Perspective

Magnetically aware actuating composites: Sensing features as inspiration for the next step in advanced magnetic soft robotics

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In this perspective, we explore the convergence of sensing and actuation in magnetic composites and its potential applications in various fields. There is a need for multifunctional mechanically flexible materials that can be easily processed into functional devices that respond to a wide range of physical stimuli, including magnetic fields. These characteristics aim for lightweight and mechanically imperceptible systems that help us to interact with technology and with each other without the need for a bulky gadget. Typically, magnetically responsive devices are constructed using materials that do not necessarily possess flexible properties, so magnetosensitive composites with tailored magnetic, conductive, and flexible properties arising from the combination of their constituents have been proposed. Such property tunability is, in turn, beneficial for achieving functional convergence. This perspective aims to address several of the challenges associated with magnetoresponsive soft composites while highlighting the synergistic convergence that will take further the applications of magnetically aware actuating composites.

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I. INTRODUCTION

Over the past century, we have witnessed the replacement of rigid technologies by soft and flexible alternatives. This transformation is evident in various domains, from the widespread adoption of safer soft contact lenses to the innovative applications of flexible electronics [1,2]. Mechanically soft materials have played a pivotal role in creating adaptive, comfortable, and responsive substitutes [1]. As in other fields, robots fabricated using soft materials have been explored, driven by the strong potential to enhance human and environmental safety, dexterity, and interactivity [3–6]. During the past decade, solid exploratory works have paved the way for possible actuation mechanisms in flexible formats, including pneumatic, hydraulic, and electroactive polymers, shape-memory polymers, and magnetic technologies [3–8]. These mechanisms are moving the field of soft robotics forward, promising significant contributions to health care, exploration, and human-robot collaboration, while also contributing to sustainability efforts [9,10]. However, realizing the full potential of these capabilities requires exploring beyond and/or revolutionizing the current actuation mechanisms, which have been the primary focus of the existing efforts. Soft robotics research has to focus on the advantages of integrating mechanically compatible sensing capabilities, thereby closing the loop between actuation and awareness of both external and internal parameters [9]. This integration enhances the functionality of soft robots, enabling precise actuation. This approach involves integrating various functionalities, such as actuation and sensing, within the same multifunctional soft material [11] to address mechanical-compatibility challenges. Focusing on soft magnetic composites, we will elaborate on their advantageous properties and how their integration into soft robotics will cover some of the most relevant challenges during the lifetime of the device.

Polymeric magnetic composites, composed of magnetic particles typically embedded in epoxy or elastomer matrices, have been known for a long time for their use as permanent magnets [12]. Endowing flexibility to these magnetic composites by using elastomeric matrices has allowed the scientific community to discover their relevance in soft robotics, providing a wireless-powered, programmable, and fast-actuating toolkit of soft materials [7,12–15].

Magnetic composites that incorporate Nd-Fe-B (0.5×10^2 emu g^{-1}) or Fe_3O_4 (0.5×10^1 emu g^{-1}) fillers demonstrate obvious enhancement in magnetic responsibility compared to purely organic plastic magnets (10^{-4} emu g^{-1}) [16,17]. From the viewpoint of mechanical stability, incorporating polymers endows the composites with flexibility and stretchability and thus susceptibility to mechanical deformation. It is remarkable that the intrinsic Young's modulus of polymeric composites changes over less than one order of magnitude with the addition of up to a 50-wt% concentration of magnetic fillers [17]. The incorporation of soft polymers not only imparts mechanical stability, providing flexibility and stretchability

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susceptible to mechanical deformation, but also maintains magnetoresponsive properties comparable to their rigid counterparts [12]. This is crucial for deformable magnetic devices.

The unique properties of these magnetic composites, characterized by magnetic responsibility and robust deformability, enable the development of unique soft-bodied robots with lightweight, durable, untethered, programmable, and ultrafast properties [18–20]), emerging as the closest synthetic system analogous to living organisms, mimicking their mechanical behavior, and going beyond them in terms of performance [21]. In contrast to the rest of the soft composites actuated by alternative mechanisms, such as pneumatic, hydraulic, or electroactive methods, magnetic soft composites exhibit rapid responsiveness to magnetic stimuli and remain impervious to electrical and optical disturbances in their surroundings. Additionally, they can be anisotropically programmed during fabrication by judiciously aligning the particles inside the composite using magnetic fields. Anisotropic composites selectively respond to different directions and polarities of the magnetic fields [6,7]. These distinctive features ensure a reliable and instantaneous actuation process. Despite these advantages, challenges exist, such as the potential to malfunction when the external conditions are changed. For example, the change of the intrinsic magnetic properties due to temperature changes can lead to modifications in the actuation pattern [22]. Addressing this challenge involves imparting self-sensing capability to soft robots by enabling communication with external controlling devices for self-guided tracking of the completion of motion [23]. To this end, magnetic composites can already be used to realize magnetic field sensors [24–29] to guide the deformation and/or movement of soft robots. To comply with the operational modes of soft robots, magnetic field sensors should be stretchable, bendable, shape-conformal, and capable of withstanding extreme mechanical deformations [30,31].

While magnetic composites can already be employed to create magnetic field sensors guiding the deformation and movement of soft robots, ensuring their functionality in the face of continuous severe deformation is crucial [2]. The continuous severe deformation will inevitably result in functional failure and a shortened life span of soft robots and magnetic field sensors. Frequent replacement of such devices will require a great deal of energy, material, time, and capital investment, contributing to environmental burdens [32]. Improper disposal of discarded devices containing toxic magnetic elements, such as Co and Ni, poses risks to the environment and to living organisms [33–35]. Therefore, it is imperative to address these problems to achieve sustainable and green advanced (magneto)electronics.

The extensive research and progress in magnetic composites have provided a versatile platform for the convergence of soft robotics and sensorics, paving the way for the development of highly embedded intelligent systems. These advancements have been curated in recent literature reviews describing the up-to-date materials, fabrication techniques, and applications of magnetic composites [21,36–41].

II. PERSPECTIVE: MAGNETICALLY AWARE ACTUATING COMPOSITES—THE CONVERGENCE OF SENSING AND ACTUATION IN MAGNETIC COMPOSITES

In recent research on magnetic composites, the focus has primarily revolved around two distinct domains: the magnetotransport phenomena of percolated composites and the actuation mechanisms of flexible magnetic materials [46] (Fig. 1). Conductive magnetosensitive composites have shown promise in the development of printed sensors [24,27,42,48] [Figs. 1(a)–1(c)], while flexible magnetic composites have gained recognition as soft actuators [18,19] [Figs. 1(d)–1(e)]. Although these fields have historically been treated as isolated domains, the methods applied in creating magnetically sensitive conductive composites can be extended to the development of soft actuators. We aim to underscore the synergies that can emerge from this convergence to create magnetically aware soft robotics that are imperceptible, self-repairable, and that report their actuation state [23,25,43] [Figs. 1(f)–1(h)]. These novel capabilities can be obtained with customized approaches such as additive manufacturing [44] [Fig. 1(i)] of environmentally friendly materials [45] [Fig. 1(j)] covering the feasibility, integration, and responsible fabrication of the proposed devices.

Similar to classical robotic systems, magnetic soft robotics need sensing and control units that are compatible with their highly deformable mechanics. Flexible and stretchable conductive magnetic composites [24,25,27] can be created using conductive magnetic fillers that can integrate sensing capabilities and at the same time provide magnetic actuation capabilities. For example, polymer composites containing ferromagnetic particles of permalloy (i.e., $Ni_{81}Fe_{19}$ alloy) have been demonstrated as printed magnetic field sensors [25,27]. At the same time, the large permeability of these types of particles can used not only for sensing but also for actuation purposes. Until now, similar examples showing magnetic origami actuators have been proposed by laminating flexible sensors for increasing the reliability of the actuation of magnetic actuators [23] but, nevertheless, there is no demonstration of fully embedded solution-processable magnetic actuation and magnetic sensing capabilities.

Currently, composites with large remanent magnetization (0.5×10^2 emu g^{-1}) that show high magnetoresistance sensitivity are lacking [16,17,24,25]. This case would be the ideal combination of the properties of the easy and programmable actuation of hard magnetic fillers and the high electrical transport sensitivity of soft

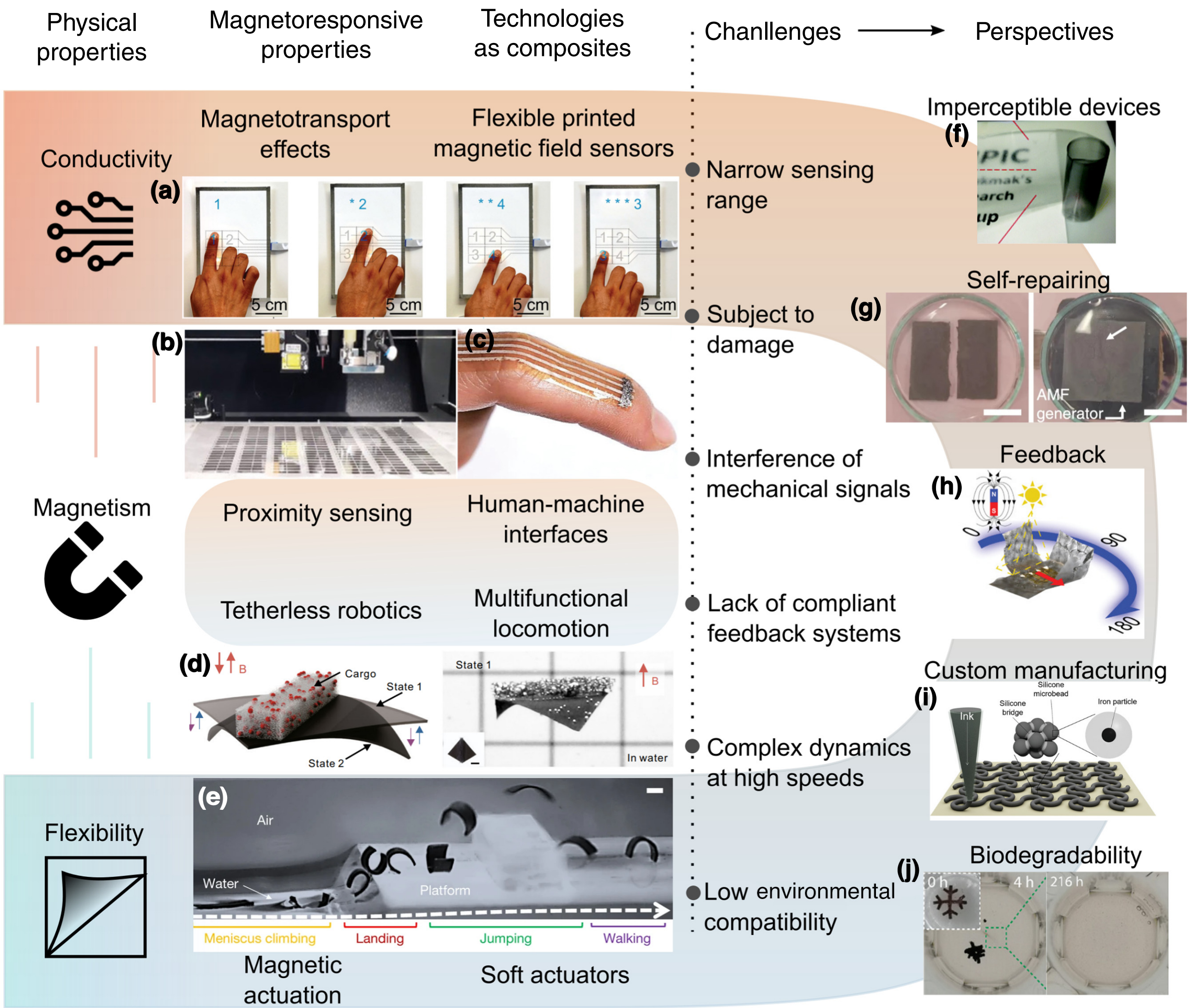

FIG. 1. Magnetic soft composites, combining both actuation and sensing functionalities, are expected to promote the performance of current magnetic soft robots by enabling feedback control through their awareness of the environment variation and proprioception. Research in magnetic composites has primarily concentrated on two distinct areas: the magnetotransport phenomena of magnetic and conductive properties (top) and the actuation of magnetic soft composites (bottom). Conductive magnetoresponsive composites have been shown in the development of magnetoresistive printed sensors for (a) interactive proximity sensing. (b) This scalable printing approach exhibit the potential for mass manufacturing of magnetically sensitive composites. (c) Endowing magnetoresistive sensors with mechanical flexibility could allow for comfortable wear on the biodynamic human body to serve as a human-machine interface. (d), (e) Soft robots made of magnetic soft composites demonstrate (d) rapid responsiveness and (e) precise manipulation capabilities. Because of the extensive compatibility in fabrication, external stimuli, and mechanical flexibility, these two research areas, traditionally separate from each other, could merge in a convergent field of magnetically aware soft actuators. Such convergence will contribute to solving the issues existing in the present research field, such as the narrow sensing range, the susceptibility to damage, the interference of mechanical signals, the lack of mechanically conformal feedback systems, the complex dynamics at high speeds, the low environmental compatibility, etc. (f)–(j) In turn, numerous applications and functionalities that were previously unavailable can potentially be unlocked, including (f) imperceptibility, (g) self-healing, (h) self-awareness, (i) structural customization, and (j) biodegradability. (a)–(d) Reproduced from Refs. [26,42,24,18] under the terms of the CC-BY Creative Commons Attribution 4.0 International license [47]. (e) Reproduced with permission. Copyright 2018, Springer Nature [19]. (f) Reproduced with permission. Copyright 2018, RSC Publishing [43]. (g),(h) Reproduced from Refs. [23,25] under the terms of the CC-BY Creative Commons Attribution 4.0 International license [47]. (i) Reproduced with permission. Copyright 2019, John Wiley and Sons [44]. (j) Reproduced from Ref. [45] under the terms of the CC-BY Creative Commons Attribution 4.0 International license [47].

magnetic composites. A possible approach is to use the piezoresistive effects of magnetically actuated conductive composites in motion [49,50]. In this case, the applied field is not measured directly but, rather, the deformations caused by the stimuli are measured. This approach will endow proprioception capabilities but does not allow the possibility of directly tracking the environmental fields that induce their actuation [46]. An alternative approach to solve this challenge is the use of multimaterial composites integrated into actuators, where the main body consists of a permanently magnetized composite with embedded magnetoresistive composite tracks. This approach would require the tuning of the mechanical properties of both composites, which, if achieved successfully, would endow the soft actuator with direct-sensing capabilities of the fields that are being used to trigger their motion. Prospectively, the most comprehensive solution consists of a fully magnetically aware actuating composite, which would integrate into a single-phase material the sensing and actuation properties of the constituent materials. These materials would eliminate the need for additional fitting of the mechanical compatibility of multimaterial approaches. Unified mechanical properties would directly translate into simplified design and an extended device lifetime and reduce the environmental burden and economic costs related to constant device replacements.

Examples of a soft machine integrated with flexible electronics to produce proprioception feedback and awareness of the environment exist in the literature [23]. Although magnetic actuators can provide a useful platform for programming the actuation inside the material, a feedback system can confirm the success of the command. This becomes especially relevant when soft actuators are actuated at high speeds. High-speed actuation can create nonlinear behavior on soft robots due to their low mechanical impedance [51]. Feedback of actuation will allow the detection of obstacles, nonlinearities, perturbations, and completeness of the commanded actuation. These integrated sensing devices might offer vast potential in rehabilitation, fine motricity tasks, and human-robot collaboration, ensuring safe and versatile interaction with users [4,52,53].

The merging of actuation and sensing requires alignment with the ongoing challenges faced by modern electronic devices, encompassing their entire life cycle from production and customization to integration, repair, and safe interaction with both humans and the environment [2]. Neglecting these considerations in the research and development phases can hinder the broader adoption of these technologies [1]. In the subsequent sections, we will describe some of the most relevant aspects that demand attention to establish magnetically aware actuating composites as a feasible and promising alternative within future soft robotics.

A. Shape, sense, actuate: From high-throughput two-dimensional printing to advancing magnetosensitive three-dimensional printing

Printing technologies have evolved beyond books and now encompass batteries, circuits, displays, or magnets [41,54–58]. This paradigm shift has enabled the printing of tools, prostheses, organs, and food [59–62]. In the frame of magnetic composites, the concept of printing holds immense potential. Utilizing industry-standard printing methods such as screen printing, high-performance magnetic sensors, and actuators can be fabricated, paving the way for integration in Internet of Things (IoT) systems, smart homes, customizable wearables, and haptic systems. Printed magnetic field sensors are already covering a wide range of the magnetic field spectra, from nanoteslas to a few teslas [25,26] and such systems can interact with high-power electromagnets, fridge magnets, and the magnetic field produced by the operation of our commonly used electronic devices.

Adding a new dimension to the printing of magnetic sensors and actuators opens up novel yet-to-be-explored possibilities for fabricating fully functional machines and systems. Three-dimensional (3D) printed formats already profit from the functionality embedded in the shape of the printed part [63]. Embedding electronic capabilities during the additive-manufacturing process allows for building intelligent parts that can measure and report their state during operation [64]. Removing the need for assembly operations might provide access to print-and-play intelligent systems. The aim of this type of structure would be to embed the magnetostatic interactions, the appropriate geometry, and the sensing capabilities that could lead to smarter 3D-printed objects. These intelligent printed parts can be designed as functional devices, programmed to perform automated tasks such as prototypes for laser-lithography sweepers or micromanipulators for precise alignment in optical setups. These examples illustrate the possibility of combining magnetostatic interactions, appropriate geometry, and sensing capabilities for smart 3D-printed magnetosensitive objects.

B. Invisible sensing, visible impact: Fully imperceptible magnetic actuators

Similarly to the way in which flexibility formats allow comfortable mechanically imperceptible magnetoelectronics [30,65,66], visually imperceptible sensors allow for integration without affecting the appearance of the target object [67]. As systems move toward multifunctional materials with visual feedback, transparency becomes even more critical. Users who directly interact with such multifunctional magnetic composites might find it easier to have a visual cue of the state of the system, without the interference of the typical black appearance of magnetic composites. The use of nanosized fillers with a high aspect ratio is a familiar strategy to achieve optically transparent conductive sensing composites [68] but has not yet been explored for magnetically aware magnetic composites.

From the sensing point of view, the interaction through magnetic fields in 3D visuals might find appealing applications for education, design, gaming, and promotional systems. Touch-screen displays rely on transparent conductors for intuitive interaction with objects on a flat screen [69]. In the same way, transparent magnetic interfaces could boost the development of touchless 3D visuals. Holographic [70] and 3D displays [71] can create more realistic digital experiences and the low interference of magnetic fields with such visuals can enable higher integration capabilities. The unconventional feature of visual transparency in magnetic field sensors opens up possibilities for creating more realistic digital experiences, which are possible through their visual imperceptibility.

Transparency holds promise in actuation applications such as optomechanical systems moving at high speeds with simple fabrication and actuation methods [72]. However, achieving transparency in magnetic composites for actuation poses challenges due to reduced filler concentration and the subsequent decrease in the effective power exerted by the composite. For this reason, the exploration of softer polymers such as gels [21], which can be actuated with smaller forces and torques, becomes crucial. While classical gels may be susceptible to environmental conditions, the emerging field of ionic gels [73] offers highly deformable yet robust materials that could potentially fulfill the vision of transparent and actuating magnetically aware composites.

C. From vulnerability to resilience: Self-healing solutions for flexible magnetically aware actuators

Magnetically aware actuating composites might face increased vulnerability to damage compared to rigid technologies [2]. Continuous deformation during actuation and exposure to harsh environmental conditions pose significant challenges to their long-term reliability. To address this issue, we highlight the relevance of the integration of self-healing capabilities into magnetically aware actuating composites. There is a current need to provide technological solutions that increase the reliability of these flexible devices in the long term even after unforeseen damage. It has been reported that self-healing polymers can enable easy repair of printed sensing composites that have cracks at the micrometer scale or that have even been torn apart [25,74,75]. In particular, the smart combination of the self-healing capabilities of the polyborosiloxane (PBS)–polydimethylsiloxane (PDMS) matrix [76] combined with the magnetic interaction of polymers creates the possibility of having a magnetically induced repair mechanism. The printed composites were conductive and had a magnetoresistive response of 0.9% owing to the anisotropic magnetoresistive (AMR) effect of the permalloy fillers. After inducing a micrometer-scale crack, the sensor fails and a high resistance level was observed. The use of alternating magnetic fields (AMF) (130 mT, 50 Hz) induced the temporary alignment, attraction, and further oscillation of the particles that reestablished the percolation of the conductive network of magnetoresistive fillers in the polymer. The repaired sensor shows the expected magnetoresistive response in the range of 1% with a high sensitivity of 35.7 $\Omega\ T^{-1}$, low noise 19 $\Omega\ Hz^{-1/2}$ at 1.8 Hz, and a high resolution of 36 nT [25].

This is an encouraging demonstration of nonmanual remote repair with AMF of magnetosensitive composites with 100% performance recovery at ambient conditions without heat treatments. Beyond the fact that printed magnetic field sensors were taken to nanotesla detection levels for the first time, the increase in reliability places this technology as an attractive approach to take flexible magnetic composites closer to day-to-day viability in use. The AMF-mediated healing can be used by command after several repetitions of device failure and can recover the sensing capabilities of the as-printed composite. Additionally, this technology can sense and actuate healing even in environmentally tough conditions such as underwater exposure, which is relevant for devices on textiles and flexible substrates [25]. Self-healing, as an unconventional feature for magnetic field sensors, might provide the possibility of creating more sensitive and more reliable magnetic composite systems. In combination with previously developed self-healable magnetic actuators, this can provide the opportunity to build highly adaptable magnetically guided systems that are aware of the environment and can adapt their shape and size to work individually and join collaboratively.

D. Going green with magnets: Biodegradable composites for ecofriendly smart composites

The successful implementation of these technologies ultimately depends on the health and environmental compatibility of the materials employed to construct magnetically aware actuating composites. The higher consumption rates, shorter life span, and few options for repair of electric and electronic equipment lead to large volumes of electronic waste (E-waste). The United Nations Global E-waste Monitor 2020 has revealed that in 2019, electronic waste hit a record high of 53.6×10^6 Mt, a 21% increase in just 5 years [77]. Toxic heavy metals and hazardous chemicals would be released into the environment if E-waste were improperly disposed of, permeating biological systems through the soil, receiving waters, and food chains [77].

The development of biocompatible and biodegradable magnetically aware actuating composites offers a set of promising routes to address this issue [78]. Biodegradable and green composites can be degraded in the natural environment with minimized hazards to health and ecosystems [79–81]. These composites enable easy recovery of the magnetic fillers through the degradation of the binder, along with the potential for efficient recovery of the fillers using magnetic fields [82]. This would enhance the recovery throughput of functional devices and minimize damage to the ambient. This approach is a shift toward a more circular and ecosustainable approach to electronic waste management that promotes the recovery and recycling of valuable and critical materials. This transition will contribute to a cleaner and healthier paradigm of making, using, and repurposing magnetic composites. This vision will require the participation of scientific, industrial, and government actors that establish the mechanisms for effective transit and recovery of these technologies.

III. CONCLUSIONS

In this perspective, we have highlighted the immense potential for magnetic soft actuators by leveraging synergies with the developments in printed magnetic field sensors. The convergence of the two fields offers promising opportunities in device fabrication, the judicious selection of functional materials, and the exploration of novel applications. The complementary synergy between magnetic field sensors and magnetic soft actuators opens up exciting prospects for enhancing the capabilities and applications of soft robotics, paving the way for robotic systems that can safely interact with humans. Nevertheless, realizing the practical utilization of magnetic soft robots to attain the aforementioned objectives remains a critical challenge. It necessitates a holistic integration of critical components such as power sources, control units, communication systems, sensing devices, feedback channels, and user interfaces [4]. For magnetic soft robots to effectively carry out tasks, these system elements must operate in harmony. For example, power sources (e.g., embedded rechargeable batteries) must function in coordination with the control unit to maintain a consistent power supply [83,84]. This involves monitoring battery levels, optimizing power consumption, and initiating wireless energy transfer to prolong the operational duration [85,86]. Considering the intricacy of this endeavor, it is imperative to gain collaboration among experts from diverse fields of materials science, magnetism, electronics, communication, and system design. Looking ahead, research endeavors should continue to push multifunctional composites and refine the integration of components to propel the field toward practical applications. With concerted efforts, the vision of magnetic soft robots seamlessly interacting with humans and their environments can transition from promise to reality.

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