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The Environmental Impact of Micro/Nanomachines: A Review

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ABSTRACT Environmental sustainability represents a major challenge facing our world. Recent advances in synthetic micro/nanomachines have opened new horizons for addressing environmental problems. This review article highlights the opportunities and challenges in translating the remarkable progresses in nanomotor technology toward practical environmental applications. It covers various environmental areas that would benefit from these developments, including nanomachine-enabled degradation and removal of major contaminants or nanomotor-based water quality monitoring. Future operations of autonomous intelligent multifunctional nanomachines, monitoring and responding to hazardous chemicals (in a "sense and destroy" mode) and using bioinspired chemotactic search strategies to trace chemical plumes to their source, are discussed, along with the challenges of moving these exciting research efforts to larger-scale pilot studies and eventually to field applications. With continuous innovations, we expect that man-made nano/microscale motors will have profound impact upon the environment.

KEYWORDS: nanomachines · nanomotors · nanoremediation · environmental monitoring · decontamination · artificial microfish · microengines · chemotaxis

The propulsion of nanoscale objects represents a major challenge and opportunity, and has thus stimulated considerable research efforts.[1−12] Nanomachines are nanodevices that perform various tasks during their movement.[1−4] Tremendous progress has been made recently toward the fabrication of micro/nano motors that can be propelled by different mechanisms, such as self-electrophoresis,[13−15] bubble propulsion,[16−21] diffusiophoresis,[22−25] and by using external stimuli such as light,[26,27] magnetic[28−32] or ultrasound[33,34] fields. Inspired by the "Fantastic Voyage" movie, early efforts in synthetic nanomachines have focused on diverse biomedical applications.[35−38] These include targeted drug delivery,[39−41] the manipulation and isolation of cells,[42−44] bioimaging,[45] nanosurgery and nanodrilling,[46] and very recently intracellular propulsion.[47] However, recent activity has illustrated that micro/nanomachines could also have a profound impact upon the environmental field. Potential environmental applications of nanomachines have grown rapidly in recent years, indicating a myriad of remediation and monitoring applications based on new motion-based phenomenon.

Environmental sustainability represents a major challenge that our planet is currently facing. Human activity during the era of industrialization has resulted in excessive emissions of hazardous pollutants into our water and air resources. Aquatic ecosystems, which are crucial for sustainable development and for human well-being, have thus been severely threatened throughout the globe. The continued environmental degradation, along with the rapid population growth and increasing demands for clean water, place urgent needs for new technologies and innovative solutions aimed at protecting our water and air resources and ensuring widespread access to clean and affordable water.[48,49]

Nanotechnology presents significant opportunities for a wide range of environmental applications, and it has already made a profound impact in the environmental field. For example, advanced nanosensors and reactive nanomaterials have been developed to facilitate the detection of major pollutants[50] or the remediation of contaminated sites,[48,51,52] respectively. These environmental applications have relied on the attractive properties of nanomaterials, including high surface area, catalytic and

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antimicrobial activity, size-dependent tunable optical behavior, and surface chemistry.[49]

While nanotechnology has been shown extremely useful for addressing environmental problems for nearly two decades, related applications of nanomachines have realized only over the past three years. These recent proof-of-concept efforts by several research groups have demonstrated that synthetic nanomachines can open the door to diverse environmental applications ranging from accelerated decontamination processes,[53,54] water-quality screening,[55] to the removal of oil spills.[56]

The objective of the present review is to highlight recent progress and future possibilities and challenges in developing functional nanomachines for environmental applications. Particular attention is given to translating recent advances in nanomachines into new environmental remediation and monitoring processes, involving autonomous tracking, isolation and degradation of pollutants, or toxicity screening. These recent proof-of-concept studies exemplify the versatility of nanomachines and are indicative of their wide range potential environmental applications. In particular, in the next sections we will illustrate how the movement of functional nanomachines adds a new and powerful dimension to environmental monitoring and cleanup protocols. Such applications benefit also from the ability of these tiny machines to penetrate otherwise inaccessible locations. We will also discuss progress toward the development of green self-degradable micromotors capable of harvesting energy directly from natural environments, and will conclude with the challenges of moving these exciting advances from the bench scale to larger-scale pilot studies and eventually to field applications.

Use of Micro/Nanomotors for Environmental Sensing and Monitoring. Environmental monitoring describes activities aimed at characterizing the quality of water and air resources by measurements of chemical, biological, or physical variables. The major goals of environmental monitoring are to establish the current status of an environment, to warn about sudden changes and incoming threats, to monitor remediation processes, and to establish trends and baselines in environmental parameters. Major challenges for such effective environmental monitoring activities are high sensitivity toward extremely low concentrations of toxic micropollutants, enhanced specificity and stability in complex natural water media (with few or no sample preparation steps), and fast response times.[49] On-site and real-time analytical measurements have been introduced in recent years to address the limitations of discrete sample collection and subsequent laboratory analysis.[57] Nanomachines could offer innovative concepts for effective environmental monitoring. For example, the large cargo-towing force of modern micromachines, along with their ease of surface functionalization, can enable new target isolation ("*Capture–Transport*") strategies.[58] Mimicking biological behavior, nanomachines could use chemotactic search strategies to trace chemical plumes to their source. Catalytic micromotors have been shown to display a chemotactic behavior in the presence of a gradient of the fuel concentration, with a directed movement and increased speed toward higher peroxide concentrations.[59,60] Changes in the swimming behavior in the presence of hazardous chemicals can offer direct and timely visualization of chemical stress. Finally, autonomous mobile nanomachines hold considerable promise for monitoring inaccessible locations or hostile environments.

For example, we reported on a motion-enabled sensing of trace silver ions based on the dramatic and specific acceleration of bimetal nanowire motors in the presence of this toxic ion (Figure 1).[61] The specific acceleration has been attributed to the underpotential deposition of silver onto a platinum segment, which increases the electrocatalytic activity. The resulting silver sensing protocol relies on the use of an optical microscope for tracking changes in the speed of the nanomotors in the presence of the target analyte (Supporting Information, Video S1). The highly selective motion-based response is characterized with a defined concentration dependence, with the speed (or distance) providing the quantitative information down to the nanomolar level. Such changes in the swimming behavior indicate promise for tracing plumes of hazardous chemicals. The silver-based acceleration has formed the basis for a motion-based detection of DNA hybridization in connection to silver nanoparticle tags.[62] Dissolution of the silver nanoparticles, in the presence of the hydrogen peroxide fuel, lead to well-defined dependence of the speed signal on the concentration of the target DNA, down to femtomolar level. Recently, Gao *et al.* reported that Ir/silica Janus micromotors can be powered by extremely low levels of hydrazine fuel (down to 1 ppb).[63] The authors observed a well-defined concentration/speed dependence that indicates considerable promise for measuring trace levels of hydrazine and tracing concentration gradients.

Mimicking life-fish toxicity testing,[55] our group introduced a simple nanomachine-based strategy for

VOCABULARY: nanomachine - a nanoscale device that performs a task; **nanomotor** - a nanoscale device capable of converting energy into movement and forces; **decontamination** - process of removing, neutralizing, or destroying harmful substances; **environmental monitoring** - activity used for characterizing the quality of our surroundings; **bubble propulsion** - a force causing movement that results from bubble detachment; **nanoremediation** - use of reactive nanomaterials for transformation and detoxification of pollutants; **Marangoni effect** - mass transfer along an interface between two fluids due to surface tension gradient;

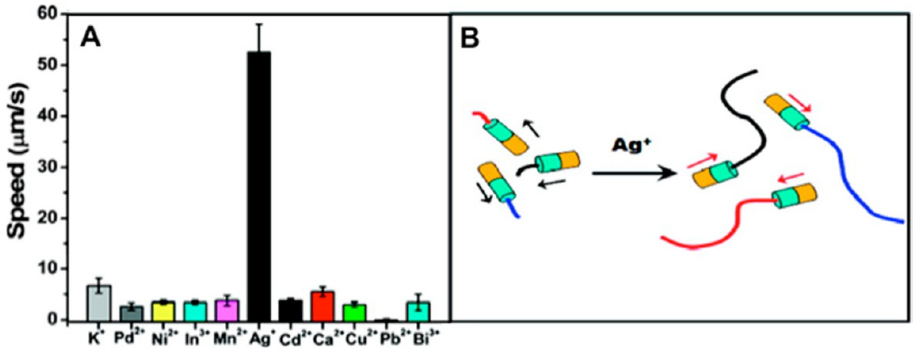

Figure 1. Motion-based sensing of trace silver ions using catalytic Au−Pt nanowire motors. (A) Selectivity: bar graph comparing the nanomotor speed in 11 different 100 μM metal−nitrate salt solutions (of K^+, Pd^{2+}, Ni^{2+}, In^{3+}, Mn^{2+}, Ag^+, Cd^{2+}, Ca^{2+}, Cu^{2+}, Pb^{2+}, and Bi^{3+}). (B) Accelerated propulsion of catalytic nanomotors in the presence of silver ions. Reprinted from ref 61. Copyright 2009 American Chemical Society.

in situ water-quality testing based on changes in the propulsion behavior and lifetime of biocatalytic microswimmers in the presence of aquatic pollutants (*e.g.*, heavy metals, pesticides). Such use of artificial fish for water-toxicity screening relies on the toxin-induced inhibition of the catalase enzyme powering the microtubular engine (Figure 2A). The observed changes in the microfish speed are analogous to changes in the swimming behavior and survival of natural fish used for toxicity testing.[55] Exposure to a toxic pollutant thus leads to a rapid decrease in the rate of bubble generation and hence of the microfish speed (Figure 2B), leading to a direct assessment of the water quality. Such use of self-propelled artificial swimmers thus allows direct visualization (optical tracking) of changes in the swimming behavior in response to the presence of chemical stress. Various organic and inorganic pollutants, such as mercury, copper, sodium azide and aminotriazole, have thus displayed a significant concentration-dependent effect upon the swimming behavior of the artificial fish. The microfish water-toxicity assay strategy thus offers direct monitoring of aquatic contaminants down to the micromolar levels and addresses major drawbacks and ethical concerns associated with live-fish toxicity bioassays.

Isolation, separation and identification of target analytes are extremely important for meeting environmental monitoring requirements.[64] Self-propelled functionalized nanomachines can offer promise for the direct selective isolation of target analytes from untreated environmental samples, thus obviating laborious sample preparation steps.[58] In particular, receptor-modified tubular microengines have been shown extremely useful for capturing, transporting and detecting a wide range of target bioanalytes. Such ability to move the receptor through the contaminated sample is unique compared to traditional bioaffinity assays.[65]

For example, our group reported a nanomachine-based "capture and transport" isolation protocol for separating the *Escherichia coli* bacteria from environmental samples.[66] *E. coli* and other pathogenic bacteria are commonly detected using traditional culture techniques, which are time-consuming and labor-intensive. The nanomachine protocol is based on catalytic tubular microengines functionalized with the Concanavalin A (ConA) lectin bioreceptor (Figure 3). Lectins are readily available glycoproteins that offer an attractive route for recognizing carbohydrate constituents of bacterial surface, *via* selective binding to cell-wall mono- and oligosaccharide components. Applicability of the microtube transporter to the direct isolation of *E. coli* from untreated seawater and drinking water samples was demonstrated.

Rapid nanomachine-based target isolation directly from raw samples can also benefit molecular biological techniques relevant to environmental water quality, as was demonstrated by Kagan *et al.*[67] For this purpose, single-strand DNA-functionalized micromotors have been used as selective transporters of complementary oligonucleotides (synthetic DNA or bacterial rRNA), allowing "on-the-fly" hybridization and selective single-step isolation of target nucleic acids without preparatory and washing steps. Tagging a secondary probe with a fluorescent tracer offers convenient visualization of the target binding and transport processes, through the optical monitoring of the resulting duplex. Rapid hybridization processes have thus been observed because of the fast movement and efficient localized convection. This selective "on-the-fly" hybridization and isolation allows for the sensitive detection of nanomolar levels of target with no interferences from a large excess of nontarget or mismatch sequences. Another motion-based DNA sensing protocol, based on enzymatic-powered asymmetric hybrid silica nanomotors, was developed by Simmchen *et al.*[68] These micromachine-enabled nucleic acid hybridization/isolation protocols could facilitate the identification of aquatic microbes and on-site microbiological water quality testing.[69]

Use of Micro/Nanomotors for Environmental Remediation. The removal and destruction of pollutants from

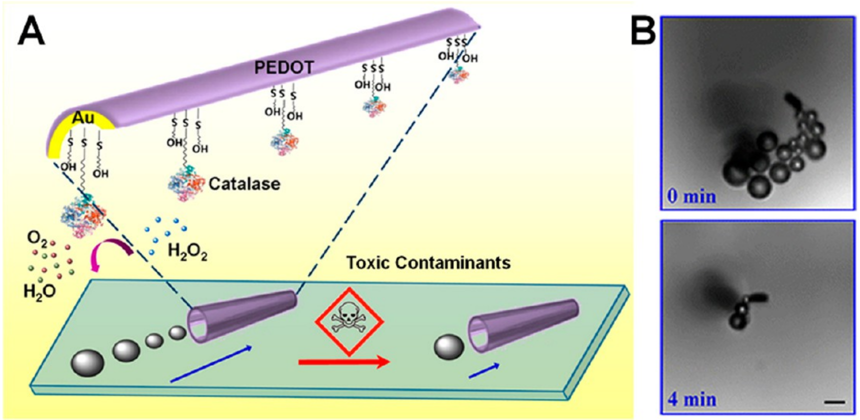

Figure 2. Artificial microfish for water quality monitoring. (A) Scheme illustrating the pollutant effect on the microfish locomotion speed through inhibition of the catalase biocatalytic layer along with the protocol used for immobilizing the enzyme at the inner gold surface of the tubular microengine. (B) Time-lapse images of the microfish recorded after 0 and 4 min swimming in a 100 μM Hg solution. Scale bar, 6 μm. Reprinted from ref 55. Copyright 2013 American Chemical Society.

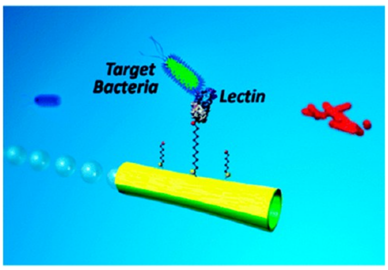

Figure 3. Lectin-modified microengines for bacteria detection. Schemes depicting the selective pick-up and transport of the target bacteria by a ConA-modified microengine in environmental matrices. Reprinted from ref 66. Copyright 2012 American Chemical Society.

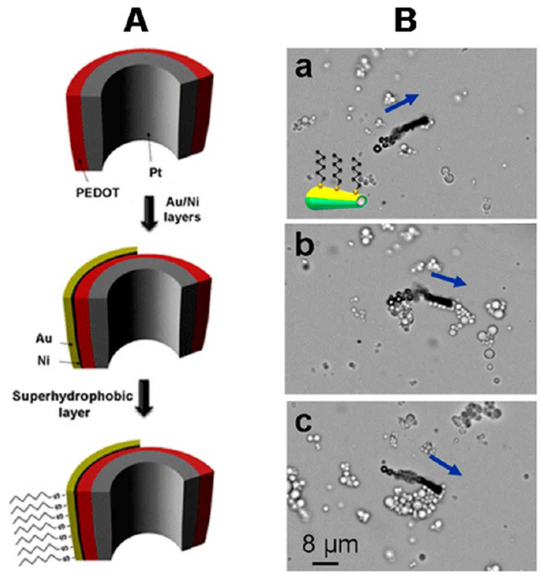

Figure 4. Self-propelled micromachines for the removal of oil droplets. (A) Cross-section of a superhydrophobic SAM-modified Au/Ni/PEDOT/Pt tubular microengine. Au and Ni layers are deposited onto the PEDOT/Pt microengines by e-beam and a superhydrophobic layer is formed on the Au surface by incubation in a 0.5 mM *n*-dodecanethiol ethanolic solution. (B) Hexanethiol-modified microsubmarine transporting a payload of multiple oil droplets. Time-lapse images at different navigation times: 11, 50, and 73 s for a, b, and c, respectively. Reprinted from ref 56. Copyright 2012 American Chemical Society.

contaminated media is an important focus of environmental sustainability.[70] The field of water purification and wastewater treatment has grown rapidly during the 1980s and 1990s because of the urgent demands for sustainable water resources and new stringent water quality regulations. Environmental remediation uses a wide variety of methods for cleaning up wastewater, contaminated groundwater, surface water, or sediments. Nanotechnology-based remediation methods (nanoremediation) entail the application of reactive nanomaterials for transformation and detoxification of pollutants toward purifying our water and air resources.[49] In particular, reactive nanomaterials have attractive properties that enable the efficient transformation of toxic pollutants.[51,52]

Nanomachines offer distinct advantages over conventional nanoremediation agents. Such tiny machines add a new dimension based on motion to decontamination processes, lead to new *in situ* and *ex situ* nanoremediation protocols, and have the potential to reduce the cleanup time and overall cleanup costs. In particular, the continuous movement of such nanoscale objects can be used for transporting reactive (water-purification) nanomaterials throughout contaminated samples, for releasing remediation agents over long distances, and for imparting significant mixing during detoxification processes. Eventually, one would expect autonomous multifunctional "remisense" nanomachines, coupling the remediation and sensing of pollutants into the same platform, analogous to theranostic particles used for biomedical applications. As will be illustrated below, these capabilities open up the door for efficient cost-effective routes for environmental remediation.

Micromachine-Enabled Removal of Contaminants. Guix *et al.* described the modification of tubular microengines with long chain of self-assembled monolayers (SAMs) toward the cleanup of oil-contaminated water samples (Figure 4).[56] Such SAM-functionalized hydrophobic microsubmarines offer high oil-adsorption ability, *via* adhesion and permeation onto its alkanethiol coating, for effective *in situ* cleanup of oil-contaminated water samples. Controlling the surface hydrophobicity, through the use of different chain lengths and head functional groups, allows tuning of the extent of the micromotor–oil interaction and the collection efficiency. The influence of the alkanethiol chain length upon the oil–nanomotor interaction was thus examined by modifying the outer gold surface of the microengine with SAMs of different chain lengths (C6, C12, and C18). The longer the navigation time, the more oil droplets are collected and confined onto the surface of the self-propelled micromotor (Figure 4B). Efficient capture and transport of multiple oil droplets has thus been observed during the navigation of the SAM-modified microengines in contaminated water samples (Supporting Information, Video S2). This proof of concept demonstration should be evaluated in larger-scale pilot studies toward addressing practical barriers associated with large oil spills. Such cleanup of larger areas would benefit from the use of swarm of multiple hydrophobic micromotors as well as of larger motors with greatly faster absolute speeds. These oil-carrying SAM modified microengines could be reused after releasing and degrading the captured oil droplets.

Molecularly imprinted polymers (MIPs) offer another promising route for "on-the-fly" removal of target pollutants. MIPs have been shown particularly attractive as selective binding medium for the removal of highly toxic micropollutants.[71] Several groups demonstrated recently that MIPs represent an attractive route for creating of specific recognition sites in synthetic self-propelled nanomachines.[72,73] Tailor-made recognition sites have thus been introduced into tubular-microengine[72] or Janus-particle[73] transporters through cavities in their polymeric material. The resulting template-imprinted micromotors offer attractive capabilities for autonomous binding, transport, and removal of the target contaminant, including isolation from raw environmental matrices.[72]

Nanomachine-Based Acceleration of Decontamination Processes. The propulsion of nanomachines can lead to enhanced fluid mixing and enhanced transport of the remediation reagents and hence to greatly accelerated decontamination processes without external forced convection (*e.g.*, stirring). Among them, bubble-propelled tubular micromotors have demonstrated a remarkable capability to induce self-stirring and fluid convection in remediation solutions and to accelerate chemical reactions.

The new concept of nanomachine-enabled decontamination has been demonstrated recently by our team in connection to the peroxide-induced oxidative

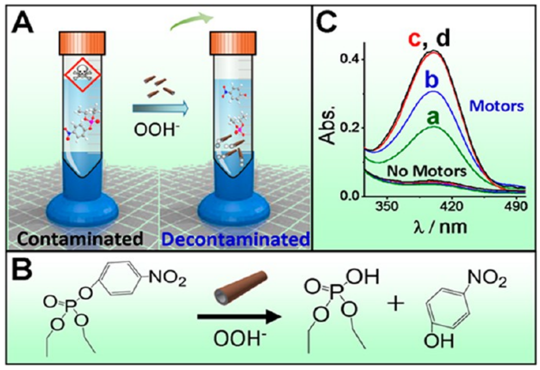

Figure 5. Micromotor-based accelerated oxidative detoxification of OP nerve agents. (A) Movement of multiple tubular microengines in the contaminated sample (containing H_2O_2 oxidant/fuel and $NaHCO_3$ as activator), leads to accelerated degradation because of an enhanced fluid motion. (B) The micromotor-based strategy allows rapid detoxification of chemical threats under mild conditions and involves the *in situ* generation of OOH^- nucleophiles. (C) The decontamination efficiency is demonstrated by measuring the absorbance of the *p*-NP reaction product at 400 nm at different reaction times (a–d). Reprinted from ref 53. Copyright 2013 Wiley-VCH.

detoxification of organophosphorous (OP) nerve agents.[53] Figure 5A illustrates the concept of micromotor-accelerated decontamination of chemical threats. It involves placing a known number of micromotors in a nerve-agent-contaminated solution, along with hydrogen peroxide (used as the oxidizing agent as well as the micromotor fuel), the peroxide activator ($NaHCO_3$ or NaOH), and the surfactant sodium cholate (NaCh), which facilitates bubble generation. The oxidative conversion of the OP nerve agent into *para*-nitrophenol (*p*-NP) was achieved under mild quiescent conditions that involve the *in situ* generation of OOH– nucleophiles with no external stirring (Figure 5B). The decrease in concentration of the OP agent was monitored spectrophotometrically by measuring the absorbance of the *p*-NP product at 400 nm (Figure 5C). The continuous movement and bubble generation induced by motors across a peroxide-activated contaminated sample results in greatly enhanced diffusion and hence leads to a higher detoxification efficiency while using significantly shorter reaction times and lower peroxide concentrations compared to common CWA neutralization processes. The accelerated decontamination achieved by the motor-induced self-stirring of the remediation solution has been attributed to the large-scale collective motion of the micromotors and to the evolution of microbubbles that leads to an enhanced mixing. The new micromotor strategy is expected to enhance the efficiency and speed of decontamination reactions of a broad range of threats and pollutants, but also of chemical processes in general. Larger-scale pilot study currently assesses the scope of the new decontamination method.

In addition to using motion-enabled enhanced diffusion (mixing) for accelerating the detoxification

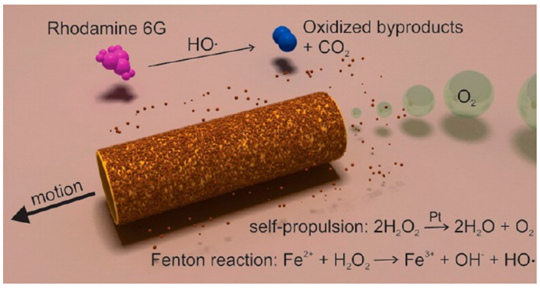

Figure 6. Tubular Fe/Pt bilayer microengines for destroying organic contaminants based on the Fenton reaction. The micromotors contain double functionality with an inner Pt layer for the self-propulsion and the outer Fe for the *in situ* generation of Fe^{2+} ions. The ions, along with the H_2O_2 fuel, generate HO• radicals that degrade organic pollutants. Reprinted from ref 54. Copyright 2013 American Chemical Society.

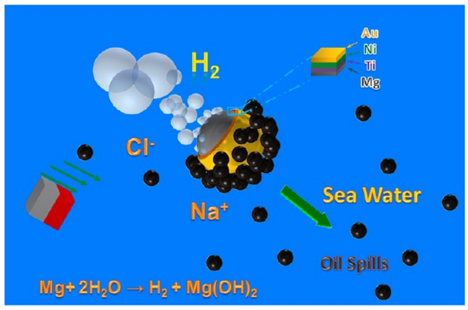

Figure 7. Seawater-powered magnesium-based Janus micromotor for environmental oil remediation. Reprinted from ref 74. Copyright 2013 Royal Society of Chemistry.

processes, it is possible to use self-propelled nanomachines, comprising of a reactive material or carrying the remediation agent, while moving through the contaminated water systems. This novel dual-function ("move and destroy") micromotor concept was demonstrated recently by Sanchez *et al.*[54] that reported the use of catalytic tubular microengines for degrading organic pollutants in water *via* the Fenton oxidation process. As illustrated in Figure 6, these attractive self-propelled bilayer micromotors are composed of rolled-up functional nanomembranes consisting of an inner Pt surface and an outer Fe surface (without further functionalization). The Fe layer leads to *in situ* generation of ferrous ions that, along with the hydrogen peroxide fuel, acts as the Fenton's reagent for the destruction of the organic compounds. In this case, hydrogen peroxide works as a coreagent for the Fenton reaction as well as the fuel to power the microengines. The motor movement was shown to induce convection and mixing of the reagents within the solution and led to a faster decontamination of the organic compounds.

Powering Micro/Nanomotors by Harvesting Energy from Natural Environments. The requirement of the common hydrogen peroxide fuel hinders practical environmental applications of catalytic micro/nanomachines, particularly those involved *in situ* operations. Extending the scope of these tiny machines to diverse environmental applications would require harvesting the energy directly from the environmental sample matrix itself. The use of environmental matrices to power micromotors would require the identification of new materials and reactions, and will obviate the need for adding external fuels.

Of particular interest for practical environmental applications are recently developed seawater-driven Janus micromotors that utilize the magnesium-water reaction for the propulsion (Figure 7).[74,75] The new micromotors consist of biodegradable and environmentally friendly magnesium microparticles coated with a nickel−gold bilayer patch for magnetic guidance and surface modification. Such seawater-driven micromotors utilize macrogalvanic corrosion and chloride pitting corrosion processes to generate hydrogen bubbles that propel the microparticles.[74] This eliminates the need for external fuels to offer efficient propulsion in untreated seawater toward diverse applications in aquatic environments. For example, the modification of Mg-based micromotors with self-assembled monolayers (SAMs) of long-chain alkanethiols has been used for "on-the-fly" collection of oil droplets. Another water-driven micromotor explores the Al−Ga alloy-based Janus particles for efficient hydrogen bubble propulsion through a process called "liquid metal enbrittlement".[76] Additional efforts should be devoted to extending the lifetime of these water-powered micromotors for addressing the requirements of practical environmental applications.

It is possible to explore other sample constituents as fuels. For example, a polyaniline/Zn tubular microengine has been shown to propel autonomously and efficiently in acidic environments without an additional fuel.[77] This acid-powered microrocket could be employed in extreme environments and used for motion-based environmental pH sensing. A hybrid Janus Pd−Al micromotor that harvests energy from the reactions of three different fuels, acid, base and hydrogen peroxide, has also been described.[78]

Macroscopic Motors and the Environment. As indicated from above discussion, considerable progress has been made for micro/nanomachines enabled environmental applications. However, various large-scale environmental applications may be hindered by the small size and limited speed of such tiny machines. Considerably larger (millimeter-scale) motors will be extremely useful for dynamic action on larger scales and for ferrying larger loading of self-released remediation agents. Various sources of energy have been used to power the motion of macroscale motors, including electrical or magnetic fields, thermal gradient and chemical fuels.[1,79−84] Among these, Marangoni effects, involving propulsion of motors along an interface based on a surface tension gradient, have received

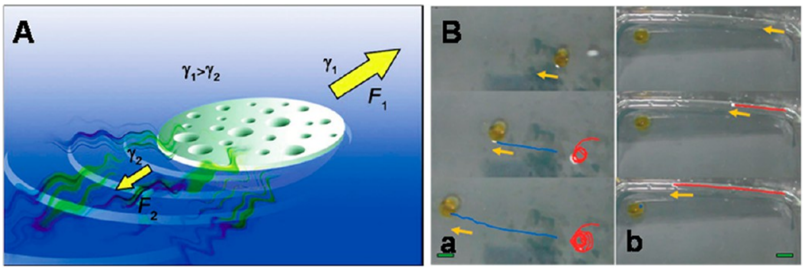

Figure 8. Polymer capsule motors for environmental oil cleanup. (A) Mechanism for the motion of the macrocapsule involving asymmetric release of dimethylformamide (DMF). Changes in the surface tension (DMF/water *vs* water) result in a forward net force which drags the capsule through the water. (B) Tracking lines of the motion of oil droplets (blue line) and SDS/PSf (a) and plain (b) capsules (red line). The oil droplet is pushed away by the SDS/PSf capsule at a significantly higher speed. The orange arrows indicate the movement direction of the capsules. Scale bar: 1 cm. Images taken at 1 s intervals. Reprinted from reference 85. Copyright 2011 Wiley-VCH.

considerable attention.[1,79] Recent efforts demonstrated that such macromotors hold considerable promise for diverse environmental applications due to their unique autonomous propulsion behavior at liquid/liquid and liquid/air interfaces without external fuel requirement.[79]

Zhao *et al.* described a millimeter-sized polymer capsule motor based on Marangoni effects with specific features and functionalities (Figure 8).[85] Such macrocapsule motor can operate at a high velocity without any external fuel. Incorporating nickel powder within the motors offered guided motion under an external magnetic field. The sodium dodecyl sulfate (SDS)/polysulfone (PSf)-polymeric capsule display long-range interaction with oil droplets, and can be used to clean the oil contaminant from the water−air interface. As illustrated in Figure 8Ba, the SDS/PSf capsule repels the oil droplets over a long distance of several centimeters and induced their movement away from the capsule because of the Marangoni effect and the resulting force. In contrast, the SDS-free capsule does not influence the movement of the oil droplets (Figure 8Bb). The long-distance interactions between the SDS/PSf capsule and oil droplets could be used to shepherd and merge several oil droplets, effectively cleaning the water surface due to the chemotaxis effect of the SDS released from the SDS/PSf capsule. More recently, Pumera group extended this activity by incorporating various levels of different-charged surfactants (SDS, cetyltrimethylammonium bromide (CTAB), Tween 20) to induce motion of the oil droplet.[86] The team recorded a gradual increase in the velocity with respect to increasing concentrations of the incorporated surfactant.

Enzyme-based bioremediation is an attractive alternative to conventional techniques for waste treatment applications. Orozco *et al.* illustrated a new motion-based biocatalytic strategy for decontaminating phenolic and azo-dye compounds based on self-release and enhanced mixing of the bioremediation agent. The method involved tubular macromotors, self-propelled by the surface-induced Marangoni effect, where the SDS release is coupled to simultaneous

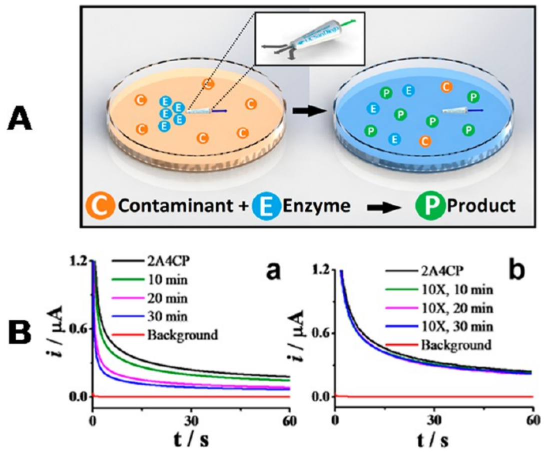

Figure 9. Biocatalytic remediation based on enzyme-releasing motors. (A) Scheme illustrating the bioremediation protocol involving gradual release and mixing of the enzyme laccase during the motor movement. Pipette tips, filled with a mixture of the enzyme solution and SDS as remediation agent and propeller, respectively, navigate in a contaminated solution. During this process, the pollutant is biocatalytically transformed into an innocuous product. (B) Current signals illustrating the accelerated dispersion of laccase and improved reaction efficiency: (a) Decreased chronoamperometric response of the 2-amino-4-chlorophenol (2A-4CP) pollutant when 6 motors, containing laccase and SDS, navigate in a 50 mL of a 250 μM 2A-4CP polluted solution for 10, 20, and 30 min. (b) Current signals for 2A-4CP obtained in control experiment involving a quiescent solution containing a 10-fold excess of enzyme or/and SDS, expected to be released from the 6 motors under similar conditions. Reprinted from ref 87. Copyright 2014 Wiley-VCH.

release of the laccase biocatalyst into the polluted sample (Figure 9A).[87] The movement of the motors was shown to generate an effective fluid convection in the sample, and hence to rapidly disperse the released enzyme (without an external stirring) and to enhance the substrate−enzyme interactions. As illustrated in Figure 9B, such enzyme-releasing motors offer a greatly improved degradation efficiency compared to quiescent solutions containing excess levels of the free enzyme (a *vs* b). This concept could be expanded to the release of a variety of remediation agents over long distances toward the decontamination of different pollutants. For example, preliminary results indicated

TABLE 1. Summary of Environmental Applications of Synthetic Motors

scope	applications	type of motors	mechanisms	ref
nanomachines for environmental monitoring	silver detection	Au—Pt nanomotors	silver-induced acceleration	Kagan *et al.*[61]
	DNA detection	Au—Pt nanomotors	DNA hybridization using silver nanoparticle tags; silver-induced acceleration	Wu *et al.*[62]
		bubble-propelled Pt-based microrockets	"on-the-fly" DNA hybridization	Kagan *et al.*[67]
		catalase-based Janus motors	DNA hybridization	Simmchen *et al.*[68]
	water toxicity screening: heavy metals, pesticides	artificial microfish (catalase-powered microengines)	toxin-induced inhibition of the biocatalytic layer	Orozco *et al.*[55]
	bacteria detection	lectin-functionalized bubble-propelled Pt-based microengines	lectin recognition of the sugar on the bacteria wall	Campuzano *et al.*[66]
	hydrazine detection	Ir/SiO_2 Janus motors	speed-concentration dependence	Gao *et al.*[63]
	pH monitoring	Zn-based and Al—Pd-based micromotors	speed-pH dependence	Gao *et al.*[77]
nanomachines for environmental remediation	oil removal	SAM modified Pt-based microengines	hydrophobic interactions with oil droplets	Guix *et al.*[56]
		SAM modified Mg-based micromotors	hydrophobic interactions	Gao *et al.*[74]
	oxidative detoxification of nerve agents	bubble-propelled Pt-based microengines	enhanced fluid transport	Orozco *et al.*[53]
	degradation of organic contaminants	bubble-propelled Fe—Pt microengines	Fenton reaction; enhanced difussion	Soler *et al.*[54]
	solid-phase extraction	MIP-PEDOT/Pt-based tubular microengines	recognition by embedded MIP sites	Orozco *et al.*[72]
		Ag-based Janus MIP microparticles	MIP recognition	Huang *et al.*[73]
macroscale motors and environments	environmental oil cleanup	polymer capsule motors	surface tension induced cargo towing	Zhao *et al.*[85] and Seah *et al.*[86]
	enhanced diffusion of pollutants	hydrophobic agglomerates of pollutants	surface tension induced self-propulsion of pollutants	Zhao *et al.*[88]
	pH monitoring	Pd nanoparticle-containing microspheres	pH taxis	Dey *et al.*[89]
	degradation of phenolic pollutants	SDS-based enzyme-releasing motors	biocatalytic degradation	Orozco *et al.*[87]

efficient removal of heavy metals (*e.g.*, lead) in connection to the release of the EDTA complexing agent.

The propagation of pollutants in water is very slow without external fluxes. Pumera *et al.* reported on the enhanced diffusion coefficients of pollutants due to the presence of self-propelled macroscopic particles during the slow asymmetric dissolution of solid agglomerates of hydrophobic particles.[88] The greatly enhanced diffusion phenomenon, observed in different natural water matrices (*e.g.*, pond and seawater), reflects changes in asymmetric surface tension. Chattopadhyay's group demonstrated the use of Pd nanoparticle-containing polymer microsphere motors for sensing pH gradients.[89] These micromotors move toward higher pH region, with increasing velocity, indicating promise for environmental pH monitoring.

CONCLUDING REMARKS AND FUTURE PROSPECTS

In conclusion, we have described recent progresses and demonstrated that nanomachines present variety of opportunities in the environmental field, ranging from accelerated decontamination to *in situ* water toxicity screening (Table 1). These early developments indicate that nanomachines enable novel environmental applications and that the movement of such tiny machines adds a new and powerful dimension to such remediation and sensing procedures. While the proof-of-concept of different potential applications has been demonstrated lately by several research groups, and new environmental capabilities of nanomachines have emerged, significant efforts are required before large-scale environmental benefits are realized. These recent accomplishments are thus only the first step toward the design of functional and sophisticated nanomachines, swimming in natural water environments and performing demanding sensing and cleanup activities. It is hoped that future efforts, in close collaboration with environmental scientists and engineers, along with new technological breakthroughs and greater sophistication of micro/nanoscale machines, will lead to rapid translation of the above research activity into practical environmental applications, hence contributing further to the environmental benefits of nanotechnology. Given the immense interest and the competitive cutting-edge research in the field of nanomachines, we anticipate exciting new ideas and applications in the future. Recent initial proof-of-concept studies are thus expected to advance into practical environmental applications.

Translating these recent and exciting advances to widespread environmental applications would require major attention to key challenges. Particular attention should be given to scale-up issues toward coverage of large contaminated areas and large volumes of wastewater. This would require additional advances and innovations and pilot studies involving different sample sizes. Larger (micrometer-to-millimeter scale) motors, swimming over long distances, and the coordinated action of swarms of numerous micromotors hold promise for dynamic action over larger scales and for transporting larger payloads of remediation agents. Many practical applications involve small samples size, *e.g.*, *ex situ* water toxicity screening or cleanup of contaminated pipes or capillaries. Attention should be given also to the influence of the environmental medium upon the propulsion behavior. While environmental samples can result in decreased speed of tubular microengines,[90] such bubble-propelled micromotors maintain efficient propulsion in most natural water media.[91] Finally, the potential toxicity of micro/nanoscale motors needs to be evaluated to prevent potential adverse environmental impacts. Recent efforts in this direction have also been devoted for designing environmentally friendly and self-degradable nanomachines based on green materials (*e.g.*, Zn or Mg).[74,75,77] Selecting nanomachines for specific environmental applications would depend on the complexity of the problem and economic costs, and require comparison to traditional technologies. Where do we go from here? The ability of nanomachines to address environmental problems is just beginning to be explored. In the not-so-distant future, we anticipate seeing self-regulated multifunctional nanomachines, capable of performing multiple tasks, "sensing, isolating and destroying" toxic pollutants and chemical threats, searching for sources of hazardous chemicals, or delivering nanosensors and nanosamplers to remote hostile locations. Swarms of micromachines could be assembled in response to hazardous conditions, used for mapping the spread of a toxic pollutant over a large area or for accelerating environmental cleanup. Our preliminary data demonstrated the organized schooling of micromotors in the presence of a hazardous material.[92] These and similar developments will have a significant impact upon the environmental field, hence addressing the environmental sustainability challenges facing our world.

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Supporting Information Available: Additional videos. This material is available free of charge *via* the Internet at http://pubs.acs.org.

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