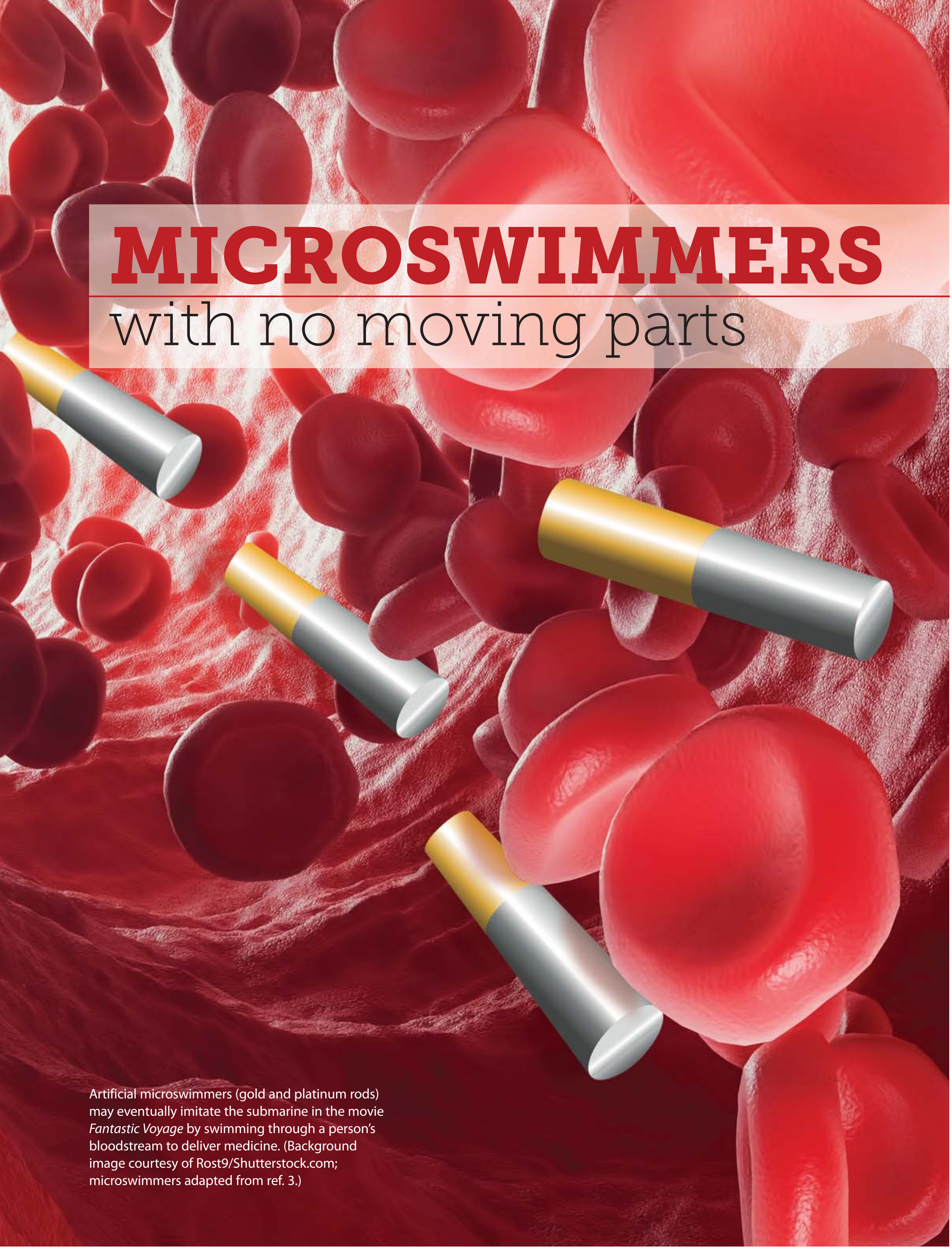
MICROSWIMMERS

with no moving parts



Artificial microswimmers (gold and platinum rods) may eventually imitate the submarine in the movie *Fantastic Voyage* by swimming through a person's bloodstream to deliver medicine. (Background image courtesy of Rost9/Shutterstock.com; microswimmers adapted from ref. 3.)

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Jeffrey Moran and Jonathan Posner

Microscopic self-propelled particles could one day be used to clean up wastewater or deliver drugs in the body.

Swimming is a ubiquitous task performed by a broad range of creatures. Large animals, like whales or humans, generally swim by imparting backward momentum to the fluid, which results in forward motion of the animal through momentum conservation. The same strategy works even for relatively small animals, such as fish and tadpoles.

Microscopic organisms, such as bacteria, that are one-tenth to one-hundredth the size of tadpoles must also swim to find nutrients and avoid predators. However, the macroscale strategy of imparting momentum to the fluid is ineffective for them because of the low Reynolds number associated with the microscopic regime, in which viscous dissipation overwhelms the effects of fluid momentum. Picture a human trying to swim in molasses: Any effort to push fluid backward and thus generate forward motion would be stifled by the fluid's viscosity. Bacteria and other microorganisms face an analogous situation because they are so small that even water's viscosity is enough to overwhelm their negligible inertia. Despite that challenge, evolution has equipped microorganisms with a range of efficient swimming strategies that have enabled microscale swimming for millions of years.

Since the turn of the 21st century, scientists and engineers have begun to develop artificial analogues to natural microswimmers. Artificial microswimmers are colloidal particles, comparable in size to bacteria and typically 1–10 μm, that can propel themselves through liquids. Whereas naturally occurring microswimmers move by actuating appendages such as cilia or flagella, artificial microswimmers usually have no moving parts and instead generate propulsive forces by interacting with their environment through chemical reactions. The particles have been designed to perform complex behaviors and tasks, so they are being actively researched for various important practical applications from water treatment to drug delivery.

People have long been fascinated by the idea of microscopic self-propelled vehicles moving inside the body. In his 1959 lecture "There's plenty of room at the bottom," Richard Feynman spoke of one day being able to "swallow the surgeon." He challenged the scientific community to design a tiny motor that moves on its own and fits inside a cube 1/64 of an inch on each side. Nonscientists have been similarly enamored with the idea. The 1966 science fiction film *Fantastic Voyage* portrays a group of scientists who shrink themselves and their submarine, *Proteus*, to the size of a cell and are able to enter the bloodstream of a colleague and remove a blood clot from his brain.

Nanotechnology took several decades to catch up to that vision; not until the early 2000s were researchers able to successfully create artificial microswimmers. Through clever micro- and nanofabrication strategies, approximately a dozen designs for self-propelled particles have now been demonstrated.

Motion at the microscale

In 1956 biochemist Peter Mitchell proposed that some microorganisms might propel themselves through nonmechanical means without using any moving parts. He postulated that if a bacterium could pump ions across its membrane asymmetrically, such that ions would be pumped out of the cell on one end and back in at the other, an electrical circuit would form,[1] as shown in figure 1. Ions would flow from the rear of the bacterium's body to the front and in the opposite direction through the fluid adjacent to the surface of its membrane. In that way the organism would be propelled forward "much as a tread is used for the locomotion of a tank," as Mitchell put it. Using asymmetric ion pumping, an organism could, at least in principle, swim without flagella.

Phoresis is the motion of cells or particles due to a gradient of a specific quantity, such as electric potential (electrophoresis), solute concentration (diffusiophoresis), temperature (thermophoresis), or another quantity. It is a common mode of transport at the micrometer scale. Phoretic self-propulsion occurs when a particle or cell generates a motion-inducing gradient on its own and then moves in response to it. Mitchell dubbed his proposed bacterial swimming mechanism "self-electrophoresis."

Since Mitchell's proposal, several scientists have sought to determine whether any microorganisms move by self-electrophoresis. In the 1980s a marine cyanobacterium called *Synechococcus* was discovered to move in seawater despite having no visible appendages. Some scientists hypothesized that *Synechococcus* swam by self-electrophoresis; however, experimental measurements later showed that the organism lacks an appreciable surface charge and therefore cannot move under the influence of an electric field, either external or self-generated. The active propulsion mechanism driving *Synechococcus*'s motion remains an open question.

Although no conclusive evidence has been found—at least, none yet—that any microorganisms use self-electrophoretic propulsion mechanisms, self-propulsion can and does occur through electrophoresis, diffusiophoresis, and thermophoresis in engineered systems. Artificial microswimmers are microscale particles that generate chemical, electric, or thermal gradients in their vicinity through chemical reactions on their surfaces. Asymmetry in at least one quantity—such as electric potential, temperature, or concentration—is required to generate motion, and that asymmetry is typically generated through a nonuniform patterning of the particle surfaces. Most phoretic artificial microswimmers have opposite-facing sides with different chemical properties and are referred to as Janus particles, after the two-faced Roman god of beginnings and endings (see the Quick Study by Steve Granick, Shan Jiang, and Qian Chen, PHYSICS TODAY, July 2009, page 68).

Motion of microbatteries

Although it has yet to be definitively observed in nature, self-electrophoresis was the first mechanism realized in artificial microswimmers. In 2004 Walter Paxton, Ayusman Sen, Thomas Mallouk, and their colleagues at the University of Pennsylvania introduced autophoretic bimetallic rods.[2] The cylindrical particles were 2 μm long and 200 nm in diameter and consisted of two connected metal segments. Typically, the metals in such swimmers are gold and platinum, although other metal combinations can also be used. Each rod acts like a tiny battery, with platinum acting as the anode and gold as the cathode. When the bimetallic rods are placed in aqueous hydrogen peroxide solutions, electrochemical reactions on each surface create an electron current through the rod from the anode to the cathode and a complementary ionic current in the electrolyte. The oxidation reaction on the anode injects positive ions into the solution, and the reduction reaction on the cathode consumes positive ions. The reaction scheme is thus similar to the one Mitchell proposed, which is depicted in figure 1.

The reactions drive the locomotion of the rods. The products from those reactions, shown in figure 2, cause the fluid surrounding the Pt end to be positively charged and the fluid surrounding the Au end to be negatively charged, which generates an electric dipole and an associated electric field in the fluid.[3] The particle is ultimately propelled by the self-generated electric field because the field exerts a force on the positive charges in the electric double layer that surrounds the rods, and on the negatively charged rod itself. The positively charged fluid flows backward toward the cathode and the negatively charged rod moves with the anode facing forward. Like batteries, the rods convert chemical energy stored in the H_2O_2 molecules into electrical energy; unlike batteries, they then convert that electrical energy into kinetic energy.

When it comes to creating a self-electrophoretic particle, there is nothing special about platinum or gold. Rods fabricated from other metal pairs also self-propel in H_2O_2, and in each case the metal with less affinity for electrons is the one that faces forward during the motion.[4] For the gold–platinum rods, the platinum end always leads. Also, a greater difference in electron affinity between the two metals results in a greater driving force for the electron transfer and a faster swimming speed. Thus, electrochemical measurements can predict the direction and speed of a bimetallic rod's self-propelled motion.

Self-electrophoresis has been observed in other engineered systems using the same physical principles but different sources of energy. For example, the same mechanism underlies the self-propulsion of carbon fibers (roughly 1 cm long and 7 μm in diameter) in glucose solutions.[5] The anode end is coated with a

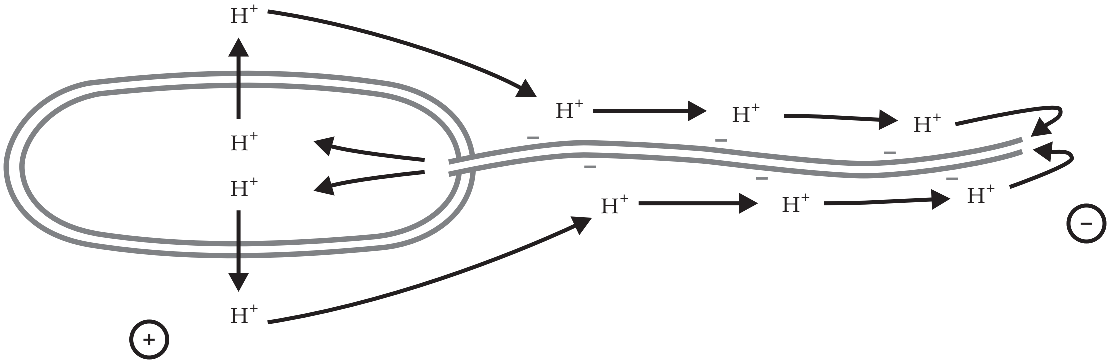

FIGURE 1. AN ELECTROKINETIC MECHANISM by which a bacterium could swim was proposed by biochemist Peter Mitchell. By pumping positive hydrogen ions out of its body and consuming them at its tail, the organism generates a stream of ions that moves alongside the outside of its body and in the opposite direction inside its body through the flagellum, completing an electrical circuit. The self-propelled motion occurs from right to left due to the negatively charged outer surface of the flagellum. (Adapted from ref. 1.)

catalyst that breaks down glucose. Electrons conduct through the fiber, and hydrogen ions migrate from the anode to the cathode end, which is coated with a different catalyst. At the cathode, oxygen combines with hydrogen ions and electrons to form water. If the electrical connection between the two ends is interrupted, the motion stops, which lends support for the self-electrophoresis mechanism.

Water and UV light can also be used as energy sources for self-electrophoresis. Titanium dioxide microspheres with one half coated in gold propel themselves in pure water under UV, with the TiO_2 end facing forward.[6] When TiO_2 is irradiated with UV, electron–hole separation occurs. The electrons that move to the TiO_2 conduction band have a high energy relative to what they would have in gold, so an electron current flows from the TiO_2 to the gold. Meanwhile, electrochemical charge transfer reactions occur on the TiO_2 and gold surfaces and produce a self-generated electric field that drives self-propulsion.

Artificial microswimmers, including self-electrophoretic ones, can be engineered to swim at greater than 1 mm—more than 100 body lengths—per second. By comparison, a Ferrari 488 sportscar traveling at its top speed of 205 mph moves only at about 20 body lengths per second. The speed of the particles generally increases with fuel concentration because the reactions driving the motion proceed faster. At high enough fuel concentrations, the velocity begins to level off, for two main reasons: First, there are a finite number of available sites on the metal surfaces for the reactions to take place; second, most of the reactions produce bubbles of molecular oxygen or hydrogen that can occlude the particle's reactive surface at fast reaction rates.

One drawback of self-electrophoretic microswimmers is that they must be operated in aqueous systems with very low salinity. Experimentally, even small concentrations of dissolved salts will substantially decrease the swimmers' velocity. Computer simulations at various salinities show that dissolved salts reduce the magnitude of the self-generated electric field, the reaction rate, and the particle surface charge. The particle velocity is roughly inversely proportional to the electrical conductivity of the solution.[7,8] Above a relatively modest salt concentration of 1 mM, the motion of platinum–gold rods is nearly indistinguishable from passive Brownian motion. Still, self-electrophoretic microswimmers are being considered for some low-salinity applications, such as motion-based contaminant sensing and removal in water samples.

A more neutral power source

Phoretic motion requires a gradient, but the gradient need not be in electric potential. Diffusiophoresis occurs when an object moves in response to a gradient in chemical concentration. When the object generates the gradient itself, the motion becomes self-diffusiophoretic. Unlike electrophoresis, diffusiophoresis can occur in gradients of neutral and charged solutes; however, most studies have focused on neutral solutes. Diffusiophoresis is thought to occur naturally in the locomotion of the bacterium *Listeria monocytogenes* and of some cells that crawl by using asymmetric polymerization of the protein actin.[9]

Self-diffusiophoresis requires that the swimmer induce a nonuniform concentration of a solute, typically through asymmetric surface reactions, as depicted in figure 3. The asymmetry in the solute concentration around the particle leads to imbalanced interaction forces between the solute and the particle. Common interaction forces between the solute and the particle stem from steric repulsion, hydration forces, van der Waals forces, and electrostatics. Those forces tend to be relatively weak, so self-diffusiophoretic swimmers typically move significantly more slowly than self-electrophoretic particles.

Swimming by self-diffusiophoresis was first proposed in 2005, around the same time research was beginning on the motion of self-electrophoretic platinum–gold rods. Ramin Golestanian and coworkers at the University of Sheffield proposed that a spherical particle could generate a neutral solute gradient if a single reactive site on its surface continually produced uncharged product particles or molecules. They calculated the resulting swimming speed as a function of the reaction rate.[10] Soon after the mechanism was proposed, a candidate system for self-diffusiophoresis was identified that used insulating microspheres, half-coated in platinum, that self-propelled with the insulating hemisphere forward and platinum-coated end backward in hydrogen peroxide solutions.[11] The dominant interaction mechanism for those swimmers is steric repulsion between the particle and the oxygen molecules generated in the peroxide decomposition reaction on the platinum surface. Since the particle–solute interaction is repulsive, the particle moves toward lower solute concentration.

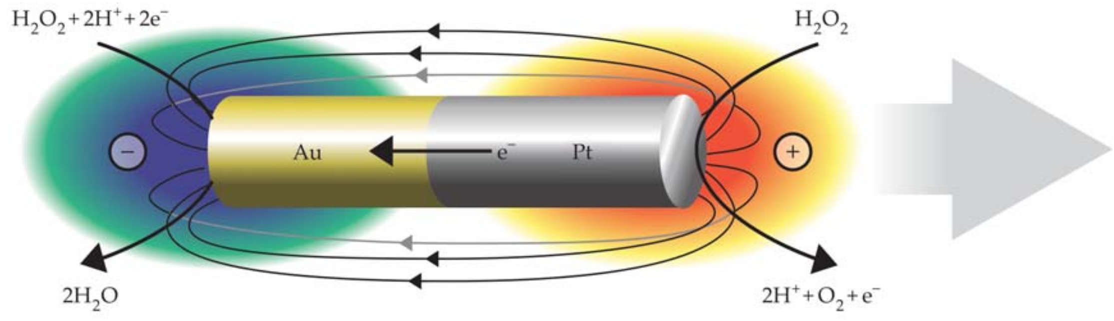

FIGURE 2. A PLATINUM–GOLD ROD UNDERGOES SELF-ELECTROPHORESIS in a hydrogen peroxide solution. The rod functions as a short-circuited electrochemical cell, with platinum acting as the anode and gold as the cathode. Oxidation and reduction reactions on the Pt and Au surfaces, respectively, lead to the establishment of dipolar charge clouds. The clouds form an electrical dipole, and an associated electric field, which then exerts a propulsive force on both the positively charged fluid surrounding the rod and the negatively charged rod itself. The result is self-generated motion of the rod from left to right. (Adapted from ref. 3.)

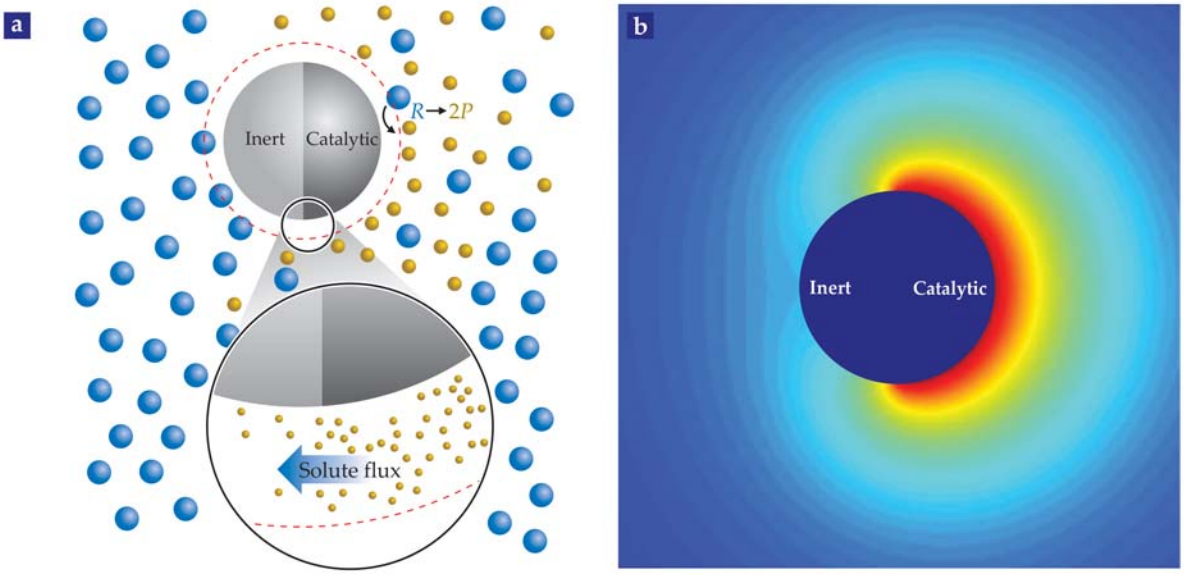

FIGURE 3. SELF-DIFFUSIOPHORESIS can propel particles in neutral solute gradients. **(a)** A Janus particle with two distinct sides is immersed in a solution containing a solute *R* (blue dots), which reacts when it contacts the catalytic half of the spherical particle, forming two product molecules *P* (orange dots). If the particle and the product molecules experience a mutually repulsive interaction force, the particle feels a net force from right to left. (Adapted from ref. 3.) **(b)** Solute molecules are asymmetrically distributed around a swimming particle. The solute molecules simply dock themselves on the catalytic hemisphere, and the repulsive interactions between the solute molecules and the inert side of the particle cause the particle to move from left to right. The colors represent the solute concentration, increasing from low (red) near the particle to high (blue) farther away. (Adapted from ref. 3 and U. M. Córdova-Figueroa, J. F. Brady, *Phys. Rev. Lett.* **100**, 158303, 2008.)

Other types of active particles have since been developed that appear more definitively to move by self-diffusiophoresis. One system uses gold–silicon dioxide Janus spheres with a catalyst on the SiO_2 hemisphere.[12] When the swimmer is immersed in a solution of monomer, the catalyst causes the monomer to form polymer chains on the catalytic SiO_2 surface and, in turn, decreases the local concentration of monomer on that side of the particle. The asymmetric polymerization results in a concentration gradient of monomer surrounding the swimmer, which causes the swimmer to propel away from higher concentration toward the polymerized end, again through repulsive steric interactions.

Some like it hot

Self-thermophoresis is the autonomous analogue to thermophoresis, the motion of particles or cells due to temperature gradients. In gases, thermophoresis is relatively straightforward. When a particle is immersed in a gas with a temperature gradient, the high-temperature gas molecules on the hotter side of the particle collide with the particle surface more frequently and with greater momentum, so that they propel the object in the direction of decreasing temperature.

In a liquid, particles typically move toward lower temperatures, as in the gaseous case; however, changing the environment by increasing the overall average temperature or by adding salt or a surfactant can cause particles to go toward warmer temperatures. The exact mechanism of thermophoresis in liquids is a topic of active research. Like other processes that occur spontaneously, it must be driven by a decrease in the Gibbs free energy of the system. Two main contributions to that energy of suspended colloidal particles are hydration forces and ionic shielding at the surface. At low temperatures and high electrolyte concentrations, the hydration entropy dominates, and motion toward warmer regions becomes thermodynamically favorable. At room temperature and in less-concentrated electrolytes, ionic shielding plays a greater role, and the particle tends to move toward lower temperatures.[13]

Like other forms of phoretic self-propulsion, self-thermophoresis typically occurs with an asymmetrically patterned colloidal particle. The prototypical self-thermophoretic system is a gold–insulator Janus particle driven by IR laser light. The particles are fabricated by taking silica or polystyrene microspheres and sputter-coating gold on half of the surface of each sphere.[14] The metallic side preferentially absorbs the IR radiation and becomes warmer than the insulating side; the temperature difference generates a temperature gradient in the surrounding solution and causes the particle to move.

The speed of laser-powered self-thermophoretic microswimmers increases with the intensity of the IR light and stops when the light is turned off. In that way, stop-and-go motion is achievable in pure water. Another advantage to not using chemical fuel is that the motion can essentially continue indefinitely as long as IR light is provided.

Complex and collective behaviors

Under a microscope, swimming particles behave similarly to microorganisms, such as motile bacteria. They change their swimming direction at regular intervals and interact with both solid surfaces and each other. Also, like some bacteria, artificial swimming particles respond to nonuniform solute concentrations, temperatures, and electromagnetic fields. Because of those apparent similarities, researchers have put significant effort into studying the complex and collective motion of artificial swimmers.

All micron-scale suspended particles undergo Brownian motion, which causes them to translate and rotate randomly due to thermal fluctuations. Self-propelled particles undergo

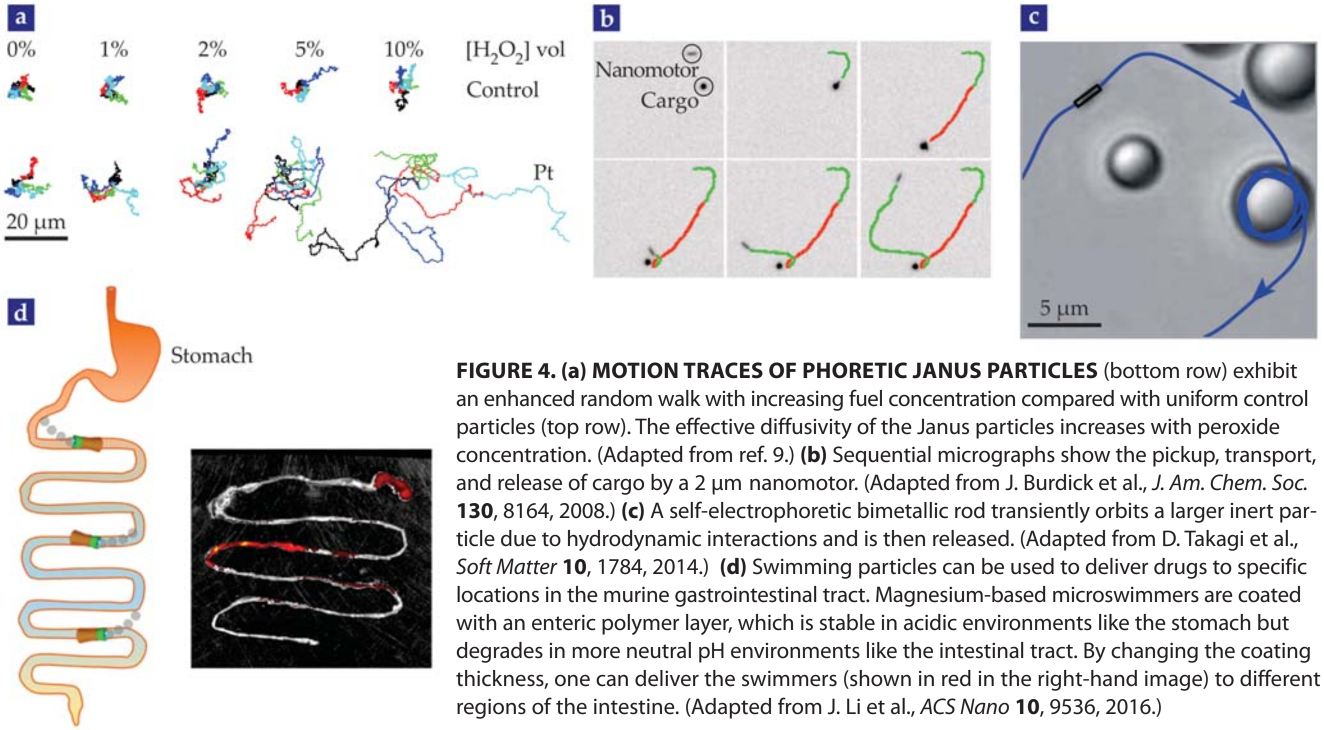

FIGURE 4. (a) MOTION TRACES OF PHORETIC JANUS PARTICLES (bottom row) exhibit an enhanced random walk with increasing fuel concentration compared with uniform control particles (top row). The effective diffusivity of the Janus particles increases with peroxide concentration. (Adapted from ref. 9.) **(b)** Sequential micrographs show the pickup, transport, and release of cargo by a 2 μm nanomotor. (Adapted from J. Burdick et al., *J. Am. Chem. Soc.* **130**, 8164, 2008.) **(c)** A self-electrophoretic bimetallic rod transiently orbits a larger inert particle due to hydrodynamic interactions and is then released. (Adapted from D. Takagi et al., *Soft Matter* **10**, 1784, 2014.) **(d)** Swimming particles can be used to deliver drugs to specific locations in the murine gastrointestinal tract. Magnesium-based microswimmers are coated with an enteric polymer layer, which is stable in acidic environments like the stomach but degrades in more neutral pH environments like the intestinal tract. By changing the coating thickness, one can deliver the swimmers (shown in red in the right-hand image) to different regions of the intestine. (Adapted from J. Li et al., *ACS Nano* **10**, 9536, 2016.)

an additional linear motion that is superimposed onto their translational and rotational Brownian motion. That superposition causes swimming particles to move persistently, but in randomly varying directions, exploring a much larger area than a freely diffusing particle would. Comparing the trajectories for Brownian particles and swimmers in figure 4a shows that the swimmers have a larger diffusivity. Over a 10-minute period, a passive particle may diffuse over a region of 35 μm^2, whereas a similar swimming particle may explore a region greater than 1 mm^2. That capability could be used for, say, delivering a drug to one region of the body with the goal of broadly distributing it.

Although most reports suggest that artificial microswimmers move in relatively straight lines with random perturbations, the particles have also demonstrated the ability to swim in tight circular patterns. They swim in circles due to an asymmetry, either unintentional or deliberate, in their geometry. As the particle swims, the asymmetry causes a larger viscous drag on one side versus the other, so the particle turns persistently in one direction. For short times circle-swimmers exhibit the large diffusivity of a swimmer, but for long times they show a reduced effective diffusivity because they swim back almost to their original position after a full revolution.

The swimming direction of a particle can be controlled using externally applied fields. A swimmer with an embedded magnetic segment will align itself in an external magnetic field. The microswimmer still generates a propulsive force on its own but moves in a direction aligned with the magnetic field. By combining those two mechanisms, a swimmer can be directed into predetermined patterns like microfluidic channels, reminiscent of when *Proteus* travels through Jan Benes's capillaries in *Fantastic Voyage*.

The swimmer could also use the same magnetic segment to pick up a paramagnetic particle and drop it off in another location, as demonstrated in several studies and shown in figure 4b. Like ants, artificial microswimmers can carry cargo more than 30 times their size. Because the swimmers are at low Reynolds number, it's the size, not the weight, of the cargo that determines its viscous drag. The cargo delivery capability has been used for purposes such as sorting cells or delivering drug-impregnated polymer particles.

Swimmers interact with surfaces and other particles through hydrodynamic, electrostatic, and phoretic forces. Long-range hydrodynamic forces can drive swimmers toward microscopic features, causing them to move along the boundaries of submicrometer-scale ledges and in intermittent circles around larger, immobilized spheres, like the rod in figure 4c. The chemical and electrical fields generated by swimmers have shorter ranges, so they can affect only interactions with nearby surfaces or particles. Interparticle interactions can be attractive, causing particles to cluster, or repulsive, causing particles to disperse, and the type of interaction can depend on the relative alignment of the particles and their associated fields.

Experiments have shown that autophoretic particles can exhibit taxis, which refers to the ability of an organism to sense an external stimulus, often light or the presence of a nutrient, and to move toward or away from it. Like some fish, engineered swimmers can exhibit positive rheotaxis, migrating against the current in a fluid flow.[15] Their upstream swimming originates from a viscous shear-flow-induced torque that persistently aligns the particles' swimming direction against the flow. Phoretic particles have also exhibited positive gravitaxis, upward swimming against the direction of gravity.[16] In that case, the particles are either geometrically or chemically anisotropic. In both cases the asymmetry leads to the displacement of the center of mass and results in a torque that aligns the particle with the gravitational field. Such behaviors could be used to design particles that can swim upstream in the circulatory

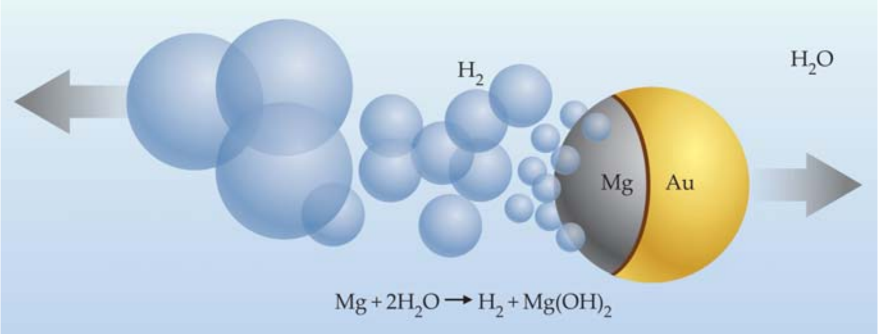

FIGURE 5. BUBBLE-PROPELLED MICROSWIMMERS generate hydrogen gas bubbles to push themselves forward. A sacrificial metal—in this case, magnesium—on one side of the particle reacts with water to form the bubbles. The gold on the other side of the particle does not react with the water. The particle experiences a recoil force on its left side from the separating bubbles and is pushed from left to right. (Adapted from ref. 18.)

system to deliver drugs or to the surface of a body of water to remediate toxic spills.

Applications in medicine and the environment

Artificial microswimmers are now being developed for biomedical applications. The *in vivo* environment poses challenges because it is often less hospitable to the mechanisms that drive swimmers' motion than pristine *in vitro* environments. Many swimmers require fuel, such as hydrogen peroxide, which would not be well tolerated by most animals at the concentrations needed for swimming. Additionally, *in vivo* conditions are typically salty—the average salt concentration in biological media is roughly 0.1 M—which precludes self-electrophoresis as a swimming mechanism.

An alternative mechanism is needed to propel microswimmers in those environments. Zinc, aluminum, and magnesium microswimmers dissolve in weakly acidic aqueous conditions and generate hydrogen gas bubbles that propel swimmers forward, as shown in figure 5. They are not phoretic, but instead are propelled forward by recoil forces generated by separating bubbles. Unlike phoretic microswimmers, bubble-propelled particles are not susceptible to dissolved salts, a major advantage for applications in organisms.

Several proof-of-concept studies from Joseph Wang's group at the University of California, San Diego, have demonstrated *in vivo* use of acid- and water-propelled bubble swimmers to deliver therapeutic payloads in mice. They use magnesium particles propelled by gas bubbles that form as the particle dissolves in stomach acid. The particles are coated with the antibiotic drug clarithromycin, which is used in humans to fight a *Helicobacter pylori* infection in the stomach and upper small intestine. In addition, microswimmers have been used to deliver drugs in the murine gastrointestinal tract (figure 4d). The process relies on clothing the swimmer with an acid-resistant enteric coating that degrades in the more neutral pH of the intestinal tract. The swimmers pass through the stomach unaltered but become motile once the enteric coating wears off; the point where the coating wears off and the swimmer becomes motile is determined by the thickness of the coating.

Christian Kastrup and coworkers at the University of British Columbia used other self-propelled microswimmers to deliver blood coagulants to severe hemorrhages in mice and pigs.[17] The particles, made from carbonate and tranexamic acid, used bubble propulsion to deliver thrombin, a blood clotting agent, several millimeters into the vasculature of a wound. When loaded with active thrombin, the particles halted severe hemorrhage in multiple animal models of intraoperative and traumatic bleeding.

Engineering swimmers to address environmental challenges, such as remediation of oil spills, may also be possible. When placed together with micron-scale oil droplets dispersed in seawater, magnesium particles asymmetrically covered with a hydrophobic coating swam through the suspension, and the oil droplets adsorbed onto the swimmers' hydrophobic surfaces.[18] The particles are propelled by breaking down seawater and generating hydrogen bubbles, similar to the gastric-acid-propelled swimmers mentioned above. The particles can be directed, collected, and reused, if they have a ferromagnetic metal incorporated into them.

Microswimmers can also clean up chemical pollutants by using them as fuel. Palladium-based swimmers in water can turn toxic nitroaromatic compounds—persistent, nonbiodegradable pollutants from industrial production of dyes and pesticides—into nontoxic by-products using a catalytic reduction reaction. Similarly, in the presence of light, TiO_2 particles use a photocatalytic reaction to destroy *Bacillus globigii* spores—a simulant of anthrax—thereby degrading an organophosphate chemical warfare agent into harmless by-products.

Engineered, self-propelled, microscopic particles that can autonomously interact with their local environment to perform useful functions sound like the basis for a hit science fiction movie. Researchers have made great strides in understanding the physics that drives them and in developing swimmers that operate in a wide range of real-world conditions. However, there are still many unanswered questions about the self-propulsion mechanisms. As the variety of artificial microswimmers grows and their general principles are better understood, the number of applications to people's health and the environment will expand rapidly.

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