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CURRICULUM VITAE

Date of Birth: January 10, 1985.
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Address of Communication: Flat No: 402, Ichapur H.I.T. Road, Sealdanga More, Opp. Tara Maa Ashram, P.O. Santragachi, Howrah – 711104, West Bengal, India.

EDUCATION

2011- 2018 **Ph.D. (Nanotechnology)**, Indian Institute of Technology Guwahati, India.

Status: Received Doctoral Degree Certificate

Ph.D. thesis: “Multimodal Propulsion of Synthetic Microbots”

Synopsis:(1) Demonstration of non-biological pH taxis of polymeric magneto-catalytic microbots. The bots were applied to sense the pH gradient while in motion. The motor size, strength of magnetic field and the imposed pH gradient were modulated to characterize the behavior and velocity of the pH driven microswimmers. (2) Formic acid driven magneto-responsive microbots for fuel cell applications. (3) Facile lithography-free fabrication of paper-based tubular microengines for cargo transportation and drug delivery. (4) Carbon soot-based micromotors for environmental remediation.

2008 - 2010 **Master of Technology (Biotechnology)**, Heritage Institute of Technology, Kolkata, W. B., India. (DGPA - 9.03/10)

Master’s thesis: “Cloning, Sequencing and Characterization of Exonuclease gene of the mycobacteriophage L1”

Synopsis: Isolation of L1 exonuclease gene was followed by PCR amplification. The amplified product was cloned in pET28a(+) vector and expressed in a bacterial system for tuberculosis infection studies.

2004 - 2008 **Bachelor of Technology (Biotechnology)**, Heritage Institute of Technology, Kolkata, W.B., India. (DGPA – 8.24/10)

Bachelor’s thesis: “Target Identification by *in-silico* studies on NS1 proteins of bird flu virus”

Synopsis: Identified a conserved domain amongst pandemic flu viruses using bioinformatics for the development of a vaccine against this conserved site.

FELLOWSHIPS/AWARDS

1. Qualified Graduate Aptitude Test in Engineering (GATE) - GATE 2007 in Life Sciences and GATE 2011 in Biotechnology.
2. Received DST-SERB International Travel grant to attend the International Symposium on Micro- and Nanomachines, Germany, 2016.
3. Received Gandhian Young Technological Innovation (GYTI)- SRISTI Award, 2019.

INTERNATIONAL JOURNAL PUBLICATIONS (SCI journals)

1. T. Bhuyan, D. Dutta, M. Bhattacharjee, **A. K. Singh**, S. S. Ghosh, D. Bandyopadhyay, "*Acoustic Propulsion of Vitamin C- loaded Teabots for Targeted Oxidative Stress and Amyloid Therapeutics*", ACS Applied Biomaterials (2019), 10, 4571. [Impact Factor: Pending]
2. T. Bhuyan, **A. K. Singh**, S. S. Ghosh, D. Bandyopadhyay, Curcumin-based magnetic microbots as efficient bactericidal agents, Bulletin of Materials Science (2019) - Submitted. [Impact Factor: 1.264]
3. N. M. Das, **A. K. Singh**, D. Ghosh, D. Bandyopadhyay, "*Graphene Oxide Nanohybrids for Electron Transfer Mediated Antimicrobial Therapy* ", Nanoscale Advances (2019), 1, 3727. [Impact Factor: Pending]
4. **A. K. Singh**, T. Bhuyan, S. Maity, T. K. Mandal, D. Bandyopadhyay, "*Multimodal Self-Propulsion of CARBOts for Water Detoxification and Oil-spill Recovery* ", Applied Materials Today (2019) – Submitted. [Impact Factor: 8.013]
5. **A. K. Singh**, S. Rarotra, V. Pasumarthi, T. K. Mandal, D. Bandyopadhyay, "*Formic acid powered reusable autonomous ferrobots for efficient hydrogen generation under ambient condition* ", Journal of Materials Chemistry A (2018), 6, 9209. [Impact Factor: 10.733]
6. T. Bhuyan, M. Bhattacharjee, **A. K. Singh**, S. S. Ghosh, D. Bandyopadhyay, "*Boolean-Chemotaxis of Logibots Deciphering the Motions of Self-Propelling Microorganisms* ", Soft Matter (2018), 14, 3182. [Impact Factor: 3.399]
7. T. Bhuyan, **A. K. Singh**, D. Dutta, A. Unal, S. S. Ghosh, D. Bandyopadhyay, "*Magnetic Field Guided Chemotaxis of iMushbots for Targeted Anticancer Therapeutics* ", ACS Biomaterials Science & Engineering (2017), 3, 1627. [Impact Factor: 4.511]
8. M. Bhattacharjee, V. Pasumarthi, J. Choudhuri, **A. K. Singh**, H. B. Nemade, D. Bandyopadhyay, "*Self-spinning Nanoparticle Laden Microdroplets for Sensing and Energy Harvesting* ", Nanoscale (2016), 8, 6118. [Impact Factor: 6.970]
9. **A. K. Singh**, T. K. Mandal, D. Bandyopadhyay, "*Magnetically Guided Chemical Locomotion of Self-Propelling Paperbots* ", RSC Advances (2015), 5, 64444. [Impact Factor: 3.049]
10. S. Kumar, **A. K. Singh**, A. K. Dasmahapatra, T. K. Mandal, D. Bandyopadhyay, "*Graphene based multifunctional superbots* ", Carbon (2015), 89, 31. [Impact Factor: 7.466]
11. S. Timung, V. Tiwari, **A. K. Singh**, T. K. Mandal, D. Bandyopadhyay, "*Capillary Force Mediated Flow-Patterns and Non-monotonic Pressure Drop Characteristics of Oil-Water Microflows ",* The Canadian Journal of Chemical Engineering (2015), 93, 1736. [Impact Factor: 1.265]
12. **A. K. Singh**, K. K. Dey, A. Chattopadhyay, T. K. Mandal, D. Bandyopadhyay, "*Multimodal chemo-magnetic control of self-propelling microbots* ", Nanoscale (2014), 6, 1398. [Impact Factor: 6.970]

PROFESSIONAL EXPERIENCE

08/2015 – Present — **Senior Research Fellow**, Centre for Excellence in Nanoelectronics & Theranostic Devices (CENTD), Indian Institute of Technology Guwahati, India
Summary: Synthetic micro/nanomotors in drug delivery and biosensing.

06/2007 – 07/2007 — **Summer Intern**, Chittaranjan National Cancer Institute , Kolkata, India
Summary: Cytopathological analysis of FNAC samples of breast cancer.

CERTIFICATION COURSES

1. "*Nanobiotechnology for healthcare*" offered by IIT Kharagpur, India (GIAN course), 2017.
2. "*Nanotechnology: A Maker's Course*" offered by Duke University, North Carolina State University & The University of North Carolina at Chapel Hill, USA (Coursera), 2018. (Online)
3. "*Biomedical Nanotechnology*" offered by Indian Institute of Technology Roorkee, India (NPTEL course), 2017.
4. "*Nanotechnology - the basics*" offered by Rice University, USA (Coursera), 2014. (Online)
5. "*Introduction to Biology- the Secret of Life*" offered by MITx, USA (edX), 2013. (Online)

SELECTED CONFERENCE PUBLICATIONS

1. **A. K. Singh,** S. Rarotra, V. Pasumarthi, T. K. Mandal, D. Bandyopadhyay, "*Formic Acid-Powered Micromotors for Fuel-Cell Technology*", National Conference on Recent Developments in Nanoscience and Nanotechnology (NCRDNN), Jadavpur University, 2019.
2. **A. K. Singh,** S. Rarotra, V. Pasumarthi, T. K. Mandal, D. Bandyopadhyay, "*Formic Acid-Powered Renewable Micromotors for Fuel-Cell Technology*", Frontiers in Chemical Sciences (FICS-2018), IIT Guwahati, 2018.
3. **A. K. Singh,** T. K. Mandal, D. Bandyopadhyay, "*Paper-based self-propelling microcleaners for efficient water purification*", 4th National workshop on MEMS/NEMS and Theranostic Devices (NWNTD-2018), IIT Guwahati, 2018.
4. **A. K. Singh**, K. K. Dey, A. Chattopadhyay, T. K. Mandal, D. Bandyopadhyay, "*Intelligent pH responsive chemo-magnetotatic microbots*", International Conference on Advances in Biological Systems and Materials Science in NanoWorld (ABSMSNW-2017), IIT BHU, Varanasi, India, 2017.
5. **A. K. Singh**, K. K. Dey, A. Chattopadhyay, T. K. Mandal, D. Bandyopadhyay, "*Alkaline pH taxis of iron-nanoparticle laden polymeric micromotors*", International Conference on Functional Materials (ICFM-2016), IIT Kharagpur, India, 2016.
6. **A. K. Singh**, K. K. Dey, A. Chattopadhyay, T. K. Mandal, D. Bandyopadhyay, "*Intelligent pH responsive chemo-magnetotatic microbots*", 3rd Indo-German workshop on Advances in Materials, Reactions & Separation processes, IIT Guwahati, 2016.
7. M. Bhattacharjee, V. Pasumarthi, J. Chaudhuri, **A. K. Singh**, H. Nemade, D. Bandyopadhyay, "Organic vapor detection using nanoparticle laden droplet and the effect of viscosity and vapor-source distance", IEEE ICEE-2016, IIT Bombay, India, 2016. DOI:10.1109/ICEmElec.2016.8074611.
8. M. Bhattacharjee, V. Pasumarthi, J. Chaudhuri, **A. K. Singh**, H. Nemade, D. Bandyopadhyay, "*Detection of organic vapors employing droplets having nanoparticles*", IEEE TechSym-2016, IIT Kharagpur, India, 2016. DOI: 10.1109/TechSym.2016.7872665.
9. **A. K. Singh**, T. K. Mandal, D. Bandyopadhyay, "*Chemically Powered Locomotion of Magneto-Catalytic Paper microjets*", International Symposium on Micro- and Nanomachines, Hannover, Germany, 2016.
10. **A. K. Singh**, T. K. Mandal, D. Bandyopadhyay, *"Self-propelling manganese dioxide nanoparticle-based Paper microengines",* Nanoparticle Assembly: From Fundamentals to Applications, Faraday Discussion, IIT Bombay, India, 2016.
11. **A. K. Singh**, K. K. Dey, A. Chattopadhyay, T. K. Mandal, D. Bandyopadhyay, "*Chemo-magnetophoretic motion of catalytic microbots*", CHEMCON 2015, IIT Guwahati, 2015.
12. **A. K. Singh**, S. Timung, D. Ranjan, S. Rarotra, T. K. Mandal and D. Bandyopadhyay, "*Vinegar Driven Micromotors for Nanoparticle Synthesis*", International Conference on. Emerging Materials: Characterization & Application (EMCA 2014), CSIR-CGCRI, Kolkata, India, 2014.
13. **A. K. Singh**, K. K. Dey, A. Chattopadhyay, T. K. Mandal, D. Bandyopadhyay, "*Chemo-magnetophoretic motion of catalytic microbots*", 6th Bangalore India Nano, Bangalore, India, 2013.
14. **A. K. Singh**, S. Sahu, B. Das, C. Banerjee, N. K. Jana, "*Cloning, Sequencing and Characterization of Exonuclease gene of Mycobacteriophage L1*", 98th Indian Science Congress, SRM University, Chennai, 2011.
15. **A. K. Singh**, N. K. Jana, "*Target identification by in-silico studies on NS1 proteins of bird flu virus*", HGM 2008 Satellite Symposium on Complex diseases: Approaches to gene identification and therapeutic management, Saha Institute of Nuclear Physics, Kolkata, 2008.
16. A. Sinha, **A. K. Singh**, A. Sett, G. Chatterjee. A. Sinha, S. Bhattacharya, S. Basu, "*Metabolic Stress in Plants due to exposure to mercuric chloride*", National Symposium on Microbial Diversity & Plant Health, B.C.K.V., Mohanpur, Nadia, 2007.
17. **A. K. Singh**, A. Sinha, A. Sett, G. Chatterjee, T. K. Ghosh, "*Bioreactor for animal cell culture*", National Conference on Impact of Biotechnology on development of human healthcare system, Heritage Institute of Technology, Kolkata, 2007.

CONFERENCES, SEMINARS, WORKSHOPS ATTENDED

1. National workshop on “*MEMS/NEMS and Theranostic Devices (NWNTD-2018)*”, IIT Guwahati (2018).
2. National workshop on “*MEMS/NEMS and Theranostic Devices (NWNTD-2015)*”, IIT Guwahati (2015).
3. National workshop on “*State-of-the-art in Microfluidics*”, IIT Guwahati (2015).
4. *“Research Conclave”*, IIT Guwahati (2015).
5. Symposium cum Workshop on “*Advances in Computational Biology and Computer Aided Drug Design*”, IIT Guwahati (2015).
6. International Conference on “*Emerging Materials: Characterization & Application (EMCA)*”, CSIR-CGCRI & NIT Durgapur, Kolkata, India, (2014).
7. Tutorial on “*Nanofabrication Technologies and Nanotech applications in Sensors and Conference-6th Bangalore India Nano*", Bangalore, India (2013)
8. IEEE workshop on “*Graphics and Presentation*”, IIT Guwahati (2013).
9. Workshop on "*Analysis of Biological Networks*", IIT Guwahati & DBT, India (2012).
10. 2nd International Conference on “*Advanced Nanomaterials and Nanotechnology (ICANN)*”, IIT Guwahati (2011).
11. National Workshop on “*Statistics in Genomics*”, Indian Statistical Institute, Kolkata, India (2010).
12. National Workshop on “*Bioinformatics and Molecular Modeling in Drug Design*”, Lucknow Biotech Park, India (2010).
13. Workshop on “*Bioinformatics in Genomics and Proteomics*”, IIT Kharagpur & DBT, India (2009).
14. International Workshop on “*Biologics: from discovery to development*”, International Centre for Genetic Engineering and Biotechnology (ICGEB) & International Union of Biochemistry and Molecular Biology (IUBMB), India (2009).
15. International symposium on “*Complex Diseases: Approaches to gene identification and therapeutic management*”, Saha Institute of Nuclear Physics, Kolkata, India (2008).

MEMBERSHIPS

- Affiliate member (ID: 582649), Royal Society of Chemistry (RSC), UK.
- Regular member (ID: EHO22), American Chemical Society (ACS), USA.
- Member (ID: S013) , Soft Materials Research Society (SMRS), India.

REFERENCES

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Acoustic Propulsion of Vitamin C Loaded Teabots for Targeted Oxidative Stress and Amyloid Therapeutics

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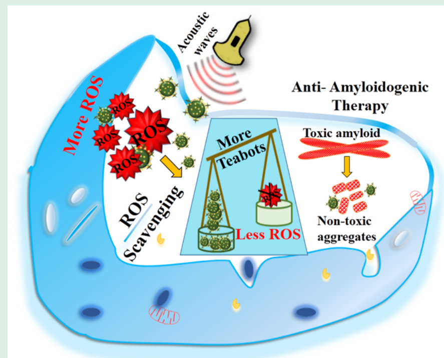
Supporting Information

ABSTRACT: We report the fabrication of ascorbic acid (AA) template nanomotors using buds of *Camelia sinensis*, undergoing fuel-free propulsion. The motors, namely, Teabots, display propulsion by converting the sound energy from the acoustic field into a mechanical one. The mesh-like structures of the anionic Teabots facilitate superior adsorption of ascorbic acid (AA-Teabots) undergoing a controlled release. The motors show antioxidant properties at the physiological pH range by scavenging intracellular reactive oxygen species. Interestingly, the percentage release of ascorbic acid is significantly higher under the influence of ultrasound exposure, as compared to the normal pH-dependent release. The motors were also efficient in the degradation of artificially synthesized toxic amyloid fibrils. The acoustic delivery of AA-Teabots could protect HEK-293 cells from oxidative injuries alongside preventing protein-aggregation derived diseases. Soon, such acoustic powered biocompatible AA-Teabots are envisioned to provide an attractive approach in proficient delivery and controlled release of therapeutic payloads at targeted zones.



KEYWORDS: *micromotor, Teabots, ascorbic acid, oxidative stress, amyloid fibers*

1. INTRODUCTION

Artificial self-propellers for the guided transport of therapeutic agents in the diseased body parts have been considered to be one of the most emerging areas of nanoscale research.[1,2] In this regard, while considerable attention has been given to the catalytic motors, various *in vivo* biomedical applications are expected to obviate the fuel-driven motions due to difficulties in modulating the motions.[3−7] In order to address these issues, efforts have been made to propel the motors under the remote excitations involving photons,[8] magnetic field,[9−11] acoustic waves,[12] and/or electric field.[13,14] In particular, recent studies have shown that the propulsion of nanomotors using safe MHz ultrasound frequencies (3−10 MHz) can enhance the prospects of the self-propelling objects in diverse biomedical applications causing insignificant harm to biological samples and the human body.[15] In a way, the rather innocuous acoustic waves enable a facile and fuel-free remote control on the motions of the self-propellers inside the biological systems alongside facilitating a controlled mechanical release.[16−20]

Apart from the driving force for the motion, another important factor in the design and development of micro- or nanoscale motors has been the materials of construction. While the majority of the first-generation self-propellers were rather synthetic devices[21−26] of late, the usage of biomaterials has become nearly mandatory at least for the motors used in the biological realm due to low cost of fabrication and biocompatibility.[27−30] In this regard, the idea of acoustically driven plant-based motors could emerge as a novel candidate for intracellular drug delivery of unharmed biological samples.[31−35] Further, under confinement and inside the viscous biological mediums, the practice of ultrasound exposure is expected to facilitate the remotely guided propulsions of such devices.[15,36] Given these backgrounds, herein, we report the fabrication of a nanomotor obtained from *C. sinensis*, which is capable of binding ascorbic acid (AA) on their surface and exhibits sustained release when driven acoustically toward oxidatively stressed cells. In particular, the proposed ultrasound (US)-propelled, L-Ascorbic acid (AA) functionalized Teabots aimed to act as an (a) excellent reactive oxygen species (ROS) scavengers in the human embryonic kidney (HEK-293) cell model and (b) efficient amyloid inhibitors.

The Teabots were synthesized from unfermented white tea buds of *Camellia sinensis* comprising of polyphenols from catechin family exhibiting antitumor and immune-stimulatory activities, lowering blood pressure, HIV treatment, and

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Graphene oxide nanohybrids for electron transfer-mediated antimicrobial activity†

Nayan Mani Das, *[ab] Amit Kumar Singh, *[b] Debdatta Ghosh[a] and Dipankar Bandyopadhyay *[ab]

The rapid increase in the prevalence of antibiotic-resistant bacterial strains poses a global health risk. In this scenario, alternative strategies are needed to combat the alarming rise in multidrug-resistant bacterial populations. For example, metal-incorporated graphene derivatives have emerged as model nanomaterials owing to their intrinsic antibacterial activity together with their biocompatibility. Interestingly, photon-activated phthalocyanine sensitizers have also shown promising physiochemical biocidal effects against pathogenic bacteria populations when conjugated with diverse nanomaterials. Herein, we report the facile synthesis of graphene oxide incorporated zinc phthalocyanine (ZnPc–GO) nanohybrids showing bactericidal activity against Gram-negative *Escherichia coli* (*E. coli*) cells, in the absence of any photo-excitation. The ZnPc–GO hybrid nanomaterials were synthesized by the *in situ* deposition of GO flakes on ZnPc-coated indium tin oxide (ITO) substrates. Two types of morphologically different ZnPc molecules, potato-chip-like α-phase ZnPc, namely ZnPc(A), and nanorod-like β-phase ZnPc(B), were used for the synthesis of the ZnPc(A/B)–GO nanocomposites. The interactions of GO with the underlying ZnPc(A/B) entities in the ZnPc–GO systems were investigated using multiple characterization techniques. It was observed that the GO flakes in the ZnPc(B)–GO nanocomposite possess stronger π–π interactions and thus show a more efficient electron transfer mechanism when compared with the ZnPc(A) counterpart. Furthermore, the *E. coli* bacterial cells with an electronegative surface demonstrated a profound adherence to the electron-withdrawing ZnPc(B)–GO surface. The death kinetics of bacteria with ZnPc(B)–GO were further investigated using surface potential mapping and Kelvin probe force microscopy (KPFM) analysis. Upon direct contact with ZnPc(B)–GO, the adhered bacterial cells showed outer cell deformation and membrane protein leakage, induced by a proposed charge-transfer mechanism between negatively charged cells and the electron-withdrawing ZnPc(B)–GO surface. These new findings may provide insights into the design of potential ZnPc–GO-based novel antimicrobial nanomaterials or surface coatings.

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1. Introduction

The abrupt rise in bacterial mutagenesis has led to widespread antibiotic-resistant infections worldwide, thereby posing a serious threat to public health.[1,2] Antibiotics are frequently administered to destroy or inhibit the growth of pathogenic bacteria. However, the exhaustive medical use and misuse of antibiotics has minimized their efficacy, which is attributed to the rapid emergence of antibiotic-resistant strains.[3] In order to avert the antibiotic-resistance crisis, researchers are exploring innovative combat strategies to restrain the spread of bacterial pathogens.[4–7] In the recent past, the bactericidal and bacteriostatic efficacy of nanomaterials has been widely studied as a promising alternative to conventional antibiotic-based treatment.[8–11] In this context, innovative methods like sensors,[12,13] bacteriophage-based systems,[14] thin film patterns[15] and microfluidics[16,17] have been employed for deactivation of pathogenic threats in the recent past. Amongst these methods, graphene-based nanomaterials[18,19] have been extensively used as antimicrobial agents for numerous biomedical applications in recent years.[20–22]

Graphene derivatives exhibit antimicrobial properties against a wide range of Gram-positive and Gram-negative pathogenic bacterial biofilms.[23,24] The ease of surface modification by direct incorporation of functional groups or extrinsic mesoscale materials on the graphene surfaces promotes their usage for antimicrobial therapy.[25,26] The antimicrobial efficacies of graphene derivatives can be further enhanced *via* surface functionalization with metals.[23] For example, metallic

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† Electronic supplementary information (ESI) available: Characterization of GO, XPS measurement data of samples, work function calculations, and FESEM images of bacteria. See DOI: 10.1039/c9na00272c

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Formic acid powered reusable autonomous ferrobots for efficient hydrogen generation under ambient conditions†

Amit Kumar Singh, [a] Saptak Rarotra,[b] Viswanath Pasumarthi,[b] Tapas Kumar Mandal[ab] and Dipankar Bandyopadhyay *[ab]

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We report the design and development of a self-propelling ferrobot composed of a collection of iron nanoparticles (FeNPs). While the propulsive thrust required for the chemotactic migration of the ferrobots was generated through the ejection of hydrogen (H_2) bubbles due to the reaction of aqueous formic acid (FA) with FeNP clusters on the motor surface, the presence of ferromagnetic FeNPs assured "on-the-fly" remote guidance using an external magnetic field. The speed of chemotactic migration of the motor was found to be highest when the rate of reaction was maximum in a 30% (v/v) aqueous FA solution. Directed propulsion of the ferrobot was also achieved by tuning the chemical gradient and magnetic field potentials across the ferrobot. Unlike previously reported systems where the FA decomposition is associated with either the production of the greenhouse gas CO_2 or the use of high temperature, the proposed self-propelling ferrobots could react with aqueous FA fuel at room temperature to produce pure H_2 gas. Thus, the ferrobots could further be employed to power a proton exchange membrane (PEM) fuel cell to rotate a portable toy fan. In this situation, while the pure H_2 gas required for the PEM cell was generated through the reaction of the FA solution with self-propelling ferrobots, O_2 gas was also produced by the catalytic decomposition of peroxide fuel using the ferrobots. Interestingly, the ferrobots and FA fuel could easily be regenerated for their repeated use towards the continuous production of pure hydrogen. The experiments uncovered the potential of the proposed ferrobots not only for the on-demand power supply to portable devices but also as a single-step commercial process to produce pure H_2 under ambient conditions and devoid of greenhouse gas emission.

1. Introduction

Nature has inspired scientists for ages in the design and development of mesoscale self-propelling objects.[1–3] In this direction, locomotives capable of performing chemotaxis,[4,5] phototaxis,[6,7] galvanotaxis,[8,9] sonotaxis,[10,11] or magnetotaxis[12,13] are expected to cause a paradigm-shift in a number of futuristic applications such as theranostics,[14–16] lab-on-a-chip operations,[17] non-invasive surgery,[18] energy harvesting,[19] sensing,[20,21] and environmental remediation.[22–24] While chemically powered motors emulate microorganisms to show *in situ* migrations due to the gradient of chemical potential surrounding them,[3–5,25] external stimuli[25] induced migrations establish high-precision remote control on these motions. In particular, the past few decades have experienced a rapid progress in the syntheses and applications of various types of such locomotives also known as micro- or nano-bots.

In this regard, the bubble-propulsive thrust generated on the surface of a microbot due to the decomposition of the peroxide fuel is perhaps the most widely studied system for artificial chemotaxis.[4,5,25] Of late, many of the microbots could show self-propulsion when water,[26,27] hydrazine,[28] inorganic acids,[29,30] alkalis,[30] citric acid,[24] sodium borohydride ($NaBH_4$),[19] acetylene,[31] urea,[32] glucose,[33] halogens[34] and alcohols[35] are employed as the fuel sources. Interestingly, Pt-black/Ti Janus micromotors have been employed in recent past to synthesize hydrogen (H_2) gas using sodium borohydride ($NaBH_4$) fuel to cater to the needs of clean energy harvesting.[19]

In this study, we report the design and development of a self-propelling object composed of a collection of iron nanoparticles (FeNPs), namely, the 'ferrobot'. The ferrobot is capable of high-speed chemotaxis through the bubble-propulsive thrust generated due to H_2 bubble ejection on the surface owing to the reaction of aqueous formic acid (HCOOH, FA) fuel with FeNPs under ambient conditions. The presence of FeNPs also provides a remote magnetic control on the chemotactic migrations of the

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† Electronic supplementary information (ESI) available: Vibrating sample magnetometry (VSM) hysteresis loop of motors, characterization of iron formate, and movies of micromotor motion. See DOI: 10.1039/c8ta02205d

Soft Matter

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Boolean-chemotaxis of logibots deciphering the motions of self-propelling microorganisms†

Tamanna Bhuyan,[a] Mitradip Bhattacharjee,[a] Amit Kumar Singh, [a] Siddhartha Sankar Ghosh*[ab] and Dipankar Bandyopadhyay *[ac]

We demonstrate the feasibility of a self-propelling mushroom motor, namely a 'logibot', as a functional unit for the construction of a host of optimized binary logic gates. Emulating the chemokinesis of unicellular prokaryotes or eukaryotes, the logibots made stimuli responsive conditional movements at varied speeds towards a pair of acid–alkali triggers. A series of integrative logic operations and cascaded logic circuits, namely, AND, NAND, NOT, OR, NOR, and NIMPLY, have been constructed employing the decisive chemotactic migrations of the logibot in the presence of the pH gradient established by the sole or coupled effects of acid (HCl-catalase) and alkali (NaOH) drips inside a peroxide bath. The imposed acid and/or alkali triggers across the logibots were realized as inputs while the logic gates were functionally reconfigured to several operational modes by varying the pH of the acid–alkali inputs. The self-propelling logibot could rapidly sense the external stimuli, decide, and act on the basis of intensities of the pH triggers. The impulsive responses of the logibots towards and away from the external acid–alkali stimuli were interpreted as the potential outputs of the logic gates. The external stimuli responsive self-propulsion of the logibots following different logic gates and circuits can not only be an eco-friendly alternative to the silicon-based computing operations but also be a promising strategy for the development of intelligent pH-responsive drug delivery devices.

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1. Introduction

Chemotaxis of microorganisms often follows a series of complex logical motions while executing different tasks.[1–3] Of late, the bio-mimetics of these natural processes have ushered the development of next-generation artificial micro or nanoscale machines for drug transport, targeted release, and other bio-medical applications.[4,5] In the recent era of automation, the programmable logic gates have also made frequent appearances in bio-photonics,[6,7] synthetic circuits,[8] smart-sensing,[9] diagnostics and therapeutics,[10] bio-computation[11] and energy harvesting.[12] A number of previous studies have shown logic gate based sensing,[13] information processing,[14] drug-release,[15] detection,[16] preparation of circuits[17] and chemical synthesis.[18] Further, the colorimetric,[19] chemi-luminescent,[20] fluorescent[21] or electrochemical[22] signals emulating a Boolean response have also been found to be suitable for a host of cutting-edge applications.

In this regard, one of the long standing challenges has been the decoding of logical operations, which are in general manifested as chemotactic migrations of microorganisms.[1,2,23] It is now well known that a series of complex but systematic logical operations have been executed for (a) bacteria moving towards a food source, namely, 'positive' chemotaxis, and away from the poison, namely, 'negative' chemotaxis;[1,2,24] (b) movement of sperms toward the egg during fertilization;[25] (c) locomotion of neutrophils against micro-organisms as a defence mechanism;[26] and (d) neuronal imaging or other motility behaviours of multicellular frameworks.[27,28]

Emulating these natural processes, of late, NAND, XOR, AND, OR, NOR, and NOT logic circuits have been developed employing the living cells.[29,30] Further, various biomaterials are employed to develop artificial logic gates targeting important applications. For example, (i) DNA-gates have been employed to identify the behaviours of base pairs;[31] (ii) RNA-bio-circuits are used for the advancement of electronic biosensors;[32] (iii) microbial logic systems have been developed for environ-mental remediation;[33] and (iv) reversible gates, such as CNOT, FREDKIN, or TOFFOLI, are constructed based on the motility of *Physarum* sp.[34] Interestingly, the logic gates with more than one input have been reported to be essential for cells to identify a combination of multiple signals, which is found to significantly

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Magnetic Field Guided Chemotaxis of iMushbots for Targeted Anticancer Therapeutics

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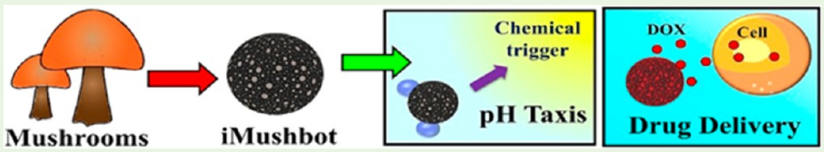
Supporting Information



ABSTRACT: We report controlled migrations of an intelligent and biocompatible "iMushbot" composed of *Agaricus bisporus*, mushroom microcapsules coated with magnetite nanoparticles. The otherwise randomly moving microbot could meticulously direct itself toward and away from the acid- and alkali-rich regions with the help of acid, acidic catalase, and alkali stimuli, emulating the chemotaxis of microorganisms. Although the catalytic decomposition of peroxide-fuel in alkali engendered the directed alkali taxis toward higher pH region, decomposition of peroxide fuel by the acidic catalase activity led to directed acid taxis toward the lower pH region. The presence of magnetite nanoparticles not only helped in improving the "activity" of the motor through the heterogeneous catalytic decomposition of the peroxide fuel but also provided a remote magnetic control on the chemotaxis. The mesoporous iMushbots having negative ζ-potential could easily be loaded with the cationic anticancer drugs, which were magnetically guided toward the cancerous cells to cause apoptosis. The iMushbots exhibited higher degree of drug retaining capacity inside alkaline pH and showed facile drug release preferentially in the lower pH environments. The experiments show the potential of the iMushbots in retaining and transporting drugs in alkaline medium such as human blood and releasing them in acidic medium such as the cancerous tissues for cell apoptosis.

KEYWORDS: *micromotor, mushroom, chemotaxis, cancer, drug delivery*

1. INTRODUCTION

Development of artificial self-propelling objects having faculties to deliver medicines in the targeted locations of human body for the therapy of life threatening diseases have attracted lot of attention in the past few decades.[1−4] A number of seminal contributions led to the fabrication of diverse micro or nanoscale robots responsive toward various stimuli which include chemical[5,6] or enzymatic[7,8] reactions, photonic[9] or acoustic[10,11] waves, thermal[12] or concentration gradients,[13−15] and electric[16,17] and/or magnetic fields,[18,19] among others. These locomotives have also been envisioned to serve diverse uses ranging from target isolation,[20,21] sensing,[22,23] therapeutics or diagnostics,[24−26] microsurgery,[27,28] transport of payloads,[29,30] and mitigation of pollutants through detoxification.[31] Presently, the biocompatible micro or nanosystems empowered with the specialties of the smart inorganic, organic, or biomaterials are poised to take over as the futuristic drug carriers to achieve therapeutic benefits with minimal side effects. The said mesoscale active systems are expected to show multifunctionalities such as the loading of the hydrophobic or hydrophilic drugs alongside the capacity to stimuli sensitive release of the same.[32] However, in the current scenario, one of the major challenges is to synthesize nontoxic biocompatible micro/nanoscale motors capable of in situ transport and on-demand release of the life-saving drugs in the targeted locations.

Thus, far, a host of state-of-art fabrication methodologies, for example, angled electron beam evaporation,[10] sputter coating,[9,12] photolithography,[30] template-assisted deposition,[33] or 3D printing[34] were employed to fabricate the multifunctional self-propelling objects. The specialties of micro- or nanoscale particles,[9,12] rods,[1,10,11] tubes[20,21] of noble metals, carbonaceous materials[16,31] and/or synthetic polymers[14,15] have been employed to impart various functionalities to the locomotion. The issues related to the cost-effectiveness and biocompatibility have also been addressed by synthesizing biomotors form the plant tissues.[35−38] For example, millimeter-scale robots[36] synthesized from the enzyme-rich plant tissues were found to self-propel in the peroxide fuel while catalase differentially decomposed hydrogen peroxide around the motor to engender the chemically powered locomotion. Microscale calcified biotubes[35] in the idioblast cells of the Dracaena sp. plant leaf coated with Fe−Ti layer were employed to perform drug

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Self-spinning nanoparticle laden microdroplets for sensing and energy harvesting†

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Exposure of a volatile organic vapour could set in powerful rotational motion a microdroplet composed of an aqueous salt solution loaded with metal nanoparticles. The solutal Marangoni motion on the surface originating from the sharp difference in the surface tension of water and organic vapour stimulated the strong vortices inside the droplet. The vapour sources of methanol, ethanol, diethyl ether, toluene, and chloroform stimulated motions of different magnitudes could easily be correlated to the surface tension gradient on the drop surface. Interestingly, when the nanoparticle laden droplet of aqueous salt solution was connected to an external electric circuit through a pair of electrodes, an ~85–95% reduction in the electrical resistance was observed across the spinning droplet. The extent of reduction in the resistance was found to have a correlation with the difference in the surface tension of the vapour source and the water droplet, which could be employed to distinguish the vapour sources. Remarkably, the power density of the same prototype was estimated to be around 7 μW cm^{-2}, which indicated the potential of the phenomenon in converting surface energy into electrical in a non-destructive manner and under ambient conditions. Theoretical analysis uncovered that the difference in the ζ-potential near the electrodes was the major reason for the voltage generation. The prototype could also detect the repeated exposure and withdrawal of vapour sources, which helped in the development of a proof-of-concept detector to sense alcohol issuing out of the human breathing system.

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1. Introduction

Integrating the special features of nanotechnology in microfluidic applications is expected to realize the dream of fabricating simple, economical, portable, and multi-tasking devices in the near future.[1–3] In this direction, a number of recent works have shown the capability of minuscule self-motile objects[4–7] to serve as sensors as well as delivery vehicles by harnessing chemical energy,[8] surface tension force,[9] photonic[10] or acoustic[11] or thermal excitations,[12] and external electric[13] or magnetic fields.[14] The modern sensors composed of field effect transistors,[15] electrochemical detectors,[16] photo-detectors,[17] piezo-materials,[18] nano-cantilevers,[19] and plasmonic[20] or microfluidic devices[21] have also been benefiting significantly from the specialties of nanowires, nanoparticles, or nanotubes of functional materials.[22–24] Importantly, these micro or nanoscale objects are also found to be equally efficient in converting the different other forms of energy into electrical energy while accomplishing their stipulated tasks.[25–28] The power density of many of the conventional macroscopic processes[3,29] such as radio frequency (~1 μW cm^{-2}), solar irradiation (~100 mW cm^{-2}), ambient illumination (~100 μW cm^{-2}), thermoelectric (~60 μW cm^{-2}) or vibration generators (~10–800 μW cm^{-3}), or ambient air flow (~1 mW cm^{-2}) are found to be comparable with some of the newly developed micro or nanoscale energy harvesters.[1–3] The vision here is to develop applications involving miniaturized self-motile systems, which can harvest high density power in addition to performing tasks simultaneously as a sensor, mixer, separator, or transporter. Herein, we show that a simple nanoparticle laden water microdroplet could simultaneously sense and distinguish different types of organic vapours alongside harvesting high density power.

The previous studies indicate that so far the most popular and robust method to detect vapours or gases has been to employ the electrochemical sensors composed of semiconductor metal oxides.[30,31] The modern sensors based on conducting[32,33] and doped polymers[34] or carbon black[35] have also been prepared for this purpose and faced the limitation with

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Magnetically guided chemical locomotion of self-propelling paperbots†

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Self-propelling microjets with multimodal chemical and magnetic controls for motion were prepared from printed waste papers coated with MnO_2 nanoparticles. Because the magnetic remote control was infused from the ferromagnetic coating of the printer ink, the nanoparticles decomposed peroxide fuel to induce locomotion by ejecting oxygen bubbles through a microjet. The paper microjets could be loaded with fluorescent rhodamine 6G (R6G), a model payload, before being remotely guided through an external magnetic field inside a fluidic environment. The reported methodology provides an economic, scalable, and biodegradable scheme for the design and development of bubble-propelled microengines, which are capable of directed locomotion.

1. Introduction

Synthesis of artificial swimmers of macro, micro or nanoscale dimension has emerged as an active area of research owing to their significant technological importance.[1–8] Synthetic self-propelling objects are expected to perform diverse tasks harnessing different sources of energy for their locomotion, which include chemical energy,[1,9] surface tension force,[7,10] photonic[11] or acoustic[12–14] or thermal excitations,[15,16] and external electric[17] and magnetic[18] fields. In particular, the minuscule self-propelling tubular microjets[19] are envisioned to serve as cargo transporters,[20] drug delivery modules,[21] agents for environmental remediation[8] and building blocks for lab-on-a-chip devices.[22]

In general, microjets are fabricated by rolling membranes of relatively inert materials such as titanium, polycarbonate and aluminum oxide coated with active metal films.[23] The rolling is done in such a manner that the inert layer forms the outer shell, while the active layers stay inside the hollow core. Interestingly, when the tubular microjets are placed in a bath of peroxide fuel (dilute hydrogen peroxide – H_2O_2), the active layers catalytically decompose H_2O_2 to produce a stream of oxygen (O_2) bubbles from the hollow inner core. The recoiling of the microengine during the ejection of the O_2 bubbles provides the required thrust for the motion. Previous studies suggest that the fabrication of microengines involves multistep processes and needs facilities such as template-assisted electrodeposition, electron beam evaporation and photolithography.[23–25] Clearly, a simpler and cost-effective methodology to fabricate microengines is more preferred option to provide further impetus to the applicability and commercialization of these devices.

In this direction, inspired by recent research studies on paper-based microfluidics,[26] we report the synthesis of a paper microjet, which shows locomotion under chemical and magnetic triggers. We employed a simple methodology to fabricate the microengines by rolling the printed waste papers in which the chemical sensitivity was obtained by depositing MnO_2 nanoparticles on the surface of the paper. These microengines were inherently sensitive towards an external magnetic field because of the presence of ferromagnetic printer ink on their surface. The target here is to recycle the printed waste papers for the fabrication of microengines after minor chemical modifications. Arguably, the results reported in this study, for the first time, highlight the directed propulsion of the paper microjets in combination with the motion triggered by the chemical excitation. An application for payload transport was demonstrated, where the paper microjets could be loaded with fluorescent rhodamine 6G (R6G) as a model cargo, and they could be remotely guided through an external magnetic field inside a fluidic environment. The reported methodology provides a scalable as well as biodegradable scheme for the design and development of bubble-propelled microengines, which are capable of directed propulsion through remote guidance.

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† Electronic supplementary information (ESI) available: Detailed experimental protocol, characterization and movies of motor motion along with detailed explanation. See DOI: 10.1039/c5ra10159j

2. Experimental methods

2.1. Materials and methods

Hydrogen peroxide (H_2O_2; 50%), ethanol (C_2H_6O; 99.9%), fluorescein sodium salt ($C_{20}H_{10}Na_2O_5$), potassium permanganate ($KMnO_4$) and Amberlite IR 120 were obtained from Merck

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Graphene based multifunctional superbots

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ABSTRACT

A versatile graphene coated glass microswimmer displayed directed motions under the influence of applied electric field, chemical potential gradient and external magnetic field. The directed chemical locomotion took place from the region of lower to higher pH with speed ~13 body lengths per second due to asymmetric catalytic decomposition of dilute hydrogen peroxide across the motor surface. The negative surface potential of graphene coated motor developed an electrical double layer in an alkaline medium which in turn engendered electrophoretic mobility towards anode when the external electrostatic field was applied. Inclusion of sparsely populated ferromagnetic iron nanoparticles on the surface of the motor offered the magnetic remote control on the motion. The coupled *in situ* and external controls enabled the motor to develop complex motions in diverse open and confined environments. For example, the motor could approach, pick-up, tow, and release a heavy cargo inside microchannel. Remarkably, the motor (~67 µg) could successfully drive out a ~1000 times heavier payload (~0.67 mg) displaying the ability to overcome the drag force of ~2619 pN with the help of coupled *in situ* and remote guidance.

1. Introduction

Directed migrations of the microorganisms are stimulated by diverse external or *in situ* excitations which include the electric field – galvanotaxis, magnetic field – magnetotaxis, light – phototaxis, or chemical potential gradient – chemotaxis. Emulating these events, miniaturized self-propelling artificial objects can be synthesized for the applications such as targeted drug delivery [1–3], non-invasive penetration [4,5], high precision sensing [6,7] and biomedical devices [8]. In particular, the chemically empowered nano-locomotives [9–17] are envisioned to perform complex *in vivo* or *ex vivo* tasks with functional efficacy similar to their biological counterparts. The recent advancements on the fabrication of self-propelling objects are directed towards this end where the nanoscale objects have shown motions under various internal and external triggers such as the gradients of magnetic field [18–20], electric field [21], temperature [22], surface tension [23,24], and electromagnetic [25] or acoustic [26] potential. The existing challenges are to infuse biocompatibility, attain multimodal *in situ* and remote control on the motion, tune the directionality at a faster response time, and add functionalities specific to the engineering processes.

Interestingly, the present decade have also experienced another important paradigm-shift in which the metal based applications have been transformed into the biocompatible

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Capillary Force Mediated Flow Patterns and Non-Monotonic Pressure Drop Characteristics of Oil-Water Microflows

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We report the capillary and frictional force mediated transitions of morphologies of an oil-water flow inside a microchannel using experiments and computational fluid dynamic simulations. A number of steady and time-periodic flow patterns were reported with the variations in the interfacial tension, exchange of inlets, flow ratio, and viscosity ratio of the phases. Transitions from slug to plug to droplet to stratified flow patterns were obtained by tuning the interfacial tension. Progressive reduction in the interfacial tension transformed big slugs into smaller plugs, plugs into droplets, and droplets into a stratified flow pattern. Interestingly, the simulations uncovered a non-monotonic and nonlinear reduction in pressure drop with the decrease in interfacial tension. The change in the pressure drop was correlated to the variation in the slug, plug, or droplet frequency of water at the outlet. The variations in the pressure drop were also associated with the transition from dripping to jetting of water droplet ejection near the channel inlet. Apart from the interfacial tension, the viscosity stratification across the phases was also found to play an important role in converting the slug flow patterns into smaller plugs or droplets. The study also reports the parametric space in which the droplet flow patterns could be obtained inside a microchannel tuning the flow and viscosity ratios of the phases alongside the interfacial tension. The reported transitions of flow patterns and the pressure drop characteristics can be of significance in improving the efficiency of future microfluidic devices.

Keywords: microchannel, oil-water flow, interfacial phenomena, flow patterns

INTRODUCTION

Interfacial morphologies of two-layer flows in microfluidic devices have been studied extensively in recent times because of their diverse applications in the areas of bio-analysis,[1] multiphase extraction,[2] emulsification,[3] MEMS devices,[4] and microreactors,[5] among many others. Microfluidic devices are considered superior to their macroscopic analogues primarily because of the availability of higher surface-to-volume ratio, smaller throughput leading to easier control of the operating parameters, and reduction in operating cost in the processes where costly chemicals are in use.[6] Microscale two-phase flows also manifest exciting interfacial flow patterns in the form of slug, plug, core-annular, stratified, bubbly, and drop-dispersions, which have fascinated many researchers. Previous studies uncovered a host of exceptional behaviours of two-phase microflows, for example: generation of microscale droplets or bubble dispersions, or chaotic interfacial patterns,[7,8] folding and swirling of interfacial threads,[9,10] formation of vesicles,[11] mixing,[12] and particle synthesis.[13] A number of review articles have summarized the various scientific and technological aspects of the two-layer flows inside microchannels.[6,14]

Among all the multiphase configurations, gas-liquid flow through a ‘Y’ or ‘T’ shaped microchannel with a circular or rectangular cross-section has been the most widely studied.[15–21] In contrast, the understanding of similar liquid-liquid configurations has started gaining attention only in recent years, because of their complex but remarkable physics. A review by Joseph et al.[22] ably distinguished the salient features of liquid-liquid flows as compared to similar gas-liquid flows while discussing the open issues in oil-water flows. It is now well understood that the typical viscosity and the density stratifications, together with weaker capillarity at the interface, make the dynamics of liquid-liquid flows rather unique. In particular, the lower interfacial tension can facilitate smaller drop formation, which can be of significance for microreactor design, formation of microemulsions, and enhanced heat and mass transfer applications.

Apart from focus on the technological aspects, the fundamental mechanisms of drop, plug, or slug release of the two-phase flows inside microfluidic devices have attracted significant attention in recent years.[23–25] The ‘dripping,’ ‘jetting,’ and ‘squeezing’ mechanisms of the droplet, plug, or slug formation due to the interplay between the inertia, capillary, and shear forces have also been investigated.[23,24] For example, Menech et al.[24] studied the steady and transient characteristics of squeezing, dripping, and jetting transitions with the variations in the capillary number. In the dripping mechanism, the droplets or plugs are ejected near the inlet, whereas the jetting involves extension of a fluid thread downstream of the channel before droplets are pinched off from the main thread. The flow morphologies originating from the dominance of capillary, viscous, or weak inertial forces and the subsequent pressure drop characteristics have also been studied in detail.[25–29] For example, Zhao et al.[26] showed the transitions in flow patterns at the T-junction of a microchannel with the Weber number by varying the superficial velocities of the oil-water phases. Tice et al.[12] showed the variations in plug flow patterns with phase viscosity changes in a liquid-liquid configuration

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Multimodal chemo–magnetic control of self-propelling microbots†

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We report a controlled migration of an iron nanoparticle (FeNP) coated polymer micromotor. The otherwise diffusive motion of the motor was meticulously directed through an *in situ* pH-gradient and an external magnetic field. The self-propulsion owing to the asymmetric catalytic decomposition of peroxide fuel was directed through a pH gradient imposed across the motor-surface, while the magnetic field induced an external control on the movement and the speed of the motor. Interestingly, the sole influence of the pH gradient could move the motor as high as ∼25 body lengths per second, which was further magnified by the external assistance from the magnetic field. Applying a magnetic field against the pH directed motion helped in the quantitative experimental estimation of the force-field required to arrest the chemotactic migration. The influence of the coupled internal and external fields could halt, steer or reverse the direction the motor inside a microchannel, rotate the motor around a target, and deliver the motor to a cluster of cells. This study showcases a multimodal chemical–magnetic field regulated migration of micro-machines for sensing, transport, and delivery inside a fluidic environment.

Introduction

Recent advances in the fabrication and characterization of small scale devices have inspired the design and development of artificial micro-swimmers,[1–8] mimicking the movements of motor-proteins[9,10] or the motility of microorganisms.[11,12] These synthetic self-propelling objects have important applications in sensing, micro- or nano-fluidic devices, drug delivery,[13] directed self-assembly,[14] and in the design of nano-machinery.[15] In particular, the catalytic decomposition of aqueous hydrogen peroxide (H_2O_2) has been widely employed for the conversion of chemical energy into mechanical motion.[16–21] An electromagnetic field can also be employed to gain external control over the migrations of the nanoscopic objects.[22–27]

Recent studies have revealed that micromotors containing gold (Au) and palladium (Pd) nanoparticles on the surface could undergo pH-directed motion inside a bath of peroxide fuel.[28,29] However, the existing challenge is to gain a multimodal internal–external control on the movements of the synthetic objects to improve their technological usefulness. In this study, we report chemo-magneto-taxis of micro-swimmers in sizes ranging between 30–250 μm, which exhibited directed motion under the coupled influence of an *in situ* pH gradient and an external magnetic field. The self-propelling motion of the motor facilitated by a pH gradient was remotely controlled through an external magnetic field. The external field could meticulously direct the motor during any stage of its migration by steering, halting or even reversing its direction of motion. A host of controlled motions including a reciprocating motion inside a channel and delivery of the motor to a cluster of target animal cells were shown by tuning the internal pH-gradient and the external magnetic field. While the pH-gradient alone could produce a remarkably high speed of ∼25 body lengths of the motor per second (∼5 mm s^{-1}), the external magnetic field could lead to an explosive increase in the speed when applied in the same direction. The application of the magnetic field against the pH gradient induced migration could decelerate and momentarily stop the motor in both open and confined environments. The minimum magnetic field required to halt a motor undergoing motion due to the pH trigger is arguably the first quantitative and experimental measurement of the magnitude of the force-field of the chemical locomotion. Concisely, this study unveils the pathways to remote-control a self-propelling micro-machine for sensing, transport, and delivery applications.

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