

Colloidal robotics

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Robots have components that work together to accomplish a task. Colloids are particles, usually less than 100 μm , that are small enough that they do not settle out of solution. Colloidal robots are particles capable of functions such as sensing, computation, communication, locomotion and energy management that are all controlled by the particle itself. Their design and synthesis is an emerging area of interdisciplinary research drawing from materials science, colloid science, self-assembly, robophysics and control theory. Many colloidal robot systems approach synthetic versions of biological cells in autonomy and may find ultimate utility in bringing these specialized functions to previously inaccessible locations. This Perspective examines the emerging literature and highlights certain design principles and strategies towards the realization of colloidal robots.

Colloidal robots (CRs) are autonomous particulate machines that employ the ‘sense–plan–act’ paradigm in colloidal conditions, with the goal of deploying microscopic robotic systems into new environments. Autonomy in this setting is defined as the ability of machines to make decisions (or ‘compute’) without external actuation and supervision. Such computations can be performed by incorporating (1) sensors for information input, (2) central processing units for logic computation and data storage, (3) communication channels for information transfer, (4) modes of actuation and locomotion and/or (5) on-board energy harvesting and/or storage units into colloidal microparticles in a modular manner (Fig. 1a–c).

The field of CR research shares similar goals with other efforts to miniaturize both electronics and robotics. CRs can be thought of as a subset of ‘micro-robotic’ systems, a term that has been used to describe sub-millimetre devices and centimetre-scale devices that may or may not be externally controlled¹. CRs are also related to ‘smart dust’², silicon-based microsystems that integrate sensing, computation and communication. Unlike CRs, these microsystems typically have

volumes larger than 1 mm³, which limits their potential applications in dispersion systems. As a result, successful implementations of these microscale electronic systems remain elusive in medicine except for environments where volume is less of a constraint, such as the mammalian digestive tract³, or where devices can be implanted^{4,5}. Properties of swimming colloidal motors, especially autonomous ‘active colloids’ are also of interest for CRs⁶. However, the colloidal motors field is primarily interested in propulsion over other computations or outputs, and has not typically investigated modular designs. Therefore, a CR that swims may also be an active colloid, and an active colloid that is modular and programmable may also be a CR.

These related technologies illustrate the trade-off that is central to the design of CR: size versus capability. Smart dust particles are relatively complex, but large. Colloidal motors are small, but relatively simple and less modular. Both small size and high complexity are being pursued in the design of CRs through the careful use of material properties, nanofabrication techniques and clever control strategies. In this Perspective we review recent breakthroughs in these areas relevant

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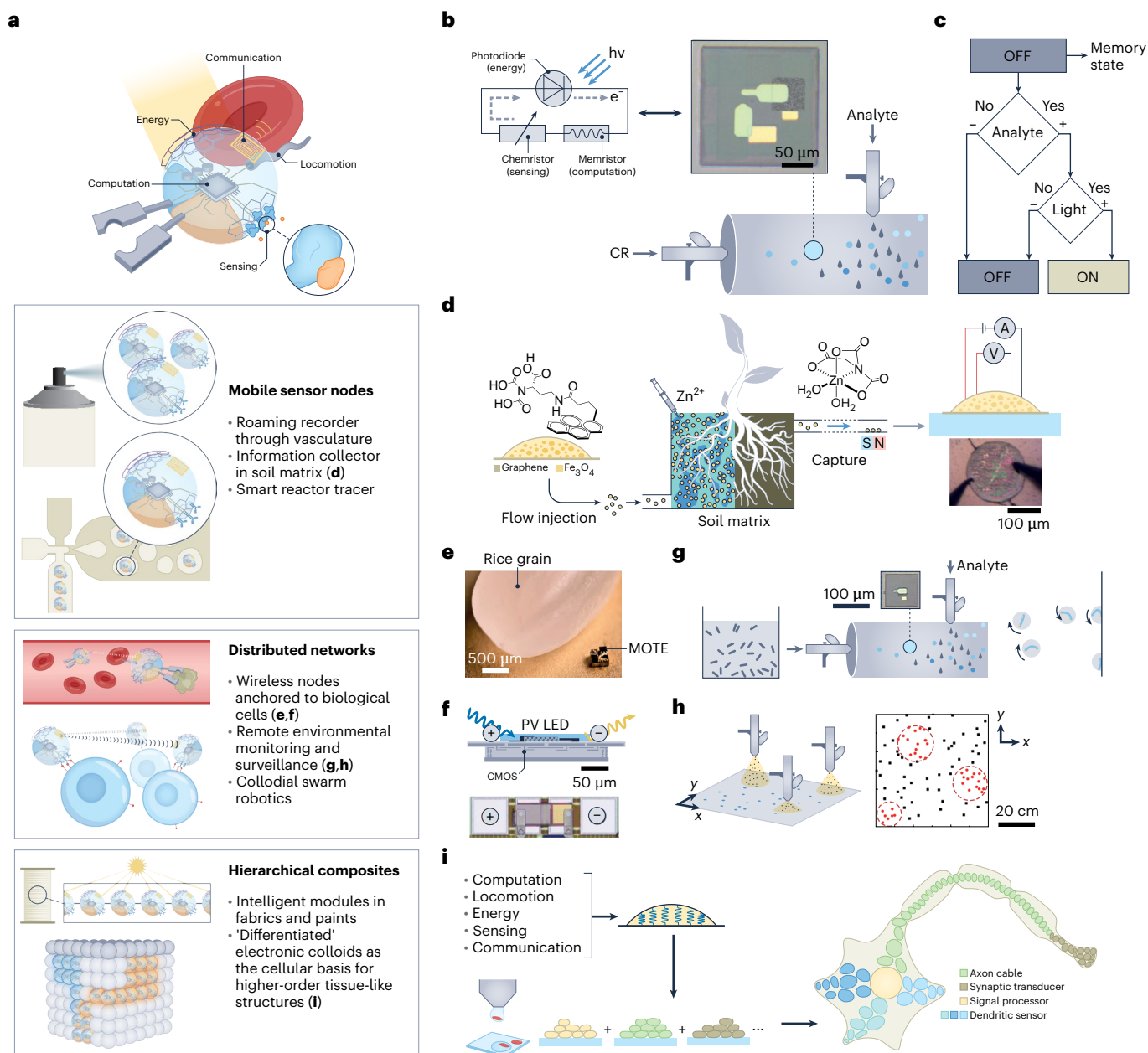


Fig. 1 | Concepts and prototypes of CRs. **a**, Schematic of a CR. **b**, Schematic of an early CR prototype being used for analyte and light detection inside a vapour chamber⁹. Left: electrical circuit diagram of the CR. The photodiode converts light into current, which turns on the memristor if, and only if, the chemristor detects a prescribed analyte. Right: top-view optical micrograph of the CR used⁹. **c**, Block diagram of the CR summarizing the combinational logic. The initial memory state OFF changes to ON only in the presence of both chemical and light signals⁹. **d**, Schematic of the deployment of surface-functionalized colloidal electronic microparticles as chemical probes to detect and record the presence of various species, such as Zn^{2+} ions in the soil matrix. Symbols S and N represent the south and north poles of the externally applied magnetic field, respectively. Inset: optical micrograph of the colloidal electronic particle during electrical readout¹⁰. **e**, A laser-powered 0.04 mm³ 16 nW wireless sensor microscale opto-electronically transduced electrode (MOTE) with integrated central processing and optical communication capable of accurate cellular temperature measurement in vitro¹⁵.

f, A 250 $\mu m \times 57 \mu m$ 1 μW MOTE capable of capturing and encoding neural signals before transmitting the encoded signals through an optical interface²⁰. **g**, Experimental schematic showing remote detection and memory storage using CRs aerosolized from their original colloidal suspension⁹. **h**, Left: schematic of large-area sensing using CRs: analytes (yellow) are sprayed at three locations over an area with dispersed CRs (teal squares). Right: digitized positions of distributed CRs. Turn ON responses (red) can be seen for those CRs exposed to the analyte, as opposed to those that have not (OFF state, black)⁹. **i**, Schematics illustrating the concept of assembling various derivatives of CRs, each specializing in a different function, into higher-order tissue-like structures. Tertiary structures can be formed from ensembles of differentiated CRs to accomplish more complex tasks at a larger scale, for instance, a synthetic neural signal transduction system. Panels adapted with permission from: **b,c,g,h**, ref. 9, Springer Nature Limited; **d**, ref. 10, Springer Nature Limited; **f**, ref. 20, IEEE. Credit: **e**, Flickr, Joseph Xu, Univ. Michigan College of Engineering.

to CR development. Many of the advances discussed might better be described as almost-CRs; we see them as enabling technologies. We also examine new applications for CRs, and conclude with a proposal of research strategies to address these applications.

Application space of colloidal robotics

CRs collect, manipulate, store and exchange information without external supervision⁷. When the highly dispersed nature inherent to a colloidal system interfaces with the autonomy and programmability

that stem from robotic and electronic systems, new capabilities emerge. We broadly classify these applications into two functional areas: (1) CRs as mobile sensory nodes and distributed information networks in confined systems, and (2) CRs as fundamental building blocks for hierarchical, higher-order functional structures.

On the one hand, CRs can be used for signal processing and transduction from enclosed spaces such as mammalian vasculature⁸, microfluidic channels and chemical/biosynthetic reactors⁹, as well as for recording from remote locations such as oil and gas conduits, rock crevices, water bodies¹⁰, soil matrix¹⁰ or the atmosphere. Operating at cellular dimensions enables an intimate, microscopic paradigm of human–matter interaction (Fig. 1d–h).

Comprising individual building blocks acting as independently controllable electronic devices, CRs also transform the way one typically thinks about functional materials. The particulate form-factor allows CRs to be integrated into coatings, fibres and many other traditional material architectures. Manufacturing electronic colloids at scale further promises the synthesis of hierarchical structures¹¹ out of individual CR particles—analogue to the way biology builds tissues out of individual cells, functional domains that consist of various combinations of differentiated CRs could be established. Some examples could be networks of artificial neurons (that is, amplified electrical signal transduction) (Fig. 1i) or synthetic myocardium (that is, with pulsatile mechanical actuation). Given these properties, CRs are well poised to solve longstanding problems in medicine, distributed computing and other disciplines.

Exploiting colloidal properties for micrometre-scale robots

As robotic devices shrink to sub-millimetre sizes, they begin to take on behaviours of colloidal systems. Typical colloidal phenomena such as Brownian motion, aggregation and sedimentation can be seen as challenging obstacles to overcome or as enabling features to be exploited for CR design. Their small size allows them to be easily dispersed. First, their thermal energy can lead to substantial displacements in position and orientation, resulting in random walks (Brownian motion) characterized by linear and rotational diffusion coefficients. As a result, CR position and orientation may change rapidly and unpredictably, which poses a challenge for a robot travelling from point A to point B if Brownian motion is commensurate with its propulsion speed. On the flip side, this means that sufficiently small CRs will diffuse to spread through a medium freely using only ambient thermal energy (for example, to monitor conditions in a chemical reactor).

A second key factor that contributes to the dispersibility of CRs is their relatively high surface-area-to-volume ratio, which leads to drag forces that are strong compared with their gravitational counterparts, ultimately resulting in slow sedimentation rates of colloidal particles. ‘Slow’ in this case depends on context: a similar settling velocity can mean that CRs stay suspended in a swimming pool for days but sediment out of a blood vessel in a few seconds. Understanding CR sedimentation behaviour is important not only in aqueous environments, but also in air, for aerosolizable electronic systems^{9,10,12}.

The relatively high CR surface area also means that particle–particle and particle–surface interactions are particularly strong. If these interactions are attractive, they can lead to potentially irreversible aggregation and surface fouling. Particle aggregates sediment out faster, and CRs stuck to surfaces will not diffuse. In some cases, the aggregation and adhesion are undesirable: one would not want CRs that monitor a chemical reactor to aggregate or adhere to the reactor surface. CRs sprayed onto a field of crops should not aggregate with each other, but ideally would adhere to the surface of the crops. On the other hand, aggregation may be a desirable characteristic in creating hierarchical assemblies of CRs. Fortunately, there are strategies to control the strength of these types of interaction, such as electrostatic¹³, steric or entropic stabilization¹⁴. The ability to program and adjust

these interparticle potentials can be exploited to realize controlled (and dynamic) self-assembly within an ensemble of CRs, achieving complex systems-level functions and desirable outcomes (for example, artificial platelets in blood).

Materials-enabled colloidal robotic modules

Here we break down CR operations into five fundamental modules (that is, computation, sensing, communication, locomotion and energy management) and discuss promising technologies that enable each of these five CR ‘organelles’.

Logic and computing

Computation is at the core of most robotic systems. It processes input signals from sensors, stores relevant information and generates output signals in response. Silicon-based electronics are the cheapest, most reliable option for building integrated circuits. Several microsystems that approach the size of a CR have been demonstrated using silicon chips for computation (Supplementary Table 1), targeting biomedical applications¹⁵, environmental sensing¹⁶ and object verification¹⁷ with power consumptions ranging from 0.4 nW to 10 μ W.

Low-dimensional materials have been explored extensively in recent years given their tunable electronic structure and high carrier mobility¹⁸. Transistors based on 2D materials and carbon nanotubes with promising device characteristics have been fabricated, enabling the development of more complex circuits¹⁹. The key advantages of using low-dimensional materials for CR applications stem from their mechanical flexibility and the chemical stability of their electronic performance.

Sensors

Sensing units enable robotic systems to be aware of their location, environment and neighbours, and subsequently trigger signalling and downstream responses. In the context of CRs, a need for modular integration renders electric sensors the most relevant. Beyond silicon-based devices, many materials (for example, 2D materials and metal oxides) can be used to make microscale electrical sensors for CR-type systems, resulting in a fairly broad field that has been subject to extensive analyses elsewhere¹⁸. Such technologies have been used to implement chemical, temperature, light and voltage sensors on devices of approximately 100 μ m in each dimension^{9,12,20–22}.

Communication

Communication between microsystems using electromagnetic, ultrasonic, optical and chemical means is summarized in Supplementary Table 2. Radiofrequency communication is the standard choice between electronic systems, yet antennas become increasingly inefficient when scaled down. Most reported radiofrequency systems at the sub-millimetre scale thus communicate data passively by back-scattering the incoming signal, especially for medical applications²³. Similarly, the lowest power consumption in ultrasound systems—which is best suited to aqueous media such as water and biological tissues—is achieved when information is transmitted back passively. This approach has been used to build a neural recording device⁴ capable of transmitting data to a receiver 9 mm away. Light can also be used to communicate between CRs, either by active light emission using light emitting diodes (LEDs) or through indirect means such as modulating the fluorescence or reflection of light²⁴. In contrast to radiofrequency systems, optical systems scale well down to the 1–100 μ m range, although such communication is only feasible through transparent media such as water or air.

A less conventional and little explored method of communication is through the emission and diffusion of molecules that serve as information carriers. Chemical communication is prevalent in nature, although it is slow and has a low bandwidth, in contrast to the previously discussed modalities²⁵. For applications with relaxed timing and sparse

information content, however, this may be a viable option. One way to implement CRs with chemical communication is to equip them with molecule reservoirs that open only when certain conditions are met^{26,27}.

Besides environmental constraints, the decision on which technology should be implemented also depends on the communication modality, such as centralized base-to-unit communication or decentralized unit-to-unit communication. An asymmetric communication strategy that combines both centralized and decentralized modalities, where information is relayed from a more complex base unit through a network of simpler CR units, could reduce energy costs by leveraging the near-field nature between adjacent CRs.

Actuation and locomotion

The ability to autonomously actuate gives CRs the ability to perform short-range position and orientation adjustments. To implement actuators at the microscale, mechanisms such as chemical adhesion and surface forces become appealing choices, as they scale more favourably than most mechanical grippers²⁸. To possess true autonomy, these actuators must be locally stimulated. Promising candidates include polymer- and gel-based actuators²⁹ and thin film bimorphs^{21,30}.

Scaling physical systems down to microscopic dimensions causes surface-born interactions, such as surface tension, drag and adhesion, to dominate over volumetric bulk effects, such as mass and inertia^{28,31}. One strategy would be to emulate similarly sized microorganisms³², which have mostly adapted appendages such as cilia, flagella or pseudopods to operate in these conditions³³. These appendages enable microorganisms to interact dynamically with the environment for active feeding, mating and avoiding predation. Researchers are increasingly able to investigate these strategies by modelling them in macroscale robotic devices^{34,35}. For example, limbless locomotion of macroscale organisms (such as sand-swimming lizards and snakes) in granular substrates³⁶ provides insights into locomotion in low-Reynolds-number or inertia-free regimes relevant for CRs.

Despite the progress in creating microscale actuators that behave much like flagella and cilia, successful microscale implementations of locomotion strategies have primarily involved driving rigid micro- and nanoparticles by several methods, including magnetic, electric, acoustic and optical fields, as well as by numerous chemical reactions. These devices, typically named nanomotors, micromotors, microswimmers or colloidal motors, are well studied and have been reviewed in great depth elsewhere⁶.

In the context of CRs, two characteristics are crucial for effective propulsion: (1) the ability to operate autonomously and (2) the ability to integrate with other CR functions. Integrating these propulsion mechanisms with other functions on a CR, such as sensors, logic and memory, remains a challenge. One solution would be to seek out propulsion mechanisms that could interface directly with on-board circuitry. To that end, electrochemical actuators integrated with simple microelectronic circuits have recently been employed as 'legs' on a walking robot²¹. Although these machines are not yet autonomous (they require a laser to be aimed at different targets on the device's surface in an alternating pattern), on-board batteries and clock circuits²⁷ could conceivably be integrated in the next generation of devices to make autonomous micro-walkers³⁷. Another well-studied propulsion mechanism, self-electrophoresis³⁸, shows promise for integration with microelectronics. Here, motion is generated as charged ions move in solution around a micromotor from an ion source to an ion sink, driven by either a spontaneous chemical reaction³⁹ or a photodiode⁴⁰. Electrons flowing through the body of the motor form a complete circuit. Further sensors and logic could be integrated into these micromotors to toggle this circuit. For example, responsive hydrogels could expand and contract to control the exposure of a catalytic patch to a chemical fuel⁴¹; dynamic shape changes (for example by the aforementioned chemical actuators) could be used for navigation⁴².

Energy harvesting and storage

Electric energy is critical to power functions such as information processing, actuation and communication, and its supply represents one of the greatest challenges for CRs (Fig. 2). Figure 2e plots the energy demand for standard computation and the energy available as a function of the characteristic length of devices. A picolitre (1,000 μm^3) energy module composed of the best commercial lithium-ion batteries would only be able to power a 100 nW load for roughly 25 s (2,500 nW s)⁴³. This is sufficient for standby circuits but not for communication. Alternatively, supercapacitors can offer energy and power densities of up to 0.4 nJ μm^{-3} and 10 pW μm^{-3} , respectively⁴⁴, while regular dielectric capacitors offer much lower areal energy densities of 25–400 fJ μm^{-2} but reach much higher power densities of up to 400 nW μm^{-2} due to the quick discharge⁴⁵—ideal for short bursts of communication or other brief, high-power tasks. The Ragone plot (Fig. 2c) visualizes this trade-off between power and energy density.

To achieve partial or complete autonomy, previous CR demonstrations have often relied on external energy harvesting. Biotic and abiotic fuel cells have been explored for the direct conversion of chemical energy, providing a typical power density of 1 pW μm^{-3} and a supply voltage of 0.5 V. Radiofrequency energy harvesters can deliver a high power density of 10–400 pW μm^{-2} at a few millimetres distance and total transmitted powers in the range of 50–2,000 mW. However, these numbers are only rough guides as smaller coils (<100 μm) typically operate in the near-field regime and downscaling probably results in lower efficiencies. The photovoltaic effect has been used extensively in autonomous microsystems for ambient and in vivo applications²⁰. Given a picolitre colloidal device, about 60 nW of power could be generated, assuming a 10% efficient solar cell operating in direct sun light ($\sim 1 \text{ nW } \mu\text{m}^{-2}$).

Finally, there are emerging technologies that may be used to power colloidal systems in the future. One promising candidate is solvent-interaction-induced electrical energy harvesting (or solvo-voltaics)⁴⁶, which is particularly relevant to CRs because it taps into their surrounding solvent environment⁴⁷. Alternatively, (mechanical) triboelectric generators could deliver power densities in the range of 0.1 pW μm^{-3} and high voltages on the order of 10 V (refs. 48).

Given the above strengths and weaknesses of each energy source (Fig. 2c,d and Supplementary Tables 3 and 4), a CR will probably integrate multiple energy storage and harvesting modules to accommodate different electric functions and situations. Assigning appropriate electricity generators to specific purposes is the key to navigating the CR across a potentially anisotropic energy landscape (Fig. 2f).

CR systems integration

The goal of CR integration is to build complex, autonomous systems from individual building blocks, analogous to combining individual organelles (for example, nucleus as CPUs, mitochondria as energy harvesters, flagella as actuators and so on) into higher-order cells (Fig. 3).

Top-down technologies

A unique feature of colloidal systems is their small final device size (approximately $100 \times 100 \mu\text{m}^2$ in-plane area) compared with traditional devices, such as a $2 \times 2 \text{ cm}^2$ CPU. This feature relaxes the defect density tolerance for the wafer-scale fabrication of CRs to be considered as an industrially viable process. Once a monolithic top-down integration approach is selected for the colloidal system, laboratory-grade methods with slightly higher failure rates may still be amenable for large-scale device production.

Lithography. Top-down lithographic fabrication is unrivalled for manufacturing complex, small parts over large areas. Today, patterns as small as 10 nm can reliably be made in industrial or academic settings via photolithography or e-beam lithography⁴⁹. Building CRs with a mature process technology like 180 nm CMOS in a commercial foundry,

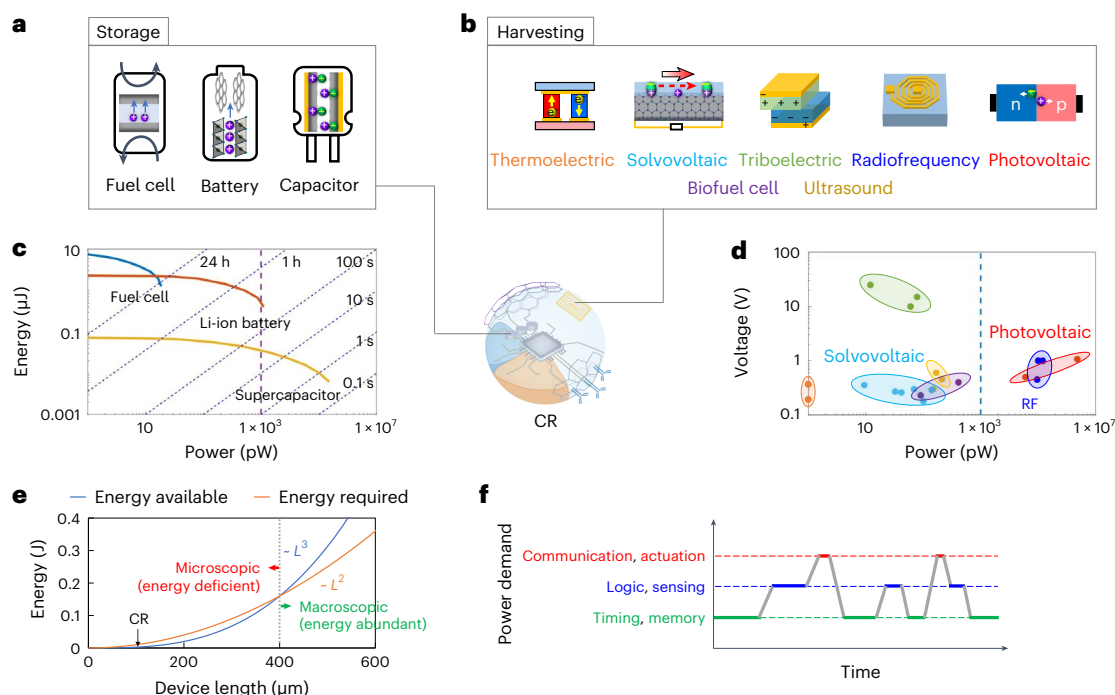


Fig. 2 | Energy harvesting and storage techniques relevant for CRs.

a,b. Potentially feasible electrical power sources on the scale of CRs, categorized by mechanism, as storage (**a**) or harvesting (**b**) devices. **c.** Ragone plot of energy storage devices for a limited volume of 1 pL. The parallel dashed lines represent the times of discharge. For example, if we discharge a Li-ion battery at 1 nW power, we follow the purple vertical line up and encounter the curve for battery between 1 hour (h) and 100 s, which means that the battery will be fully discharged in several hundred seconds. The plot is based on a conservative estimation of the energy and power densities, with data from ref. 43, which also shows that with an optimized configuration and structure of electrodes, the maximum accessible

power density may be boosted by 10 times⁴³. **d.** Voltage–power characteristics of energy harvesting devices. Data points are colour coded to devices shown in **b**. The power output is estimated for a structure with a surface area of $600 \mu\text{m}^2$, which is the maximum attainable for a $10 \times 10 \times 10 \mu\text{m}^3$ CR. The solar cell is supposed to have 10% efficiency and operate under 1 Sun illumination (about 1 kW m^{-2}). **e.** Estimated energy required for standard computation (20 transistors per μm^2 with each transistor consuming 5 nW at 3.4 GHz for a computation time of 10 s) and electrochemically stored energy ($2.5 \text{ nJ } \mu\text{m}^{-3}$) plotted as a function of device length (denoted as L). **f.** Example duty cycles of CRs utilizing different energy sources at different power demands (log scale).

for example, simultaneously inherits the cost, scale and complexity benefits currently available. Yet even with this powerful tool, key challenges remain. First, lithography makes planar patterns, and it is not obvious how to build 3D parts such as arms, legs or bodies for robots. Second, robots do not just sit idly on a wafer: they need to be released into the world at large. This requires striking a balance between locking parts in place during top-down fabrication to achieve high resolution and fully freeing the resulting devices later. Third, lithography can only pattern materials on the wafer. Many other functional materials for robot components require additional methods for heterogeneous integration. Miskin et al. recently demonstrated a promising step in augmenting lithographic fabrication²¹. Photolithography was used to pattern simple silicon circuits, wiring and electrically controllable actuators massively in parallel. To make legs and arms, the authors leveraged origami concepts, building 2D lithographic patterns that could later self-fold into 3D arms and legs. With this mixture of methods, they demonstrated swarms of 10,000 electronically integrated microscopic robots deployed at yields of ~90% (ref. 21).

Printing. With recent advances in precision printing, additive manufacturing emerges as a viable and inexpensive alternative to traditional manufacturing methods. Unlike lithography, additive printing builds complex scaffolds through deposition, layer by layer. State-of-the-art printing processes achieve a minimum feature size of about $1 \mu\text{m}$ with alignment accuracies of several micrometres and printing speeds of about 1 m s^{-1} (Fig. 3a,b)⁵⁰. For features that do not require nanometre precision, printing is inexpensive and compatible with a large variety of materials (nano- and microparticles

of various shapes, thermoplastics, ceramics, polymer composites, low-dimensional materials and so on). In practice, various ‘inks’ have been used to establish active or passive circuit elements, such as transistors, capacitors, resistors, memristors and so on (Fig. 3c)^{10,41}, suggesting that printing represents a complementary tool for CR fabrication⁵¹.

Heterogeneous integration. The top-down assembly of separate as-made modules, categorized here as heterogeneous integration, encompasses a wide spectrum of techniques from traditional integrated circuit methods such as wire bonding, flip chip reflow soldering, through-chip via and wafer bonding to recent materials transfer innovations such as fluidic self-assembly⁵², dry transfer printing (Fig. 3d)⁵³ and wafer-scale layer-by-layer assembly⁵⁴. These approaches allow optimization of the best material for each building block and decouple fabrication constraints such as high-temperature processing or chemical compatibility for each building block (Fig. 3d).

Bottom-up technologies

Defect self-assembly. It is well understood that material fracture can be controlled using a prescribed strain field, and the governing Griffith criterion has been demonstrated to operate down to the nanometre limit within 2D lattices such as graphene⁵⁵. Recently, Liu et al. successfully implemented the strategy of strain-induced guided ‘autoperforation’ on 2D material sheets (monolayer graphene, MoS_2 and hBN), demonstrating the feasibility of using controlled defect self-assembly as a reliable and scalable nanofabrication method

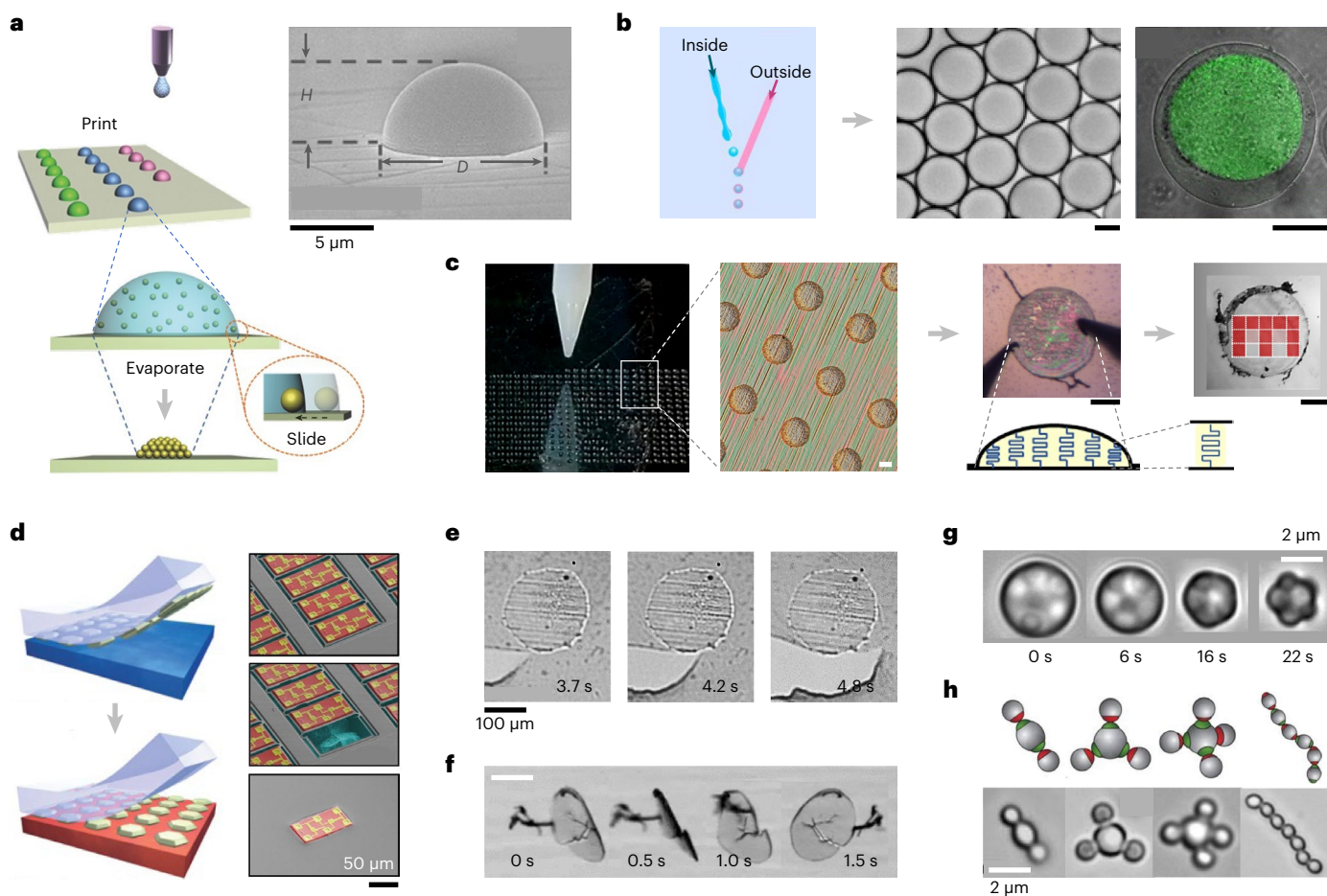


Fig. 3 | Selected examples of device integration relevant for colloidal robotics applications. **a**, Schematic of the process of assembling inkjet-printed polystyrene beads into aggregated dome-shaped arrays. Inset: scanning electron micrograph (SEM) image of one such printed polystyrene micro-dome⁷⁷. H , height; D , diameter. **b**, High-throughput in-air microfluidic inkjet printing of micro-suspensions of multiple compositions⁵¹. Left: schematic of the technique that generates core-shell structures. Middle: spherical particle suspensions viewed via optical microscopy. Right: multi-material (alginate-dextran-tyramine in H_2O) core-shell particles. Scale bars, 50 μm . **c**, Inkjet printing of CR particles consisting of graphene-encapsulated memristor arrays. Left: optical image (inset: micrograph) of the printed microparticles. Middle: top view of the electrical characterization of a single microparticle (after solution lift-off and recapture). Bottom-right inset: schematic cross-section of the vertical memristor array. Top right: a 15-bit microparticle memristor array written via electronic probes. An 'M'-shaped vertical conductivity map (red, ON; clear, OFF) overlaid on top of the microparticle tested. Scale bars, 50 μm (ref. 10).

d, Left: schematic of transfer printing⁵³. Right: false-colour SEM images of an array of devices, shown in sequence, after undercut etching (top), after removal of a single device from the donor wafer (middle) and after transfer printing of this device onto a receiving substrate (bottom)⁷⁸. **e**, Optical micrographs showing fracture and crack propagation of the graphene layers around a single polymer disk¹⁰. **f**, Optical micrographs of an autoperforated graphene-encapsulated polymer disk rotating and translating in solution in a laminar flow field. Scale bar, 100 μm (ref. 10). **g**, Optical micrographs during evaporation of the toluene, showing the emulsion encapsulation process⁵⁸. **h**, Bright-field images (bottom) and schematics (top) showing colloidal 'molecules' self-assembled from 'patchy' particles preprogrammed with complementary green and red DNA patches⁵⁹. Panels adapted with permission from: **a**, ref. 77, Wiley; **b**, ref. 51 under a Creative Commons licence CC BY-NC 4.0; **c**, ref. 10, Springer Nature Limited; **d**, ref. 53, Springer Nature Limited, and ref. 78, National Academies of Science; **e**, **f**, ref. 10, Springer Nature Limited; **g**, ref. 58, AAAS; **h**, ref. 59, Springer Nature Limited.

applicable to the production of electronic colloids¹⁰. This autoperforation strategy forms the basis for various types of autonomous micro-electronic devices capable of analyte detection and memory storage (Fig. 3c)^{10,41}.

Origami- and kirigami-based folding methods. Given that most electronic material systems and fabrication methods are optimized for circuits in 2D, self-folding strategies, such as those inspired by origami and kirigami, have opened the door to creating complex 3D structures using planar fabrication techniques³⁰. Recent work has demonstrated successful folding of polymers, silicon oxides, shape-memory alloys and 2D materials³⁰ at various length scales⁵⁶. Origami robots capable of autonomous actuation³⁰, supervised locomotion and reprogrammable self-assembly have also been fabricated en masse⁵⁷. With its

inherent scale-free character, origami as a framework for metamaterial design has found utility across many geometric scales (nanometre, micrometre, millimetre).

Controlled colloidal self-assembly into electronic clusters. Self-assembly is a process in which a system of pre-existing components spontaneously organizes into an ordered structure with no external supervision. Well-defined structures can be synthesized via processes such as emulsion encapsulation (Fig. 3g)⁵⁸ and DNA hybridization (Fig. 3h)⁵⁹. Alternatively, colloids can be precisely organized and linked by a 'lock and key' mechanism⁶⁰, patchy surface hydrophobicity⁶¹ or wetting forces (colloidal fusion)⁶². On the other hand, dynamic, or out-of-equilibrium, assembly has also been examined extensively⁶³. Although the literature on the self-assembly of colloids is constantly

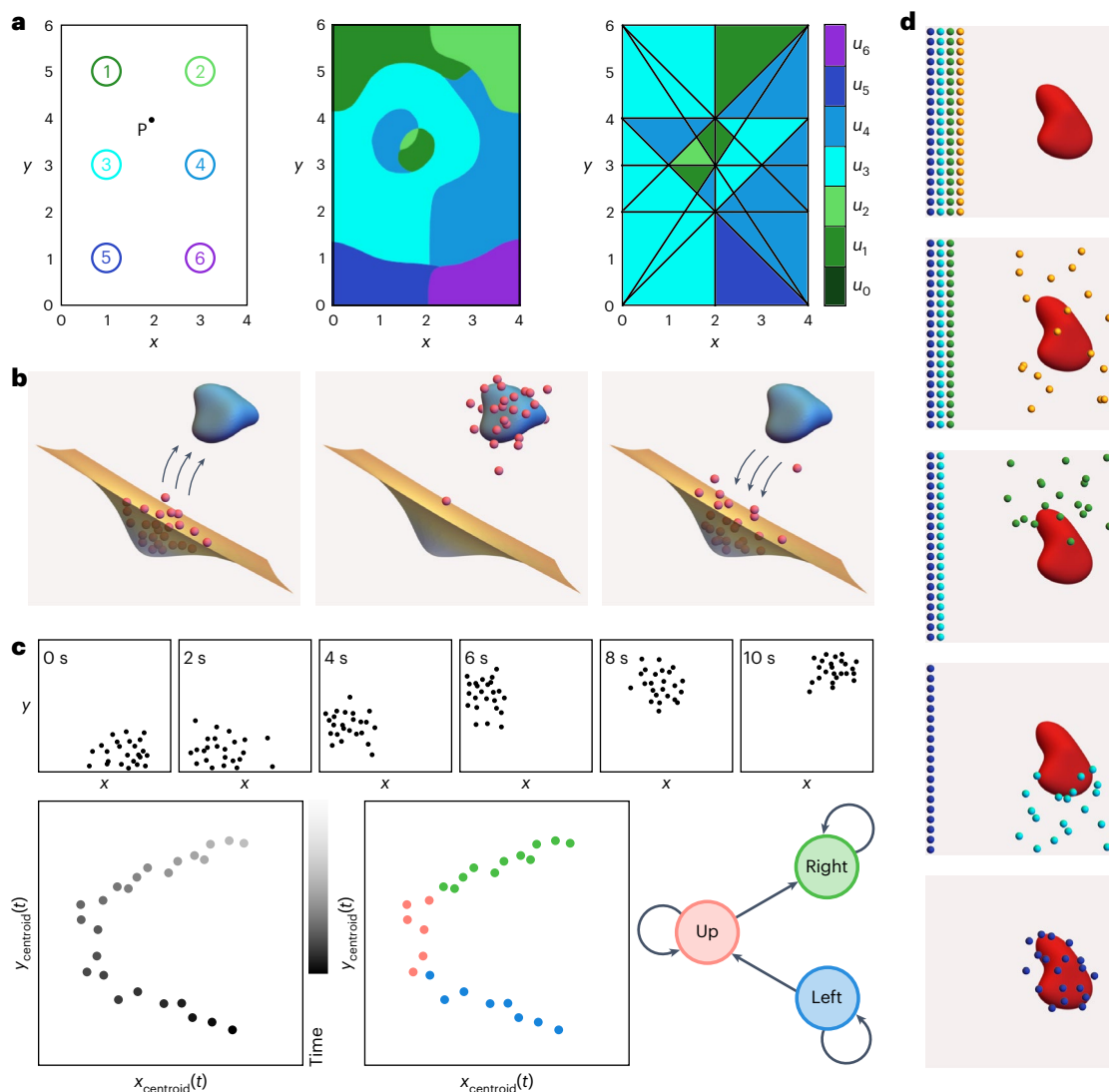


Fig. 4 | Distributed computation and control in systems of CRs. **a**, Model-based control. Left: a planar environment with six different chemical sources. A CR can locomote by choosing one of seven different control modes: being attracted towards a chemical potential at one of the six different sources ($u_1, u_2, u_3, u_4, u_5, u_6$) or staying still (u_0). The goal of the device is to move towards point P. Middle: a continuous control policy that indicates which control mode (shown with different colours) to use at each location in the 2D environment. The discrete control output is a function of continuous input variables $u(x, y)$. Right: a discretized control policy, where the environment is divided into regions that can be differentiated using chemical comparators. The discrete control output is a function of discrete input regions $u(r)$. **b**, The ensemble of CRs (pink) needs to find an energy source (blue object), but in doing so diffuses. The CRs

therefore need to then exploit environmental characteristics (the yellow pocket in the environment) to reassemble themselves into a non-diffuse ensemble. **c**, Simulated position for individual motile robots at distinct time (t) (top) can be simplified to the ensemble's motion (bottom left), which can then be abstracted as three modes of transport (bottom middle) that can be encoded in a finite state machine (bottom right) on-board each CR. The small number of behaviours that describe the complex dynamics lead to minimal computation elements that the CR will need to employ to execute control. **d**, Illustration of a sequence of rollouts that could enable CRs to learn in a massively parallel fashion. Starting from an initial set of randomized rollouts, the policy can be updated through inter-robot communication, eventually leading to a policy that successfully encapsulates an object. Panel **a** adapted with permission from ref. 72, Springer Nature Limited.

growing⁶⁴, programmed-clustering^{32,65} of individual parts into CRs with logic and full electronic autonomy is still in its infancy⁶⁶.

Systems of autonomous CRs

Programmable matter refers to a physical computing medium composed of simple, homogeneous entities that is instantly reconfigurable and robust to failures⁶⁷. One can differentiate between passive and active programmable matter systems. In passive systems, individual entities rely on structural properties and environmental interactions as means of locomotion and reconfiguration. They may, in some cases, have limited computational abilities to make decisions and communicate. Prominent examples of passive systems include population

protocols (which can be viewed as an abstraction of chemical reaction networks)⁶⁸, DNA/molecular computing and tile self-assembly models⁶⁹ and slime moulds⁷⁰.

CRs fall into the category of active systems, in which the individual entities can control their actions and/or movements to achieve tasks. Other examples of physical active systems arise in the context of swarm robotics and self-reconfigurable modular robotics⁷¹. Control is traditionally implemented through reconfigurable computing to calculate optimal or near-optimal decisions on the basis of an objective. However, as a system shrinks, reconfigurable computing takes a greater percentage of the physical volume and energy budget, eventually becoming impractical. Consequently, CR control will be constrained

to use limited computational resources, and its control authority (in the form of simple circuits⁹, logical operators and individual elements of non-volatile memory)¹⁰ will be internally represented in terms of discrete material states—despite operating in and interacting with continuous environments. Figure 4a shows a control architecture—the result of a hybrid, optimization-based approach⁷²—that is capable of moving a CR to a specified location. The robot autonomously achieves its goal in continuous space, using only discrete actuation, discrete sensing and a few logical operators⁷².

If individual CRs can be controlled, ensembles of such agents should be able to achieve broader objectives. The control of systems of simple agents has been explored in many areas, such as the ‘short-cut bridging’ behaviour of army ants, where a stochastic, distributed algorithm enabled particles to self-assemble into bridges optimized for length and cost⁷³. Similar global behaviours have been demonstrated in algorithms for compression or expansion, and separation or integration of heterogeneous agents⁷⁴.

Emergent behaviour and learning in CRs

To make control decisions, a CR directly modelling physical states is impractical. Instead, such model can be discovered as an emergent phenomenon, implying that an ensemble of robots must detect, and exploit, emergent behaviours to accomplish its goals^{32,65,75}. In Fig. 4b, CRs search for a blue object but diffuse as they do so. They must detect and exploit the yellow energy potential to avoid dispersion. Instead of each agent modelling specific states such as position, velocity, energy levels and details of its surroundings, the robots can make decisions on the basis of broader characteristics, such as the characteristics of neighbours, as in self-organizing particle systems.

To discover emergence, the ensemble might assume a very limited model that involves only a small number of unknown behaviours. In Fig. 4c, a simulated ensemble of individually stochastic CRs on average moves in only a few ways. There are three distinct dynamic behaviours (translating left, up and right) that the ensemble can choose on the basis of sensory inputs. Once these emergent behaviours are identified⁷⁶, they can be used in different combinations to execute tasks.

Despite the computational limitations of individual CRs, an ensemble can be used in parallel reinforcement learning. As illustrated in Fig. 4d, devices of different colours (representing distinct rollouts) attempt to encapsulate an object. Rather than one agent attempting the task many times, many agents attempt the task once and communicate their results to each other. Later rollouts of agents update their policies in response to rewards reaped by earlier attempts⁸. Even with limited computation, these examples illustrate how CRs can exhibit emergent intelligence. Instead of being a consequence of exquisite sensing, actuation and computation, autonomy can be accomplished as the result of flexible interactions between an ensemble of CRs and their environment.

Outlook

The field of colloidal robotics builds on the existing concepts of smart dust and microrobots, whereas smart dust particles have been non-mobile microsystems in the cubic millimetre range, and microrobots are often ‘supervised agents’ that require external control. CRs are the next logical step in this development and raise the bar in terms of specifications. At a nanolitre size or less, they are at least 1,000 times smaller than smart dust or microrobots, and are designed to not settle out in fluidic environments, enabling a variety of new applications.

This emerging field is highly interdisciplinary and building CRs cannot be trivially accomplished by simply scaling down or up contemporary technologies designed for larger or smaller systems. There are length-scale-specific physical phenomena that make this infeasible and require rethinking of fabrication approaches and operational designs. To do so, it helps to generalize the concept of CRs as a microsystem

that combines a subset, if not all, of the following building blocks: (1) computation, (2) sensing, (3) communication, (4) locomotion/actuation and (5) energy harvesting.

Integrating all these functions in a nanolitre ($10^6 \mu\text{m}^3$) volume is a challenging task. While monolithic integration of all components is desirable, no single materials class will suffice to realize all functions. A heterosystem featuring a diverse set of materials that best fit each function is ultimately desirable for assembling autonomous CRs. The path forwards is clear: we simultaneously need a diverse set of material constructs (components such as sensors, timers and actuators) that bring together highly specialized, yet energy-efficient, functions and modular system designs that allow one part of the CR to work with the other. Standardization of inputs and outputs is therefore important to foster collaboration between materials science researchers working on individual modules.

As micro- and nanoscale machines develop as solutions to challenging problems, it is important to highlight the risks and repercussions of colloidal robotics. The burden is on the scientists, engineers and business owners who use the technology, as well as policymakers, to uphold the integrity of the ethical and responsible usage of the technology. As we initially deploy CRs in non-laboratory settings, enclosed spaces should be explored first (with appropriate filtering, where the larger sizes of CRs relative to nanoparticles will be advantageous). One additional advantage of using functional 2D (or other nanostructured low-dimensional) materials as opposed to traditional bulk conductors and semiconductors for electronic circuitry is the reduced potential toxicity concerns (in that a single atomic layer of a 2D material, for example, can deliver similar functionality to its bulk counterparts with much less material). This highlights the necessity of exploring non-traditional materials for these forthcoming applications using CRs. In this Perspective we have highlighted several pioneering efforts that have used CRs as distributed information probes in several otherwise inaccessible locations. Many opportunities lie ahead for disruptive innovations in medical, environmental monitoring and surveillance, as well as other creative uses of such technologies.

References

1. Jafferis, N. T. et al. Untethered flight of an insect-sized flapping-wing microscale aerial vehicle. *Nature* **570**, 491–495 (2019).
2. Warneke, B. et al. Smart dust: communicating with a cubic-millimeter computer. *Computer* **34**, 44–51 (2001).
3. Kalantar-Zadeh, K. et al. A human pilot trial of ingestible electronic capsules capable of sensing different gases in the gut. *Nat. Electron.* **1**, 79–87 (2018).
4. Seo, D. et al. Wireless recording in the peripheral nervous system with ultrasonic neural dust. *Neuron* **91**, 529–539 (2016).
5. Piech, D. K. et al. A wireless millimetre-scale implantable neural stimulator with ultrasonically powered bidirectional communication. *Nat. Biomed. Eng.* **4**, 207–222 (2020).
6. Fernández-Medina, M. et al. Recent advances in nano- and micromotors. *Adv. Funct. Mater.* **30**, 1908283 (2020).
7. Gómez-Martínez, R. et al. Silicon chips detect intracellular pressure changes in living cells. *Nat. Nanotechnol.* **8**, 517–521 (2013).
8. Pervan, A. & Murphey, T. Bayesian particles on cyclic graphs. In *IEEE/RSJ International Conference on Intelligent Robots and Systems* (IEEE, 2020).
9. Koman, V. B. et al. Colloidal nanoelectronic state machines based on 2D materials for aerosolizable electronics. *Nat. Nanotechnol.* **13**, 819–827 (2018).
10. Liu, P. et al. Autoperforation of 2D materials for generating two-terminal memristive Janus particles. *Nat. Mater.* **17**, 1005–1012 (2018).
11. Thorkelsson, K., Bai, P. & Xu, T. Self-assembly and applications of anisotropic nanomaterials: a review. *Nano Today* **10**, 48–66 (2015).

12. Cortese, A. J. et al. Microscopic sensors using optical wireless integrated circuits. *Proc. Natl Acad. Sci. USA* **117**, 9173–9179 (2020).
13. Velegol, D. *Colloidal Systems* (CreateSpace, 2016).
14. Fan, F. & Stebe, K. J. Size-selective deposition and sorting of lyophilic colloidal particles on surfaces of patterned wettability. *Langmuir* **21**, 1149–1152 (2005).
15. Wu, X. et al. A 0.04 mm³ 16 nW wireless and batteryless sensor system with integrated Cortex-M0+ processor and optical communication for cellular temperature measurement. In *2018 IEEE Symposium on VLSI Circuits* 191–192 (IEEE, 2018).
16. Funke, D. A. et al. A 200 µm by 100 µm smart dust system with an average current consumption of 1.3 nA. In *2016 IEEE International Conference on Electronics, Circuits and Systems* 512–515 (IEEE, 2016).
17. Usami, M. An ultra-small RFID chip: µ-chip. In *Proc. 2004 IEEE Asia-Pacific Conference on Advanced System Integrated Circuits* 2–5 (IEEE, 2004).
18. Fiori, G. et al. Electronics based on two-dimensional materials. *Nat. Nanotechnol.* **9**, 768–779 (2014).
19. Shulaker, M. M. et al. Three-dimensional integration of nano-technologies for computing and data storage on a single chip. *Nature* **547**, 74–78 (2017).
20. Lee, S. et al. A 250 µm × 57 µm microscale opto-electronically transduced electrodes (MOTEs) for neural recording. *IEEE Trans. Biomed. Circuits Syst.* **12**, 1256–1266 (2018).
21. Miskin, M. Z. et al. Electronically integrated, mass-manufactured, microscopic robots. *Nature* **584**, 557–561 (2020).
22. Hempel, M. et al. SynCells: a 60 × 60 µm² electronic platform with remote actuation for sensing applications in constrained environments. *ACS Nano* **15**, 8803–8812 (2021).
23. Weinstein, R. RFID: a technical overview and its application to the enterprise. *IT Prof.* **7**, 27–33 (2005).
24. Withey, P. A. et al. Strain paint: noncontact strain measurement using single-walled carbon nanotube composite coatings. *Nano Lett.* **12**, 3497–3500 (2012).
25. Park, J. et al. Trend and challenges of the diffusion-based molecular communication for nanonetworks. In *2015 International Conference on Information and Communication Technology Convergence* 1362–1364 (2015).
26. Farra, R. et al. First-in-human testing of a wirelessly controlled drug delivery microchip. *Sci. Transl. Med.* **4**, 122ra21 (2012).
27. Yang, J. F. et al. Memristor circuits for colloidal robotics: temporal access to memory, sensing, and actuation. *Adv. Intell. Syst.* **4**, 2100205 (2022).
28. Ceylan, H. et al. Mobile microrobots for bioengineering applications. *Lab Chip* **17**, 1705–1724 (2017).
29. Palagi, S. et al. Structured light enables biomimetic swimming and versatile locomotion of photoresponsive soft microrobots. *Nat. Mater.* **15**, 647–653 (2016).
30. Miskin, M. Z. et al. Graphene-based bimorphs for micron-sized, autonomous origami machines. *Proc. Natl Acad. Sci. USA* **115**, 466–470 (2018).
31. Purcell, E. M. Life at low Reynolds number. *Am. J. Phys.* **45**, 3–11 (1977).
32. Blackiston, D. et al. A cellular platform for the development of synthetic living machines. *Sci. Robot.* **6**, eabf1571 (2021).
33. Wan, K. Y. & Jekely, G. Origins of eukaryotic excitability. *Phil. Trans. R. Soc. Lond. B* **376**, 20190758 (2021).
34. Aguilar, J. et al. A review on locomotion robophysics: the study of movement at the intersection of robotics, soft matter and dynamical systems. *Rep. Prog. Phys.* **79**, 110001 (2016).
35. Aydin, Y. O. et al. in *Robotic Systems and Autonomous Platforms: Advances in Materials and Manufacturing* (eds Walsh, S. M. & Strano, M. S.) 109–127 (Woodhead, 2019).
36. Maladen, R. D. et al. Undulatory swimming in sand: subsurface locomotion of the sandfish lizard. *Science* **325**, 314–318 (2009).
37. Reynolds, M. F. et al. Microscopic robots with onboard digital control. *Sci. Robot.* **7**, eabq2296 (2022).
38. Moran, J. L. & Posner, J. D. Phoretic self-propulsion. *Annu. Rev. Fluid Mech.* **49**, 511–540 (2017).
39. Wang, Y. et al. Bipolar electrochemical mechanism for the propulsion of catalytic nanomotors in hydrogen peroxide solutions. *Langmuir* **22**, 10451–10456 (2006).
40. Wang, J. et al. A silicon nanowire as a spectrally tunable light-driven nanomotor. *Adv. Mater.* **29**, 1701451 (2017).
41. Liu, A. T. et al. Autoperforation of two-dimensional materials to generate colloidal state machines capable of locomotion. *Faraday Discuss.* **227**, 213–232 (2021).
42. Dou, Y. & Bishop, K. J. M. Autonomous navigation of shape-shifting microswimmers. *Phys. Rev. Res.* **1**, 032030 (2019).
43. Pikul, J. H. et al. High-power lithium ion microbatteries from interdigitated three-dimensional bicontinuous nanoporous electrodes. *Nat. Commun.* **4**, 1732 (2013).
44. Choi, J.-H. et al. High capacitance and energy density supercapacitor based on biomass-derived activated carbons with reduced graphene oxide binder. *Carbon* **132**, 16–24 (2018).
45. González, A. et al. Review on supercapacitors: technologies and materials. *Renew. Sustain. Energy Rev.* **58**, 1189–1206 (2016).
46. Liu, A. T. et al. Direct electricity generation mediated by molecular interactions with low dimensional carbon materials—a mechanistic perspective. *Adv. Energy Mater.* **8**, 1802212 (2018).
47. Liu, A. T. et al. Solvent-induced electrochemistry at an electrically asymmetric carbon Janus particle. *Nat. Commun.* **12**, 3415 (2021).
48. Cao, X. et al. Triboelectric nanogenerators driven self-powered electrochemical processes for energy and environmental science. *Adv. Energy Mater.* **6**, 1600665 (2016).
49. Hasan, R. M. M. & Luo, X. Promising lithography techniques for next-generation logic devices. *Nanomanufact. Metrol.* **1**, 67–81 (2018).
50. Gao, M., Li, L. & Song, Y. Inkjet printing wearable electronic devices. *J. Mater. Chem. C* **5**, 2971–2993 (2017).
51. Visser, C. W. et al. In-air microfluidics enables rapid fabrication of emulsions, suspensions, and 3D modular (bio)materials. *Sci. Adv.* **4**, eaao1175 (2018).
52. Chung, S. E. et al. Guided and fluidic self-assembly of micro-structures using railed microfluidic channels. *Nat. Mater.* **7**, 581–587 (2008).
53. Meitl, M. A. et al. Transfer printing by kinetic control of adhesion to an elastomeric stamp. *Nat. Mater.* **5**, 33–38 (2005).
54. Kang, K. et al. Layer-by-layer assembly of two-dimensional materials into wafer-scale heterostructures. *Nature* **550**, 229–233 (2017).
55. Zhang, P. et al. Fracture toughness of graphene. *Nat. Commun.* **5**, 3782 (2014).
56. Rogers, J. et al. Origami MEMS and NEMS. *MRS Bull.* **41**, 123–129 (2016).
57. Felton, S. et al. A method for building self-folding machines. *Science* **345**, 644–646 (2014).
58. Manoharan, V. N., Elsesser, M. T. & Pine, D. J. Dense packing and symmetry in small clusters of microspheres. *Science* **301**, 483–487 (2003).
59. Wang, Y. et al. Colloids with valence and specific directional bonding. *Nature* **491**, 51–55 (2012).
60. Sacanna, S. et al. Lock and key colloids. *Nature* **464**, 575–578 (2010).
61. Lee, H.-Y. et al. Self-assembly of nanoparticle amphiphiles with adaptive surface chemistry. *ACS Nano* **8**, 9979–9987 (2014).
62. Gong, Z. et al. Patchy particles made by colloidal fusion. *Nature* **550**, 234–238 (2017).

63. Grzybowski, B. A. et al. From dynamic self-assembly to networked chemical systems. *Chem. Soc. Rev.* **46**, 5647–5678 (2017).
64. Xu, Z. et al. Transmembrane transport in inorganic colloidal cell-mimics. *Nature* **597**, 220–224 (2021).
65. Kriegman, S. et al. A scalable pipeline for designing reconfigurable organisms. *Proc. Natl Acad. Sci. USA* **117**, 1853–1859 (2020).
66. Gracias, D. H. Forming electrical networks in three dimensions by self-assembly. *Science* **289**, 1170–1172 (2000).
67. Toffoli, T. & Margolus, N. Programmable matter: concepts and realization. *Phys. D* **47**, 263–272 (1991).
68. Angluin, D. et al. Computation in networks of passively mobile finite-state sensors. *Distrib. Comput.* **18**, 235–253 (2006).
69. Patitz, M. J. An introduction to tile-based self-assembly and a survey of recent results. *Nat. Comput.* **13**, 195–224 (2014).
70. Reid, C. R. & Latty, T. Collective behaviour and swarm intelligence in slime moulds. *FEMS Microbiol. Rev.* **40**, 798–806 (2016).
71. Yim, M. et al. Modular self-reconfigurable robot systems [grand challenges of robotics]. *IEEE Robot. Autom. Mag.* **14**, 43–52 (2007).
72. Pervan, A. & Murphey, T. D. Low complexity control policy synthesis for embodied computation in synthetic cells. In *WAFR 2018: International Workshop on the Algorithmic Foundations of Robotics* (eds Morales, M. et al.) 602–618 (Springer, 2020).
73. Andrés Arroyo, M. et al. A stochastic approach to shortcut bridging in programmable matter. *Nat. Comput.* **17**, 723–741 (2018).
74. Cannon, S. et al. A local stochastic algorithm for separation in heterogeneous self-organizing particle systems. In *Proc. 2019 APPROX-RANDOM 1–22* (Schloss Dagstuhl - Leibniz-Zentrum für Informatik, 2019).
75. Kriegman, S. et al. Kinematic self-replication in reconfigurable organisms. *Proc. Natl Acad. Sci. USA* **118**, e2112672118 (2021).
76. Savoie, W. et al. A robot made of robots: emergent transport and control of a smarticle ensemble. *Sci. Robot.* **4**, eaax4316 (2019).
77. Kuang, M. et al. Inkjet printing patterned photonic crystal domes for wide viewing-angle displays by controlling the sliding three phase contact line. *Adv. Opt. Mater.* **2**, 34–38 (2014).
78. Chang, J.-K. et al. Materials and processing approaches for foundry-compatible transient electronics. *Proc. Natl Acad. Sci. USA* **114**, E5522–E5529 (2017).

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