solutal buoyancy and diffusiophoretic motion. By tuning the relevant parameters, one mechanism can dominate over the other. In particular, the diffusiophoretic mechanism dictates that the particles move along the direction of the highest chemical gradients. Since the reactants are consumed at the sheet and the products diffuse towards the edge of the chamber, the highest chemical gradients point away from the sheet. Hence, when diffusiophoresis dominates, the particles are located relatively far from the sheet (Fig. 2B) and appear to flee from the crab-shaped object.

The particles can, however, be trapped if the effects of solutal buoyancy are dominant, as in Fig. 2C–D. The four crabs are now completely mobile (the centers of sheets are no longer constrained in the simulations). The chemically generated fluid not only molds the shape of the sheets, but also propels their movement in the chamber. The crabs are driven towards each other by the collective effects of the inward flow; as crabs get closer together these effects are amplified, increasing the magnitude of the fluid velocity around the self-organizing sheets. The enhanced flow draws the particles toward the aggregate and hence the four "collaborative" mobile sheets can catch and trap the prey, performing a task that one alone could not achieve. Moreover, since the active sheets are both reconfigurable and self-propelled, they can perform self-sustained operations that would not be possible with stationary chemical pumps or non-deformable chemical motors. The active sheets also act as sensors since the catalysts on the surface only respond to the appropriate reactants [10].

These examples illustrate how computational approaches that capture phenomena extending over nano and micro-scales and different energy domains (e.g., opto-thermo-mechanical and chemo-mechanical) can guide the fabrication of portable microfluidic devices that operate in a relatively autonomous manner, alleviating the need for extraneous mechanical or electrical equipment [11]. A remaining challenge is designing devices that incorporate catalytic cascades, where the product of one reaction is the reactant for the next, and thus could autonomously perform multi-step processes prevalent in chemical synthesis. Notably, the chemical industry is rapidly undergoing a transition from large manufacturing plants to microscale devices, which enable more sustainable and efficient processing. Such multi-scale computer simulations can greatly facilitate and accelerate this transition.

Declaration of Competing Interest

Authors do not have any conflict of interest to declare.

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