A guide to design the trajectory of active particles: From fundamentals to applications

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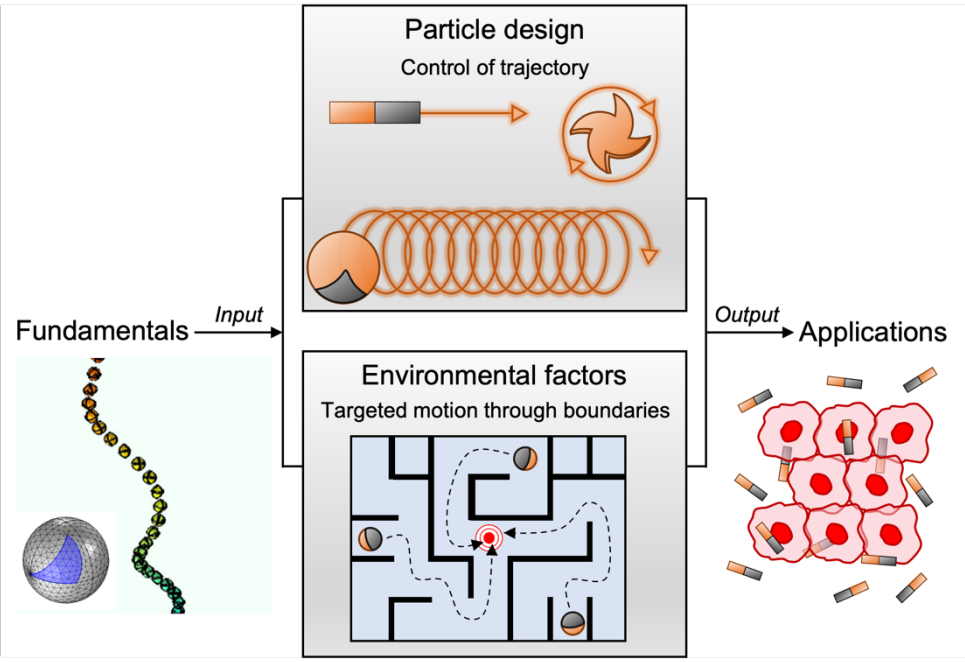

Abstract

Active particles convert external energy into motility, displaying a variety of dynamical features. Recent progress in the field has marked a shift in focus from understanding the origin and sources of active motion to controlling the dynamics and trajectory of individual microswimmers. This Review explores the advancements made in a two-fold perspective – the role of particle design and that of external factors. Our main goal is to highlight the guiding principles which determine active particle trajectory. These include, on the one hand, the role of the morphology of active particles and their assemblies in driving translation, rotation, and corresponding coupling between the two. On the other hand, the effect of environmental parameters such as the presence of physicochemical heterogeneities including interfaces, suspended obstacles, and boundaries on the modality and trajectory of active colloids. We discuss the potential of using active particles in biomedical and environmental applications through recent examples.

Keywords: Active particles; Self-propelling colloids; Colloidal dynamics; Motion trajectory; Complex environments; Biomedical applications

1. Introduction

Active colloids are composed of micro- and nano-sized particles that convert external energy into motion within a continuous medium. These particles may be fueled by chemical gradients [1] in their suspending medium or be powered by external fields applied remotely [2]. The resulting motion and corresponding dynamic phenomena have raised much interest and contribute to the development of the area of active and responsive matter. From a fundamental perspective, active particles have proved an invaluable tool as a model system to study non-equilibrium behavior of living matter such as micro-organisms [3]. In addition, the analogy between synthetic and living active matter has been fruitful in a dual way. On the one hand, the motion of birds [4], fish [5], and other swimming micro-organisms [6,7] has inspired the fabrication of biomimetic artificial active particles with improved motility. On the other hand, controlled experiments using synthetic microswimmers have elucidated the mechanism of motion of certain bacteria [8]. The technical application of active particles has great potential lying in the ability of tiny, self-propelling objects to travel and perform tasks in complex environments [9,10]. The expectation is to advance the functionality of active particles towards application in areas such as *in vivo* drug delivery [11] and toxic waste removal [12]. One key hurdle to the successful translation of active particles toward technology lies in achieving practical control over their dynamics.

To exploit the motility of active particles for practical microscale applications, it is important to consider their defining characteristics. In the absence of an external force, colloidal particles display Brownian dynamics, i.e., random motion originating from thermal energy. A passive particle is defined by purely diffusive motion and is described by a mean square displacement expressed by the Einstein-Smoluchowski equation. Active particles acquire unique dynamics that originates from the energy source powering their motion. These particles travel along self-generated gradients in chemical potential, which would not otherwise exist in systems of passive particles. Such directed motion is subject to reorientation resulting from thermal fluctuations. The conversion of external energy may enhance the mean square displacement of active particles yet controlling the trajectory of motion remains a challenge. Various modes of active motion show different degrees of control over the dynamics, ranging from superdiffusive behaviors [13,14] analogous to run-and-rumble bacteria to highly directed motion in external fields [15]. Many particles displaying self-propulsion have non-trivial interactions with each other that affect their colloidal stability. This often leads to collective behaviors such as motility-induced phase separation and the formation of dynamic, activity-dependent states. In this Article, we mostly limit our discussion to the dynamics of single particles and redirect the reader to previous reviews[16–18] for in-depth analysis of collective behaviors.

Real-world application of active particles requires the highest achievable programmability in control over the directed motion (Fig. 1a). The recent literature reports a variety of modes of motion with unusual trajectories [19–24] and complex interactions with the surrounding environment [25–30]. We aim to partially unravel some key emerging rules that provide a useful toolset for controlling the motion of active particles. These concepts can be classified into two broad categories:

1. Factors intrinsic to the particle itself, i.e., its design and properties including shape, patch, and assembled structure (Fig. 1b).
2. Factors external to the particle, i.e., environmental parameters relating to the interaction of particles with external fields, complex suspensions, and boundaries (Fig. 1c).

By surveying recent progress in these areas, we focus on how these factors affect motion to find guiding principles for the design of active particle trajectories. We conclude by discussing interesting emerging applications and commenting on the future of the potential technology provided by active particles.

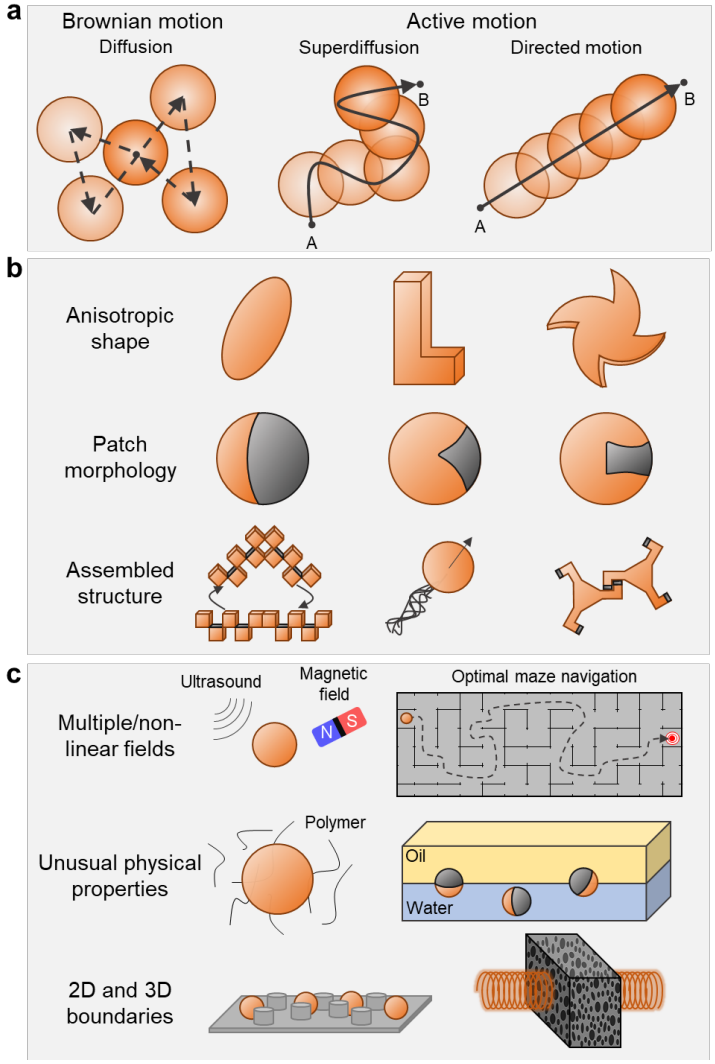

***Figure 1**: Rationale and structure of the article. **a** The dynamics of particles moving by Brownian diffusion differ from the dynamics of active particles. The trajectory of self-propelled particles displays random superdiffusion but can also lead to more directed motion. **b** Properties of microswimmers that affect trajectory of motion include shape, surface chemistry and assembled configuration i.e. factors internal to the particle. **c** Factors external to the active particle affecting its motion include environmental heterogeneities such as energetic landscape, unusual suspension properties and rough and porous media.*

2. Trajectory control by particle design

Breaking the spatial symmetry of forces acting on a particle is a fundamental requirement to successfully convert external energy into an active form of motion. This is true irrespective of the many available energy sources (Fig. 2a), thus making particle design fundamental to the study and application of active colloids. Such design must contend with the limitations inherent to locomotion in a low Reynolds (Re) number environment. This describes the fluid regime in which suspended microparticles experience viscous forces that are several orders of magnitude stronger than inertial forces. When Re << 1, reciprocal motions of a swimmer do not produce net displacement according to Purcell's 'scallop theorem' [31]. There are two main design routes that employ symmetry breaking to achieve active motion: (1) swimmers that self-propel by generating and moving across local chemical potential gradients, and (2) swimmers that perform nonreciprocal geometrical deformations or reorientations that result in translation. Strategies for successful deployment of self-propelled particles always involve designing a symmetry-breaking element within their morphology. Thus, the motion and trajectory of an active swimmer is encoded in either its surface properties, its shape, or a combination of the two.

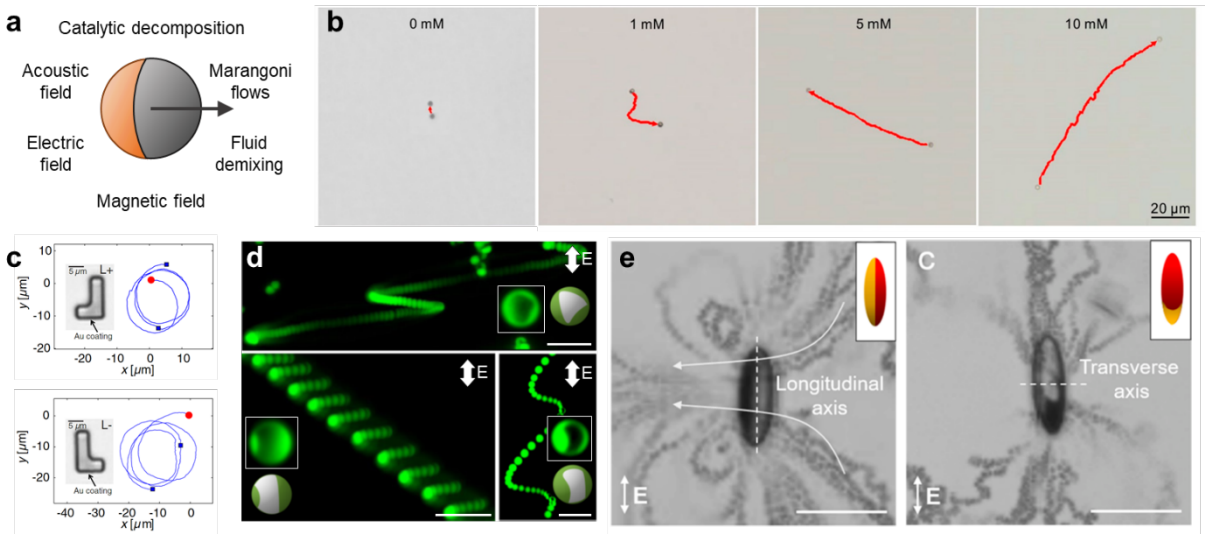

***Figure 2**: Self-propulsion of active particles in linear and non-linear trajectories. **a** Schematic of propulsion modes of a Janus particle. **b** Janus particle half-coated with multiple urease layers that are immobilized on a thin layer of Au, propelling in solutions of increasing urea concentration. The motion-trace reflect propulsion over a timeframe of 5 seconds, showing increased speed with increasing urea concentration. Reprinted with permission from [33]. Copyright 2020, American Chemical Society. **c** Circular motion traced by L-shaped microparticles propelling via a heat-induced fluid demixing. The handedness of the orbits depends on the chirality of the particle, i.e., clockwise for the L+ configuration and counterclockwise for the L- configuration. Reprinted with permission from [45]. Copyright 2013, American Physical Society. **d** ICEP of PS microspheres with low-symmetry Au patches in non-cylindrical helical trajectories in an ac electric field. The insets show micrographs and schematics of the particle morphologies corresponding to each motion. Scale bars: 25 μm. Reprinted with permission from [19], through a Creative Commons CC BY license. **e** Induced-charge electro-osmotic (ICEO) flows around patchy microellipsoids shown by superimposing images indicating the motion of tracer particles (0.75 μm in diameter) around immobilized active particles. Scale bars: 10 μm. Reprinted with permission from [20], through a Creative Commons CC BY license.*

2.1 Linear trajectory

The original design of an active colloidal particle was the bimetallic gold (Au) – platinum (Pt) rod developed by Paxton et al. [13,32]. This particle achieves motion via a self-electrophoretic mechanism by consuming hydrogen peroxide (H_2O_2) in the suspending medium. The Au and Pt ends of the rod respectively reduce and oxidize the H_2O_2 causing a flow of protons around the particle that propels it in a linear trajectory. The active motion originates from the ability of the particle to consume fuel by generating a localized gradient of ions along the surface. This seminal work demonstrated the role of chemical heterogeneity and shape asymmetry in driving the self-propulsion of active particles. Catalytic reactions are an effective propulsion mechanism also for spherical particles, with the condition that their surface presents a form of chemical heterogeneity. That was the case of the Pt-coated Janus particle used by the group of Golestanian to demonstrate self-propulsion [14]. The Janus configuration is a type of particle design presenting a hemispherical patch of different composition or surface chemistry. In this case, a spherical polystyrene (PS) microparticle is coated with a thin layer of Pt to induce the localized reduction of H_2O_2 fuel on the metal patch only. The imbalance of solute concentration between the two hemispheres results in linear motion across the gradient. Since these seminal works, the mechanism of catalytic active motion has been upgraded with the addition of light triggers. In fact, Pt can be replaced with photocatalytic metals and metal oxides such as hematite (Fe_2O_3) and titania (TiO_2) to only consume H_2O_2 under blue and UV light exposure, respectively. This is a promising route for several applications, such as environmental remediation, as it impedes unwanted fuel consumption and provides temporal control over self-propulsion. Catalytic active particles are also evolving towards a more biocompatible route by employing enzymatic reactions to drive equivalent motions. This often requires the asymmetric deposition of enzymes on particles to enable diffusiophoresis from concentration gradients produced from localized reactions. One or multiple layers of urease can be immobilized on the gold hemisphere of Janus particles through biotin-streptavidin crosslinking [33]. The formed enzymatic active particle swims by consuming urea, as shown in Fig. 2b.

Alternative mechanisms to induce active linear motion of microparticles have been discovered and implemented over recent years. These include thermophoretic propulsion driven by localized temperature gradients [34] as well as so-called Marangoni swimming that exploits fluid flows generated by a gradient in surface tension [35]. A highly promising class of active particles are those powered by external energy sources such as acoustic [36], electric [19] and magnetic [37] fields. All these tools can induce active motion in linear trajectories by following localized energy gradients and coupling it with fluid fluxes that occur by particle design with encoded symmetry breaking. For example, polystyrene particles half-coated with a layer of Au can self-propel in electric fields via induced-charge electrophoresis (ICEP). As theorized by Squires and Bazant [38] and subsequently demonstrated by Velev and co-workers [15], such metallodielectric PS-Au Janus particle is able to swim in conductive media by translating with the polymer half facing forward. This occurs due to the difference in charge relaxation times of the electrical double layer around the PS and Au halves of the particle. The electric field polarizes the conductive side more strongly than the polymer side, effectively shearing more of the adjacent ions. At low enough field frequencies, the concentration imbalance of ions in the two hemispheres results in fluid flows that causes the self-propulsion of the particle. Note here that the energy source for the particle motion is an external electric field which, as is the case with other fields like magnetic, imposes a global energy gradient that isotropic particle can follow passively. Active motion in such fields is only achieved when the polarized particles follow a

gradient local to their immediate surroundings. For instance, a water-suspended PS sphere in an electric field would move parallel to the field gradient via dielectrophoresis, i.e., a form of passive motion. By contrast, a metallodielectric Janus particle generates and moves along its own gradient that is orthogonal to the global field gradient in active ICEP motion (Fig. 2a). More detailed treatment of particles in alternating current (ac) electric fields can be found in a recent review [39]. Rotating magnetic fields also trigger active motion in corkscrew-like linear trajectories due to the continuous torque-driven reorientation of helically shaped particles [40,41]. The main condition for linear translation of active particles is the breaking of rotational symmetry along one axis, leading to propulsion over the orthogonal axis. This defining design principle applies irrespective of the energy source that powers the active motion.

2.2 Non-linear trajectories

Additional manipulation of particle morphology can enable highly non-trivial dynamics with complex trajectories [22,42–44]. Such non-linear motions arise when the symmetry of the microswimmers is further reduced, leading to translation and rotation of the particles. The coupling of rotational and translational motion gives rise to non-linear trajectories which can be circular in 2D geometries [45,46] and helical in 3D [19,20]. Kummel et al. [45] reported an exemplary result using these design principles to construct diffusiophoretic active particles that move in circular orbits, shown in Fig. 2c. They use photolithography to fabricate *L*-shaped microparticles, which they selectively coat with a thin Au layer on the side of the short segment of the arms of the particles. These are subsequently suspended in water and 2,6-lutidine suspension at the critical concentration and illuminated with light. As the sample cell containing the suspension is irradiated, the Au patch is heated above the critical point, which induces local demixing of the solvent [47]. The ensuing fluid flow causes propulsion along the local concentration gradient and in circular orbits. The Au patch is key in breaking rotational symmetry and lies at the origin of the diffusiophoretic mechanism. Concurrently, the *L* shape of the particle breaks mirror symmetry such that viscous forces act asymmetrically to rotate the particle causing the circular trajectory. The coupling between the translational and rotational contributions in the motion is evidenced by their linear relationship: as the light intensity is increased, the particle's angular velocity scales linearly with its translational velocity.

Our group recently investigated helical propulsion of patchy particles propelled by ICEP [19,20]. Employing PS particles coated with asymmetric Au patches, we observed helical motion arising from the simultaneous translation and circular motion along orthogonal axes. Particles with anisotropic patches were fabricated by glancing angle vapor deposition [48] of Au with partial control over the fractional patch area. While Janus configurations maintain mirror symmetry and propel linearly, the obtained triangular patches are chiral, thus adding the rotational element to the motion. The coupled linear and angular speeds of the active particle scale linearly with the square of the applied electric field, as predicted by ICEP theory [38]. Conversely, the characteristics of the helical trajectory such as radius and pitch are independent of the external field strength, and instead result from the precise geometry of the patch. The handedness and direction of the motion is determined by the initial orientation of the patch with respect to the global field. The rotational diffusion dependence of the direction and handedness are the most prominent elements of stochasticity in an otherwise engineered motion. The combined translational and rotational components of the helical motion enable more efficient scanning of 3D space in analogy with flagellated micro-organisms [49]. This can facilitate motion through complex environment as demonstrated via experiments that showed helical motion allows to

tunnel across a porous membrane with more than twice the probability as linear motion. Further modifications to the patch geometry give rise to even more complex trajectories [42] such as the non-cylindrical helical motions shown in Fig. 2d. By designing active particles with combined anisotropy in shape and patch, one can navigate the intriguing albeit complex parameter space to engineer highly complex motions (Fig. 2e). We recently reported an example of the range of trajectories that a patchy ellipsoidal particle can undergo when propelled via ICEP [20]. Depending on their patch shape, these active ellipsoids can self-propel in linear, circular, and helical trajectories.

2.3 Spinners, rollers, and self-assembled swimmers

Active particles with high degree of asymmetry could transduce the external energy into rotation with a diversity of mechanisms that depend on the particle morphology [43,44,50]. Such dependence of rotation on shape can inform the design of standalone spinners with functional properties and gear-like components of active structures. In addition, rotating particles can also translate when placed at close distance with a substrate [51] (Fig. 3a-b). In this case, the substrate aids in breaking rotational symmetry due to the anisotropy in the hydrodynamics surrounding the particle near and away from the substrate. Thus, it becomes increasingly more important to understand the design rules that guide the active rotation of microparticles. Brooks et al. reported the self-electrophoretic rotation of finned platinum (Pt) microdisks in H_2O_2 solutions (Fig. 3c) [52]. The asymmetry in the shape of these lithographically fabricated particles determined the direction and speed of rotation. Right- and left-handedness in the chiral particle morphology leads respectively to clockwise and counter-clockwise rotation. Increasing fin asymmetry increases angular speed due to the enhanced transport of the H_2O_2 decomposition products near sharp edges. This causes an imbalance in half-reaction between the particles' anodic edges and cathodic faces and the subsequent flow of ions. Surprisingly, the number of fins does not appear to affect the angular speed significantly. Particle systems displaying active rotation provide useful models to understand design rules that correlate symmetry with specific motion trajectory.

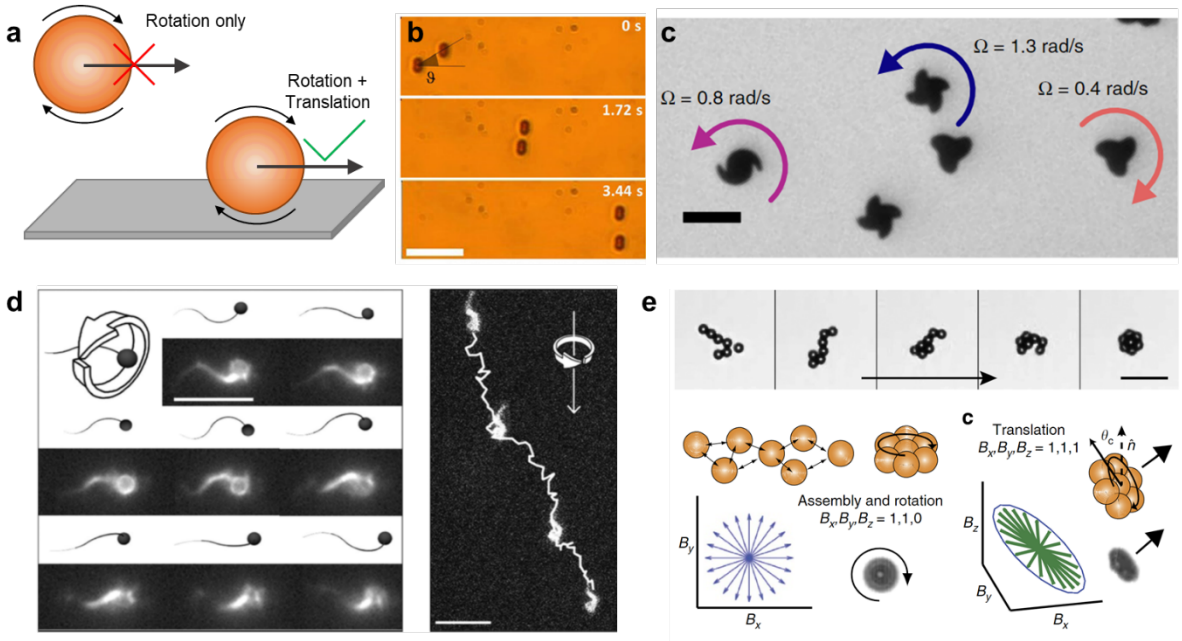

Figure 3: *Active rotation and assembled swimmers.* ***a*** *Schematic of rolling from active rotation near a substrate. A symmetric rotating particle can only translate near a substrate, which acts to break its rotational symmetry causing imbalance in the fluid flows near the substrate and in the bulk fluid.* ***b*** *Hematite microparticles actuated by an out-of-plane rotating magnetic field leading to their translation along a substrate. Scale bar: 10 μm. Reprinted with permission from [51], through a Creative Commons CC BY license.* ***c*** *Multiple designs of platinum microdisks in 'twisted star' shapes with varying number of fins and chirality. Chirality determines handedness of rotation and the angular velocity increases with the degree of anisotropy of the fin. Scale bar: 20 μm. Reprinted with permission from [52], through a Creative Commons CC BY license.* ***d*** *DNA-flagellated magnetic bead propelled by a rotating magnetic field orthogonal to the swimming direction. The schematic and respective fluorescence micrographs show the origin of the propulsion in the non-reciprocal motion of the DNA bundles acting as flagella. Scale bar: 10 μm. Reprinted with permission from [8], through a Creative Commons CC BY license.* ***e*** *Assembly and translation of rotating 'microwheel' composed of seven superparamagnetic microparticles. The schematic showcases the use of two orthogonal rotating magnetic fields in x- and y-direction to induce assembly and a third field in z-direction to tilt the wheel and induce rolling on a substrate. Scale bar: 20 μm. Reprinted with permission from [62], through a Creative Commons CC BY license.*

So far, we have discussed the design of microswimmers composed of an individual particle that contains the symmetry-breaking element for active motion. Another set of active structures are assemblies composed of multiple colloidal elements that only propel when combined. In analogy with many biological materials and swimmers, the individual components cannot achieve motion which is only triggered by their bottom-up assembly to break symmetry [53]. Such structures can either be permanently bound into rigid, static constructs or they can be dynamically connected in a transient state with a reversible mechanism. The choice of permanently bound multicomponent structures is a strategy to confer anisotropy to an active particle in a modular fashion [54–58]. This approach employs bottom-up assembly techniques to construct swimmers capable of achieving non-reciprocal motions, e.g., when actuated by external fields [59,60]. For example, Maier et al. [8] attached artificial flagella to 1 μm iron oxide microspheres and powered their motion with a rotating magnetic field (Fig. 3d). This biomimetic approach inspired by flagellated micro-organisms allows to controllably investigate the role of morphology on the characteristics of motion. They find faster swimming in assemblies made with bundles of tubes with variable stiffness, which achieve high amplitudes while maintaining the ability to bend along the length of the flagellum. Active assemblies can also be dynamic, i.e., they are combined via a transient interaction and lose their structure interchangeably as the interaction is modified [61]. This is often achieved via input of external energy with mechanisms that induce specific assembly and subsequent active motion. One notable example comes from Marr and coworkers through a programmed combination of multiple rotating magnetic fields [62,63], shown in Fig. 3e. First, paramagnetic particles are assembled into a '*microwheel*' shape using two orthogonal fields. This rotationally symmetric structure spins under the influence of the rotating magnetic fields. Subsequent introduction of a third precessing field tilts the structure causing it to roll on the substrate at high speeds. The creative combination of directed assembly and active motion mechanisms can pave the way to the design of complex colloidal devices [64] capable of navigating 3D space and performing multiple functions [65].

3. Environmental effects on trajectory

The motion of active particles is inextricably linked to factors that are external to its design. These may involve the energetic environment underlying its potential and kinetic energy, or the interactions with the suspending fluid, other particles, boundaries, and obstacles. Such environmental effects modify the local energy landscape of active particles with drastic consequences on the trajectories and dynamics of motion. Understanding these aspects is crucial to deploy functional active particles in relevant environments such as biological media or crowded and multiphase mixtures. This is a growing area of inquiry dealing with factors other than the particle design that affect active motion, which we briefly classify into three categories. (1) We first look at direct changes in energetic environment of an active particle due to non-linear field exposure or the use of multiple external fields. (2) Secondly, we consider the effect of suspensions with unusual properties such as non-Newtonian fluids or liquid crystals. (3) We then look at the role of boundaries, interfaces, and environments with complex morphologies.

3.1 Non-linear and multiple external fields

Active particles exposed to an energy source varying in time and space modifies their dynamics as they adapt to continuously changing local energy gradients. Thus, the mode of energy input can become a useful tool to engineer the motion of microswimmers [66]. This has been successfully shown by modulating the trajectory of active particles based on the strength and frequency of externally applied fields [67–69]. Rollers driven by magnetic and electric fields adapt their motion to several regimes associated with different field characteristics. Hematite ellipsoids roll along a surface under the influence of a rotating magnetic field [51], as seen in Fig. 3b. While the particle moment rotates synchronously with the external field, the rolling speed increases linearly with the field frequency. After a critical frequency, the rotating ellipsoids fall out of sync with the external field and enter a ‘back-and-forth’ regime of motion. Rollers actuated by electric fields show analogous changes in dynamics depending on the strength of the applied field. These so-called Quincke rollers are particles suspended in dielectric media, which rotate in response to an imbalanced charging of the particle surface under a dc electric field [70]. Above a certain threshold of field strength, Quincke rollers have been found to enter an oscillatory regime due to transient charge moments within a conductivity gradient that reverse the direction of motion [71].

Frequency of ac electric fields can also revert the direction of ICEP-driven active motion of metallodielectric Janus particles [67,69] (Fig. 4a): this happens at very high frequency (i.e., > 10^5 Hz) due to the proximity of the metal patch with the conductive substrate in a new mode of propulsion termed self-dielectrophoresis [44]. Time-varying external fields increase the versatility of active particles by providing a larger parameter space to access multiple modes of motion within the same system comprising of particles, suspending medium, and external field setup [72,73].

Automation and optimization of active motion using feedback control methods is a promising route to guide active motion. Feedback control methods developed in the group of Bevan [74,75] can be aimed at improving the navigation of active particles whose direction is stochastically determined by the rotational diffusion of the swimmer. Through feedback control mechanisms, active motion can be triggered only when the particle is oriented in the optimal direction (Fig. 4b). Such guided active motion is reminiscent of the optimized chemotaxis of run-and-tumble bacteria like *E. coli*, with the ability to independently scan the environment while

moving towards a target. Combining multiple energy sources is an equivalent step towards improving control of active particles, by exploiting specific advantages of each technique [76–78]. This is often achieved by pairing the motion driven by catalytic activity with an external energy source that is chemically inert such as a magnetic or an acoustic field [79,80]. Such pairings allow particles to move by multiple local gradients in cooperation or competition, spontaneously or programmatically changing directions. For example, bimetallic rods propelled by H_2O_2 can reverse their direction when exposed to ultrasound [81] while photocatalytic spheres with an iron oxide core can be steered with an external magnet [76].

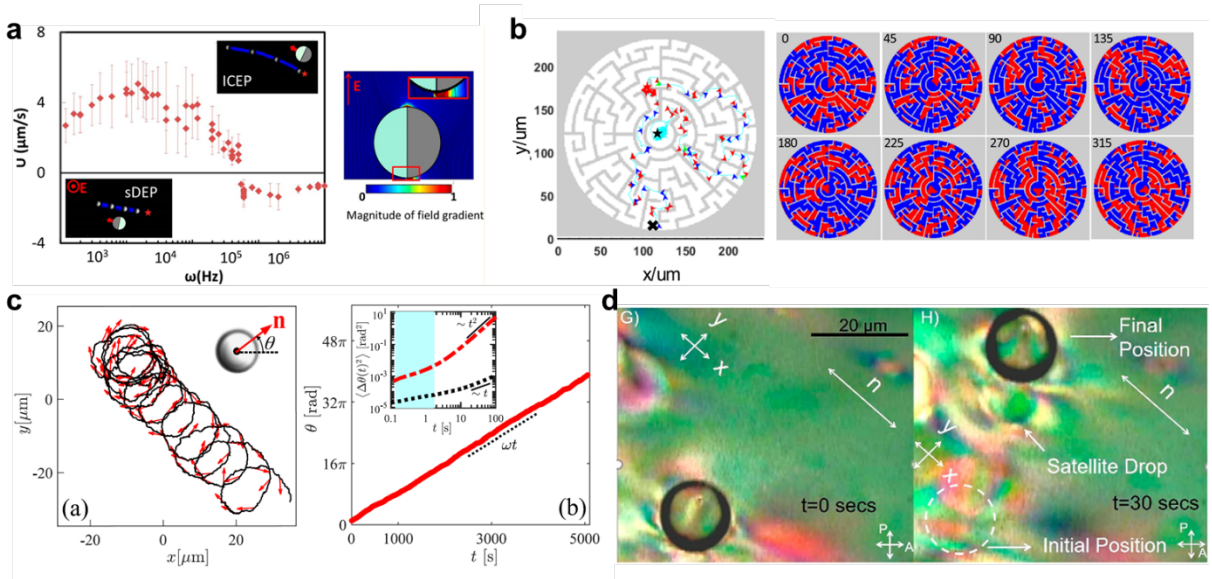

Figure 4: *Complex fields and unusual suspension properties.* ***a*** *Change in translational velocity of gold-coated Janus particle exposed to an out-of-plane ac electric field. At frequencies lower than ~1000 kHz, the active particle moves by induced-charge electrophoresis (ICEP) with the gold patch in the back. At higher frequencies, motion is driven by self-dielectrophoresis with direction inverted and the gold patch in the front. To the right, a COMSOL simulation showing the electric field gradient between substrate and patch, responsible for self-dielectrophoresis. Reprinted with permission from [67]. Copyright 2016 American Chemical Society.* ***b*** *Simulation of optimal navigation of self-propelled rods in a maze based on a feedback control policy that activates motion to promote favorable collisions, alignment with maze features and tracing the shortest path between start and end points. To the right are the optimal control policies used to navigate the maze depending on the orientation of the rod in 45˚ increases. Reprinted with permission from [74]. Copyright 2018 American Chemical Society.* ***c*** *Trajectory of carbon-coated silica sphere in a viscoelastic fluid, driven by local fluid demixing due to heating of the metal patch using a laser and thus showcasing a persistent circular motion. The plot on the right shows the linear behavior of the particle orientation over time. Reprinted with permission from [88]. Copyright 2021 American Physical Society.* ***d*** *Polarized optical micrographs of 5CB droplet translating in DSCG medium due to Marangoni stresses. Motion takes place in the direction perpendicular to the director. Reprinted with permission from [96]. Copyright 2020 American Chemical Society.*

3.2 Unusual suspension properties

Many prospective applications of active particles involve their dispersion in media that are more physico-chemically complex than a continuous aqueous or organic phase. Among others are biological fluids such as mucus, saliva, and blood which are colloidal and macromolecular aqueous mixtures. The presence of nanoparticles and polymers can have dramatic effects on the dynamics of self-phoretic active particles [82–84]. At the single particle level, such crowding

effects have been shown to dampen the dynamics of Pt-coated Janus particles in H_2O_2 solution [25]. Concurrently, excluded volume effects can drive active particles to cluster together transiently with non-linear dependence on depletant concentration [86]. Kalil et al. showed that clustering of catalytic Janus particles increases at low polymer concentrations and, conversely, decreases with increasing depletant fraction. Small amounts of polymer do not slow the dynamics of the active particles and instead contribute to clustering via short-ranged depletion attractions. On the other hand, larger volume fractions of polymer decrease the speed of the active particles, consequently reducing clustering. Polymeric suspensions with viscoelastic properties add a layer of complexity to our understanding of active particle dynamics. This is evident in the divergence of results observed when testing the effect of macromolecular and colloidal crowding on living active matter. For example, the translational velocity of E. coli was found to increase in viscoelastic media while its angular speed decreases, attributed to motion-induced polymer stretching. Patteson et al. [87] showed that the swimming of bacteria exhibits less 'tumbles' and proceeds for longer straight runs in polymer solutions. This appears to be in contrast with the dampening of dynamics described earlier for self-electrophoretic particles. In addition, reports by Narinder et al. [26,88] describe a transition from linear to circular motion of light-activated Janus particles swimming in a viscoelastic fluid. They use an external laser to induce local fluid demixing near the carbon-coated hemisphere of silica particles, finding an increase in rotational diffusion with increasing intensity of the laser (Fig. 4c). Such effects point to a critical difference in the role played by the presence of additional solute in the cases of self-phoretic and wobbling swimmers. Kamdar et al. [89] provided evidence that the enhancement of E. coli motility occurs in concentrated colloidal suspensions as well. They proposed that the bacteria's trajectory is straightened by hydrodynamic torques experienced near solid boundaries. Thus, increasing suspended solute concentration is equivalent to increasing the surface available to rectify bacterial wobbling and increase its speed and the persistence length of its runs.

The unusual physical properties of complex media are not always a hindrance and can instead be exploited for the creative deployment of active particles. A notable example is the ability to bypass the scallop theorem and use reconfigurable swimmers in non-Newtonian fluids [90]. This was recently shown using microswimmers with scallop-inspired shape comprising a hinge and flapping arms. These are actuated by external magnetic fields in shear-thickening and shear-thinning fluids [60]. As noted above, propulsion in low Re fluids generally requires non-reciprocal motions. However, geometrically reciprocal motions can still lead to net translation when the fluid viscosity changes non-linearly with the shear rate. This was shown when the arms of the microscallop open and close at different rates, generating local viscosity gradients in the surrounding non-Newtonian suspension. Liquid crystal suspensions provide analogous in-built anisotropy that can be advantageous to active motion if applied properly [91–95]. Nayani et al. [96] recently reported the transport of an active nematic droplet through a liquid crystal phase, driven by Marangoni flows [97]. The isotropic hexane droplets are diffused and self-propelled preferably in the direction parallel to the director of the surrounding disodium cromoglycate (DSCG) nematic. This is due to the lower hydrodynamic resistance in this direction which has approximately half the effective viscosity of the perpendicular direction in continuous DSCG. Surprisingly, in the case of a 4-cyano-4′-pentylbiphenyl (5CB) nematic droplet swimming in the same continuous liquid crystal, the preferred direction of motion was perpendicular to the director (Fig. 4d). This is attributed to the biasing of the 5CB orientation within the droplet due to van der Waals interaction between liquid crystal phases inside and

outside the droplet. The anisotropy of liquid crystals allows to extract work from otherwise randomly moving active particles. For example, a droplet containing randomly swimming bacteria and emulsified in a thermotropic nematic will move along complex trajectories defined by the director of the liquid crystal environment [92]. Complex media with non-linear properties can be beneficial to active motion as a route to tailoring dynamics and trajectories all the while removing some of the limitations on design such as symmetry-breaking and non-reciprocal motions.

3.3 Interfaces, boundaries, and morphologically complex environments

Realistic environments for the application of active matter can be morphologically diverse, with boundaries that affect the motion of particles. Navigating multiphase environments implies dealing with the specific properties of fluid interfaces such as oil and water. By observing bacteria near such interfaces, Deng et al. identified multiple modes of active motion [98]. They placed *Pseudomonas aeruginosa* near a water-hexadecane interface and found that motile adsorbed bacteria move in curvilinear and 'pirouette' trajectories that are associated with drag asymmetries unique to the interface, seen in Fig. 5a. Concurrently, some bacteria spent most of their time in the aqueous phase, 'visiting' the interface for brief periods of time before escaping it back towards the water. These findings are reflected in recent reports that use synthetic microswimmers near oil-water interfaces. Janus configurations have a particular affinity to adsorb at such interfaces due to their surfactant-like structure, with each half binding preferably to one phase. Fei et al. studied the motion of magnetic Janus particles at the water-decane interface [99]. The PS particles coated with a thin hemispherical layer of nickel moved upon exposure to a precessing magnetic field, in unique zig-zag and curved trajectories otherwise unseen in bulk fluid (Fig. 5b). Jalilvand et al. observed the behavior of Pt-coated silica particles at the water-decalin interface where the Pt cap faces the oil and the silica side faces the water [100]. They found that the induced motility of the particles in the presence of H_2O_2 favors the rotation of the Pt cap away from the interface preventing the adsorption. The analogy between living and active materials is facilitating the understanding of the motion and the binding/unbinding behavior of active particles at interfaces.

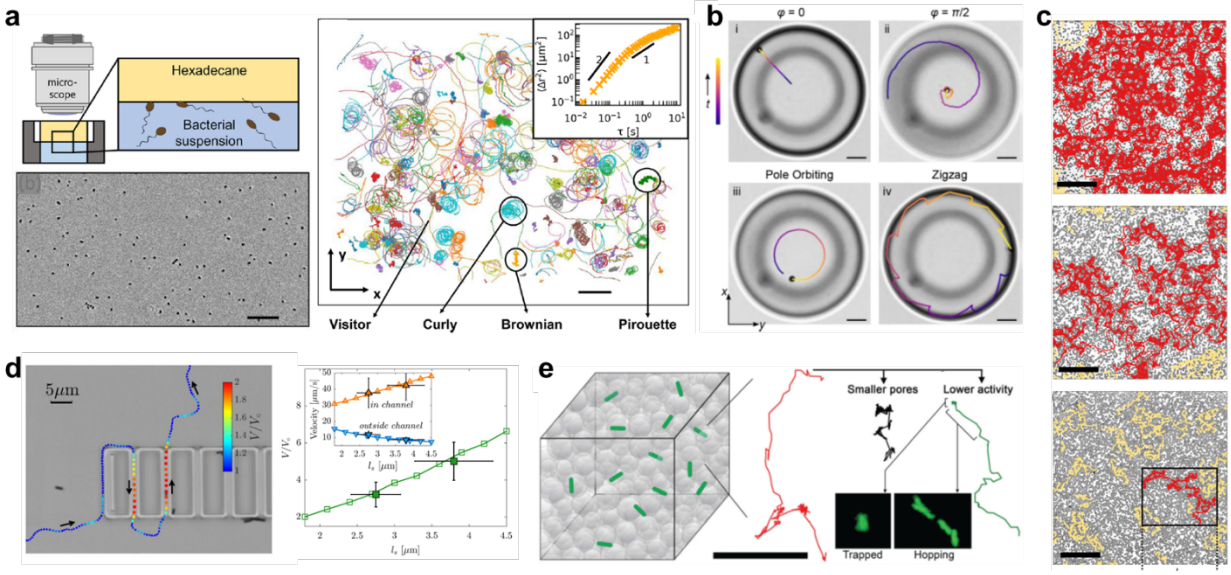

Figure 5: *Interfaces, confinements and obstacles.* ***a*** *Motion of P. aeruginosa at hexadecane-water interface. At the left are a schematic (top) of the experiment and a typical micrograph of the interface*

obtained (bottom). To the right are multiple trajectories of individual bacteria showcasing the motion of visitors as well as Brownian, curly and pirouette motion of interfacially trapped cells. Scale bars: 20 μm. Reprinted with permission from [98]. Copyright 2020 American Chemical Society. ***b*** *Trajectory of magnetic Janus particles at the decane-water interface moving in different trajectories: orbiting the drop equator with a static field (top left); circling towards the drop pole in a rotating field (top right); orbiting the pole (bottom left) and moving in zigzags (bottom right) in a precessing field depending on the relative frequencies of the field components. Scale bar: 15 μm. Reprinted with permission from [99]. Copyright 2020 American Chemical Society.* ***c*** *Trajectories of Quincke rollers in random 2D obstacle lattices. Images show the decrease in percolating trajectories with increasing density of obstacles. Scale bar: 500 μm. Reprinted with permission from [105]. Copyright 2017 American Physical Society.* ***d*** *Trajectory of catalytic bimetallic rod swimmer in lithrographed channel. The color coding indicates the increase in velocity when swimming in the confinement. The plot indicates the increase in the swimmer velocity with its length. Reprinted with permission from [106]. Copyright 2016 American Physical Society.* ***e*** *Dynamics of motile E. coli investigated inside transparent porous environment shown in the schematic (left). The representative trajectory shows the trapping-hopping mode of motion and its dependence on pore size and cell activity. Scale bar: 20 μm. Reprinted with permission from [109]. Copyright 2019 Royal Society of Chemistry.*

The impact of physical boundaries on the motion of active particles has important consequences in applications in crowded and porous environments [101–103]. Most of our understanding of the role played by such boundaries is based on observation of swimmers moving on microstructured substrates [104]. Obstacles in these 2D environments are found to either hamper or enhance the motion of active particles, depending on their density, size, and spatial distribution. Tracking the motion of Quincke rollers in random lithographed post lattices, Morin et al. [105] observed a slowing in dynamics with an increasing density of obstacles. At low obstacle density, the trajectories of the active particles form a single percolating cluster while, at intermediate densities, they form disconnected clusters. Above the percolation threshold of obstacles, the rollers become localized within a void region (Fig. 5c). Boundaries have a different effect when they are geometrically ordered [106,107]. For instance, Liu et al. [106] showed how engineered confinements such as microchannels could increase the velocity of catalytic swimmers by up to 5 times, as shown in Fig. 5d. This enhancement in particle speed is attributed to the screening of the local gradient in electric potential which leads to stronger electro-osmotic flows on the swimmer. Translating findings from 2D to 3D environments has proven challenging due to the difficulties in tracking the dynamics of single active particles. Nevertheless, some initial results have been reported using dense packings of swollen hydrogel particles which are transparent and enable imaging at sufficient resolutions. Battacharjee et al. [108,109] observed the trajectory of E. coli in this type of porous media, finding that the run-and-tumble motility typically observed in bulk fluids is replaced with a ‘hopping-and-trapping’ motion (Fig. 5e). Collisions of the cell with the pore walls truncates runs into ‘hops’ while the rapid tumbling reorientations are replaced with transient trapping as the cell slowly reorients its flagellum until it is able to hop again. Many applications of active particles involve their interaction with soft boundaries and the confinement of active components inside deformable shells is a popular concept of active protocells [110]. Vutukuri et al. showed that self-propelling particles, embedded inside giant unilamellar vesicles, generate shape-morphing forces that deform the membrane into morphologies otherwise inaccessible in equilibrium conditions [111].

4. Applications

Reported applications of active particles span many areas, with some highly creative demonstrations [112,113], such as energy generation [114] and microscopic welding [115]. However, the most promising deployments of active particles are in the areas of biomedical engineering and environmental remediation. Both of these are highly amenable to exploiting the motion of micro- and nanoscopic particles in complex and confined suspensions. In such applications, active particles are envisioned and utilized as micro- and nanorobotic devices with the ability to deliver, collect and organize cargo at molecular and colloidal length scales. In this section, we report recent applications of active particles, achieved with a degree of control over trajectories and interactions with the immediate environment.

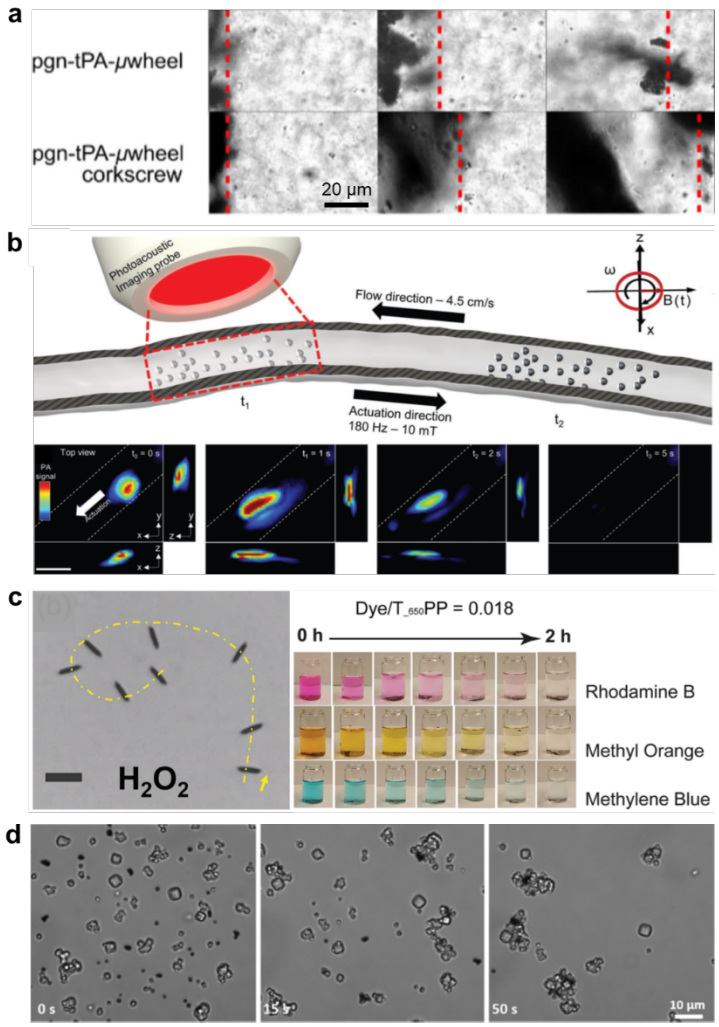

Figure 6: *Biomedical and environmental applications of active particles.* ***a*** *Timelapse of lysis of plasma clots using magnetic active particles. The dashed red line indicates the fibrin front and the increase in penetration of the particles when translated linearly (top) and with corkscrew-type rotation (bottom). Reprinted with permission from [119]. Copyright 2021 Wiley.* ***b*** *Schematic (top) and timelapse example (bottom) of photoacoustic imaging of active particles motion inside of tubing. Magnetically actuated rollers move against flow direction and their flow is visualized based on the contrast in photoacoustic signal. Scale bar: 3 mm. Reprinted with permission from [120], through a Creative Commons CC BY license.* ***c*** *Motion of tubular particle composed of co-axial TiO_2 and bimetallic PtPd nanotubes in 5% H_2O_2 solution*

(left) and corresponding degradation of organic dyes (right). Scale bar: 30 μm. Reprinted with permission from [122]. Copyright 2016 Wiley. ***d*** *Removal of debris from washing powder using TiO_2-based Janus particles half-coated with Au, suspended in 0.1% H_2O_2. Phoretic attraction is triggered by UV light to form removable aggregates to purify the liquid. Reprinted with permission from [124]. Copyright 2019 American Chemical Society.*

4.1 Biomedical applications

Manipulation of active particles in the human body offers the possibility of minimally invasive medical procedures, with applications in the targeted delivery of drugs or remote microsurgery [116–118]. The preferred actuation mechanisms are acoustic and magnetic fields due to their contactless applicability and chemical inertness. Disharoon et al. recently used superparamagnetic beads to target and lyse plasma clots [119]. They coated magnetic microparticles with mesoporous silica material and loaded these with plasminogen and tissue plasminogen activator which are responsible for fibrinolysis. These particles are translated towards the clots and rotated using external magnetic fields. This active method enables the lysis of plasma clots at rates 40-fold higher than the biochemical limits and in areas that are inaccessible by mechanical thrombectomy (Fig. 6A). One key challenge in using active particles for *in vivo* applications is the reliable localization and visualization within the body. Different techniques are available to detect the position of particles, such as magnetic resonance or photo-acoustic imaging. These require sufficient contrast in magnetic and optical properties between the material of interest and the background. Magnetic Janus particles are being investigated for their potential localization using existing medical imaging equipment, with actuation using external magnetic fields. Materials such as the $L1_0$ phase of FePt alloys have the right properties for both remote actuation and localization. Bozuyuk et al. demonstrated the endovascular navigation of silica microparticles coated with a thin layer of co-deposited Fe and Pt [120]. These rollers successfully moved against fluid flows and can be visualized with both MRI and photo-acoustic imaging (Fig. 6B). The ability to visualize motion, combined with the loading and triggered release of drugs from Janus particles [121], represents significant advancement in the use of active particles for drug delivery.

4.2 Environmental applications

Self-propelling particles hold promise for several applications in environmental remediation, such as the removal of heavy metal ions, organic pollutants, and particulate matter. Light-activated swimmers are of particular interest since environmental use of UV light is already very common for the purification of water by disrupting regulatory mechanisms of DNA in microorganisms. Thus, photo-activated motile particles are heavily investigated to increase the capability of water decontamination and purification by light irradiation. While photocatalytic active swimmers such as TiO_2-based self-propelled particles produce hydroxyl radicals which decompose organic molecules. Mushtaq et al. fabricated a versatile TiO_2-PtPd-Ni nanotube via template-assisted electrochemical synthesis [122]. These particles are activated not only by UV light, but also by visible and natural sunlight via decomposition of H_2O_2. This is due to the coupling of TiO_2 with PtPd, which modifies its bandgap and improves photocatalytic activity. These active particles were able to degrade model organic pollutants such as rhodamine B with 100% removal efficiency under natural sunlight (Fig. 6C). The decontaminant role of active particles may be exploited for the removal of other suspended particulate matter. This is relevant to the emergence of microplastics as a pollutant in many aqueous environments [123], which can be removed with photocatalytic or magnetic interactions (Fig. 6D) [124]. The major

challenge to the use of active particles for environmental applications is to develop fuel-free alternatives to the current methodologies [125].

5. Conclusion

The past few years have seen a marked improvement in our understanding of the factors that guide the trajectory of active particles. We described some key particle design principles that can direct linearity and non-linearity of active motion, yet much remains to be explored. Fabrication of particles that allow experimental investigation of such morphology-motion relationship is challenging. Therefore, modular approaches that seek to construct microswimmers from individual units is a promising avenue. Assembly may allow access to designing active structures that move not only by non-reciprocal deformation but also by self-phoretic mechanisms with complex trajectories. Transient self-assembled swimmers bring us closer to the concept of colloidal machines and synthetic cells long sought after.

Studying the parameters that are external to particle design has proven very fruitful. Many factors such as viscoelasticity and porosity of the surrounding environment appear to play highly non-trivial roles in the modality and trajectory of motion. Nevertheless, the breadth of this parameter space is such that it is difficult to decouple effects in biologically realistic environments that are complex both in morphology and charge distribution [25]. We showed examples of converging concepts from motile micro-organisms and active particles. However, there is still an ongoing effort to connect some diverging results obtained with living and synthetic swimmers [126].

Combining the progress made in both the areas of internal and external factors driving specific motion, will be key to the technological success of active particles. In fact, the recent fundamental advancements have gone hand-in-hand with a general increase in reported lab-scale applications. Such progress makes it possible to tailor particle design for use in more realistic settings with awareness of how such environments will affect motion. Porous environments may be better navigated by helically moving particles, and, at the same time, interfacial trapping of particles may be used to guide otherwise impossible motions [127]. An improved understanding of swimmer-environment interactions also enables to apply active motion to trigger phenomena such as cargo release [128–130] and localized polymerization [131].

While still far from the sophistication of nature’s motility, active particles are becoming more diverse, efficient, and versatile. As we uncover the principles and mechanisms that determine individual particle trajectory, we are still learning to translate them in controlling collective swimming. From the motion of many individual active particles, often emerges a swarm-like behavior with somewhat independent dynamics. Thus, a major future challenge will be to gain control over the trajectory of active swarms composed of individual active particles. Progressing towards hierarchical active matter, we envision the decoupling of the motion of multiple collectives moving independently to perform multiple functions simultaneously.

Declaration of competing interests

The authors declare no competing financial interests.

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8* Example of the role of biomimetic design of active particles in elucidating the mechanical role of flagella in motile bacteria.

19* Demonstration of the role of patch morphology in driving complex 3D motion via ICEP.

42* Analytical investigation of the role of particle symmetry in motion driven by ICEP.

44* Key demonstration of the variable modes of activation of colloidal spinners and their assembly, via AC electric fields.

65* Thought-provoking perspective on the rationale for design of versatile microbots as opposed to single-task particles.

75* Model development for controling the trajectory of active particles using artificial intelligence.

76* Example of the versatility of hybrid active colloids that can be manipulated with light and magnetic fields.

89* Clarification of the mechanism of bacterial swimming rectification in colloidal suspensions

95* Initial study of the correlation of active motion trajectory with the properties of nematic liquid crystals.

101* Demonstration of the potential in exploiting geometric constrain to steer and guide trajectories for active particles.

108* Observation of unique mode of bacterial transport in porous media, via hopping and trapping.

111* An experimental demonstration of dynamic shape-morphing ability of vesicles due to active motion of particles enveloped within the deformable giant unilemellar vesicle.

121* Report of roller active particles able to flow upstream and target cancer cells, delivering drug molecules.