



Sustainable and untethered soft robots created using printable and recyclable ferromagnetic fibers

Wei Tang^{1,2,3} · Yidan Gao^{1,2} · Zeyu Dong^{1,2} · Dong Han^{1,2} · Vadim V. Gorodov⁵ · Elena Y. Kramarenko^{4,5} · Jun Zou^{1,2} 

Received: 14 November 2023 / Accepted: 16 June 2024 / Published online: 16 August 2024
© Zhejiang University Press 2024

Abstract

Integrated printing of magnetic soft robots with complex structures using recyclable materials to achieve sustainability of the soft robots remains a persistent challenge. Here, we propose a kind of ferromagnetic fibers that can be used to print soft robots with complex structures. These ferromagnetic fibers are recyclable and can make soft robots sustainable. The ferromagnetic fibers based on thermoplastic polyurethane (TPU)/NdFeB hybrid particles are extruded by an extruder. We use a desktop three-dimensional (3D) printer to demonstrate the feasibility of printing two-dimensional (2D) and complex 3D soft robots. These printed soft robots can be recycled and reprinted into new robots once their tasks are completed. Moreover, these robots show almost no difference in actuation capability compared to prior versions and have new functions. Successful applications include lifting, grasping, and moving objects, and these functions can be operated untethered wirelessly. In addition, the locomotion of the magnetic soft robot in a human stomach model shows the prospect of medical applications. Overall, these fully recyclable ferromagnetic fibers pave the way for printing and reprinting sustainable soft robots while also effectively reducing e-waste and robotics waste materials, which is important for resource conservation and environmental protection.

Wei Tang, Yidan Gao, and Zeyu Dong have contributed equally to this work.

✉ Elena Y. Kramarenko
kram@polly.phys.msu.ru

✉ Jun Zou
junzou@zju.edu.cn

¹ State Key Laboratory of Fluid Power and Mechatronic Systems, Zhejiang University, Hangzhou 310058, China

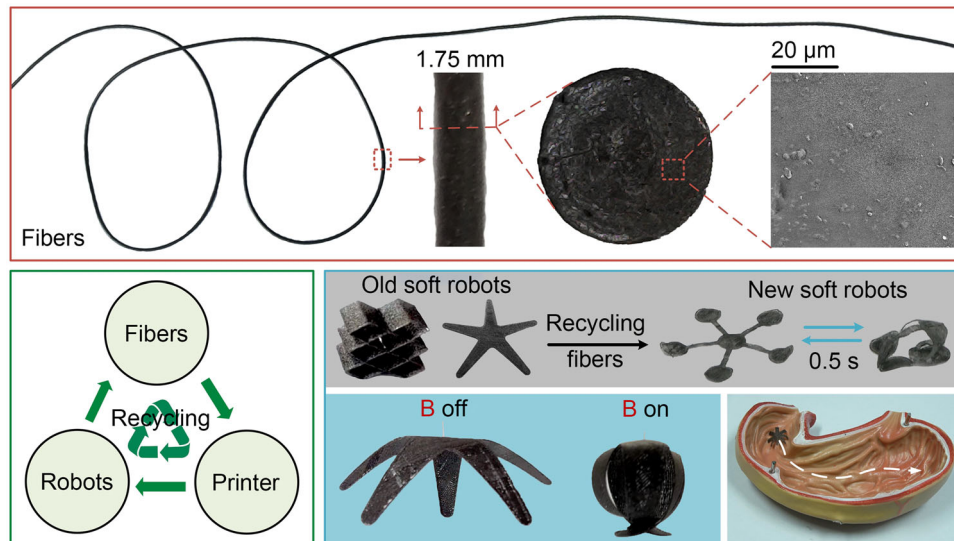
² School of Mechanical Engineering, Zhejiang University, Hangzhou 310058, China

³ Institute of Process Equipment, College of Energy Engineering, Zhejiang University, Hangzhou 310027, China

⁴ Faculty of Physics, Lomonosov Moscow State University, Moscow 119991, Russia

⁵ Enikolopov Institute of Synthetic Polymeric Materials of Russian Academy of Sciences, Moscow 117393, Russia

Graphic abstract



Keywords Ferromagnetic fibers · Sustainable soft robots · Three-dimensional printing · Recyclable soft materials · Medical applications

Introduction

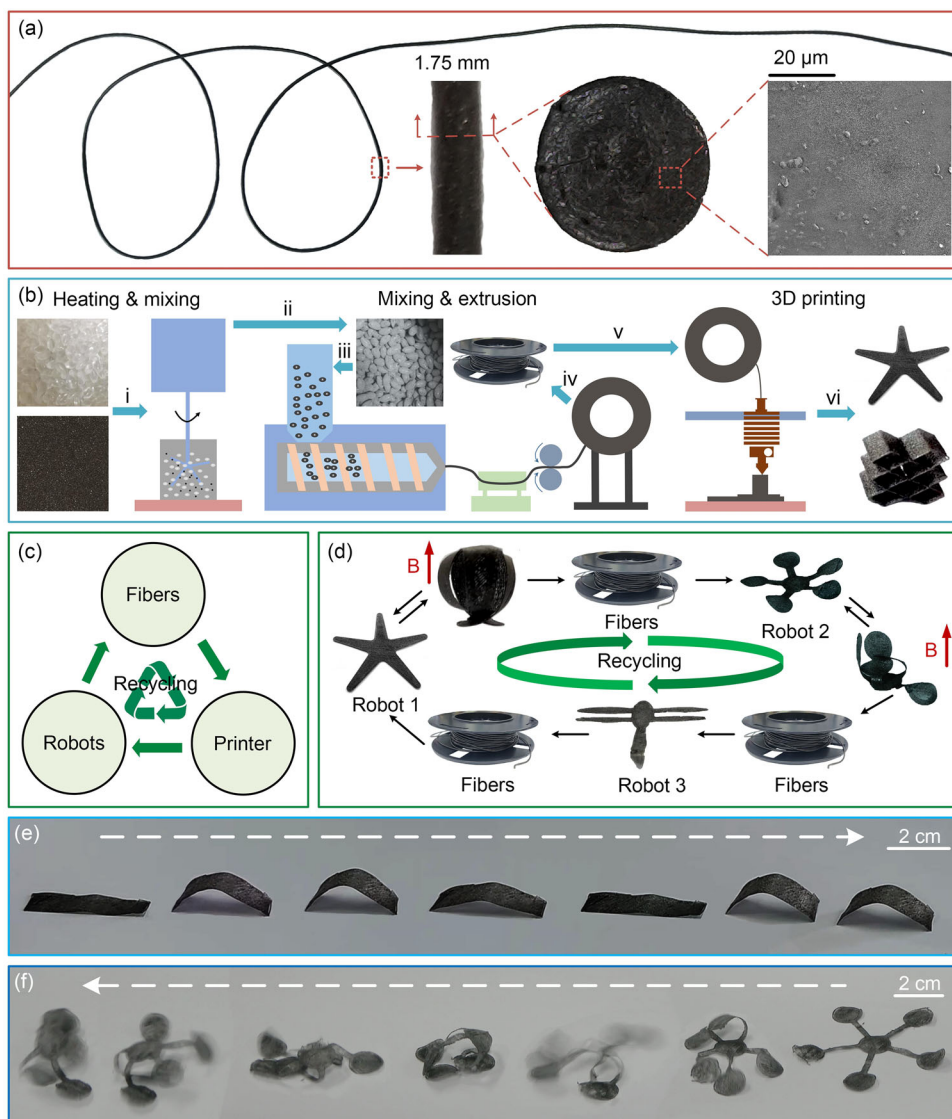
Robots are employed in many fields, including manufacturing, medical devices, agriculture, education, and consumer products, due to their high productivity, safety, and convenience. However, the rising usage of robots also brings material waste and unsustainable disposal techniques [1]. In 2019 alone, electronics generated 53.6 million metric tons of unrecycled waste, a number that is expected to grow by nearly 2 million per year [2]. Consequently, environmental issues related to e-waste and robotic waste materials have attracted increasing attention worldwide [3, 4]. Therefore, it is crucial to use sustainable materials while creating robots [5], and recyclable robotics materials [6, 7] offer a promising alternative.

Single-composite materials typically offer good recycling potential [8]. However, employing a single-composite material to create a traditional rigid robot [9–11] may be impractical. In contrast, the recent emergence of soft robots [12–21]—and particularly magnetic soft robots [22–26]—suggests a new possibility. Typically, magnetic soft robots are made of a single magnetic composite material, often consisting of Ecoflex or polydimethylsiloxane (PDMS) mixed with ferromagnetic particles. These have significant application prospects for medical treatments [27–29]. These robots can be deformed in many ways, and their movements can be actuated by magnetic fields. However, Ecoflex- or PDMS-based soft robots cannot be directly recycled or reprocessed at the end of their life by melting them down into a raw

material capable of being used for a new product. Instead, they require additional processing stages (i.e., depolymerization, polymerization, and re-manufacturing) and may be disposed of as waste, posing a potential environmental hazard [30–32]. In addition, most magnetic soft robots are made using casting methods [33–35], which results in relatively simple and primarily two-dimensional (2D) structures, thereby restricting their use for complex use cases. Some robots can also be printed using the direct ink writing (DIW) method [36–40]; here, the printed structure is primarily two-dimensional, with only a few robots having a simple three-dimensional (3D) structure. However, this kind of printing requires an additional nozzle to print support molds [41]. Thus, after the printing is complete, it is necessary to remove the mold; however, it is challenging to print complex 3D structures due to the ink is extremely easy to collapse. Therefore, integrated printing of magnetic soft robots with complex structures using recyclable materials to achieve sustainability of the soft robots remains a persistent challenge.

Here, we propose a class of ferromagnetic fibers (Fig. 1a) that are printable and recyclable, and can, therefore, be used to fabricate sustainable untethered soft robots. These ferromagnetic fibers, which are based on thermoplastic polyurethane (TPU)/NdFeB hybrid particles, are extruded via a single-screw extruder. They have a diameter of 1.75 mm, making them suitable for commercial desktop fused deposition modeling (FDM) 3D printers. We analyzed the effect of

Fig. 1 Recyclable ferromagnetic fibers for sustainable robots. **a** Ferromagnetic fibers are printable and soft. Scanning electron microscope images show the cross-sections of the ferromagnetic fibers. **b** The processes by which ferromagnetic fibers and soft robots are manufactured. A single-screw extruder was used to extrude the ferromagnetic fiber. **c** The ferromagnetic fiber, printer, and robot form a circular system. **d** Detailed circular process. **e** A soft robot when crawling. **f** A soft robot when rolling



the TPU/NdFeB hybrid particle doping ratio on the diameter and density of formed ferromagnetic fibers, as well as their printability, mechanical performance, and recyclability. We then assessed the impact of ferromagnetic fibers on 3D-printed soft robots and verified their recyclability. In addition, an FDM 3D printer was used to demonstrate the feasibility of printing 2D and complex 3D soft robots with ferromagnetic fibers, and a magnetic field was applied to remotely actuate robot deformations. Finally, once the soft robots had accomplished their tasks, we demonstrated the sustainability of the ferromagnetic fiber-based soft robot paradigm by reprinting new robots designed for new applications. We believe that the use of fully recyclable ferromagnetic fibers provides an effective way to print and reprint sustainable soft robots. Moreover, we note that this method is simple to use and can be easily commercialized.

Results

Recyclable ferromagnetic fibers for sustainable robots

The diameter of the ferromagnetic fiber is standard since it is made to be suitable for commercial desktop 3D printers. The matrix materials for ferromagnetic fibers were chosen from three TPU particles with different harness, whose main parameters are shown in Table S1 (Supplementary Information). The main parameters of the NdFeB particles used as the magnetic filler for ferromagnetic fibers are shown in Table S2 (Supplementary Information). The fabrication process of ferromagnetic fibers is shown in Fig. 1b and Movie S1 (Supplementary Information). First, the TPU particles and NdFeB particles were dried at 60 °C for 8 h. Then, TPU particles and NdFeB particles at a certain mass ratio were

heated to 70 °C and put into a blender for thorough mixing. Next, the TPU/NdFeB particle mixture was put into a single-screw extruder to extrude ferromagnetic fibers. By setting the appropriate temperature, extrusion speed, and pulling speed in the single-screw extruder, ferromagnetic fibers with a diameter of about 1.75 mm were obtained, which can be used in commercial desktop FDM 3D printers. Finally, the ferromagnetic fibers were used to create magnetic soft robots using an FDM 3D printer (Fig. 1b and Movie S2 in Supplementary Information).

Interestingly, soft robots printed with ferromagnetic fibers are fully recyclable, and the ferromagnetic fiber itself is also recyclable. This is made possible by a circular relationship between ferromagnetic fibers, the robot printer, and the robot (Fig. 1c). In this circular system, the printer creates the robot by printing ferromagnetic fibers. Once the robot is no longer needed, it can be recycled back into ferromagnetic fibers to begin a new cycle. An example is shown in Fig. 1d; here, Robot 1 is manufactured using ferromagnetic fibers, and once it completes its task, it is recycled back into ferromagnetic fibers. These fibers are then used to create Robot 2, which—after fulfilling some other task—can be recycled into ferromagnetic fibers once more. Furthermore, the structure and design of magnetic soft robots are highly dependent on their specific task, function, and application. These robots can have diverse shapes and functionalities and can be actuated by an external magnetic field. For example, Figs. 1e and 1f and Movie S3 (Supplementary Information) show the untethered motions of two sustainable soft robots, including crawling and rolling motions.

Characterization of ferromagnetic fibers

A single-screw extruder (Fig. S1 in Supplementary Information) comprising a main machine, water cooler, pulling machine, and reeling machine was used to extrude ferromagnetic fibers. The single-screw extruder operates as shown in Movie S1 (Supplementary Information) and/or as described in Sect. “Methods”. Briefly, a TPU/NdFeB particle mixture was first fed into the extruder from a hopper, where high temperatures caused it to form into a mixed molten state within the barrel of the screw section. This material was then extruded by the main machine of the extrusion die, pulled by the pulling machine, cooled in a water cooler tank, and then molded into uniform, continuous ferromagnetic fibers by the reeling machine. Using several experiments, we determined the ideal extruder parameter set Sect. “Methods” for a 90-A-TPU-based matrix. Overall, we came to the conclusion that the extruder’s temperature control I level should be set to about 205 °C, temperature control II level should be set to about 200 °C, the extrusion speed should be set to about 9.5 r/min, and the pulling speed should be set at about 30 r/min, which can stabilize the extrusion of fibers with a

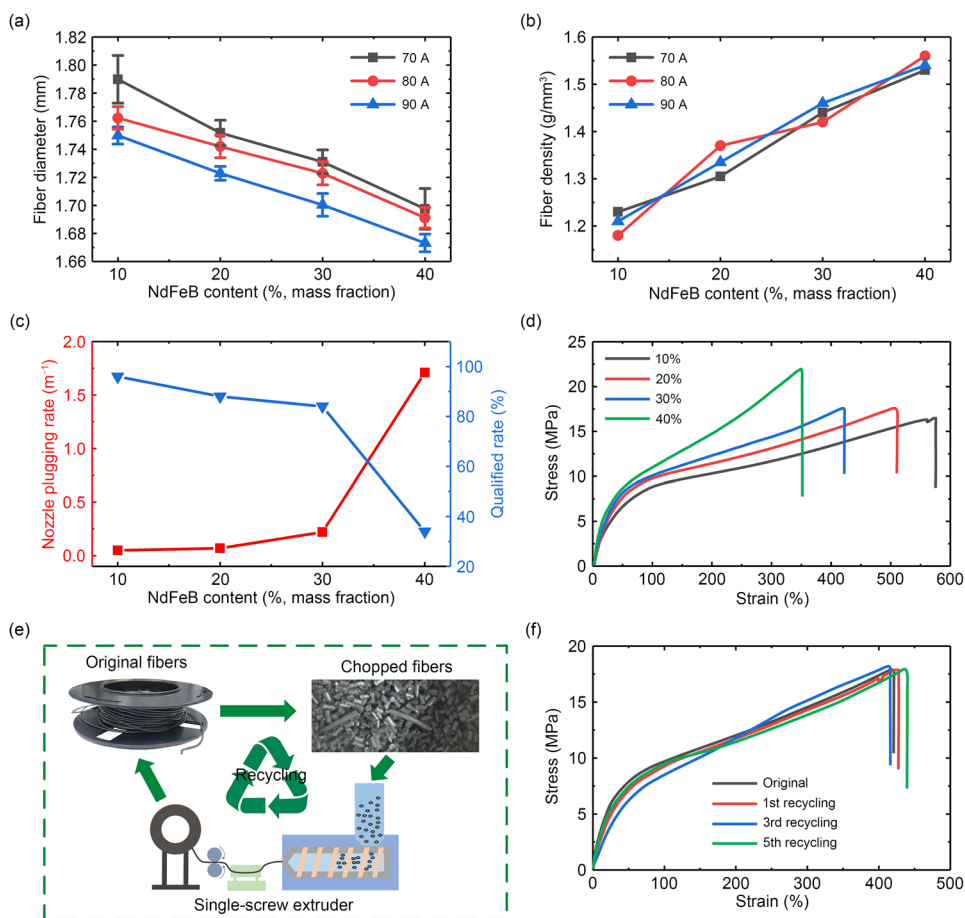
diameter of about 1.75 mm. Scanning electron microscope images (Fig. 1a) show the cross-sections of the ferromagnetic fibers. Figure S2 (Supplementary Information) shows energy-dispersive X-ray spectroscopy characterization of a cross-section.

Since the diameter of the ferromagnetic fiber affects printing, we analyzed its diameter. Accordingly, the diameters of ferromagnetic fibers made from TPU/NdFeB with different TPU hardness and magnetic powder (i.e., NdFeB) content levels were measured using Vernier calipers. Each spool of ferromagnetic fibers was tested 20 times at intervals of 0.5 m. As shown in Fig. 2a, the mean value of these 20 tests was taken to represent the diameter of the ferromagnetic fibers analyzed (Fig. 2a). Further analysis of measurement data revealed that, when all other extruder parameters (i.e., extrusion temperature, extrusion speed, and pulling speed) were held constant, the diameter of ferromagnetic fibers decreased with increasing TPU hardness, but decreased with increasing NdFeB content. These effects were due to the fact that increasing TPU hardness causes a notable decrease in viscosity; moreover, adding NdFeB powder reduced the viscosity of the mixed material when the hardness of TPU was held constant. In such a case, the front of the extrusion mold will melt more fluidly and be easier to stretch under constant traction force of the pulling machine due to the reduced viscosity of the mixed material. In addition, it was discovered that as the NdFeB content increased to >30% (mass fraction) during the preparation process, the uniformity and smoothness of the resulting fibers significantly declined. This was caused by greater NdFeB content being correlated with greater dispersal in sampled ferromagnetic fiber diameter, i.e., lower uniformity.

Figure 2b shows the density of TPU/NdFeB-based ferromagnetic fibers with different TPU hardness and NdFeB content. These results show that ferromagnetic fiber density increased as NdFeB content increased. In addition, TPU hardness had little effect on the density of the ferromagnetic fibers when the extrusion temperature, extrusion speed, and pulling speed of the extruder were held constant. This is due to the fact that NdFeB has a far higher density than TPU; hence, when extruding a fiber, the mass per unit length of the fiber increases much more with increasing NdFeB content than with increasing TPU content.

Ferromagnetic fibers are highly suitable for commercial desktop FDM 3D printers. Printing ferromagnetic fibers requires printing at temperatures just above the melting point and at low printing speeds so that they can be fully cooled. Shapes can be formed when the fibers showed high viscosity, ease of stretching, and softness at high temperatures. It was found that about 50 °C for the printing platform, about 205 °C for the printing temperature, about 15 mm/s for the printing speed, 0.2 mm for the layer height, 100% infill density, and

Fig. 2 Characterization of ferromagnetic fibers. **a** Diameter of ferromagnetic fibers with different NdFeB content and thermoplastic polyurethane (TPU) hardness levels (data are expressed as mean \pm standard deviation, $n=9$). **b** Density of ferromagnetic fibers with different NdFeB contents and TPU hardness levels. **c** Nozzle plugging rate and qualified rate at different NdFeB content levels. **d** Stress–strain curves of ferromagnetic fibers with different NdFeB content levels. Three samples were prepared for each test. **e** Recycling of ferromagnetic fibers. **f** Stress–strain curves of recycled ferromagnetic fibers at different cycles



0.4 mm for the nozzle diameter were ideal parameters for printing ferromagnetic fibers (90 A).

Next, ferromagnetic fibers with NdFeB contents of 10%, 20%, 30%, and 40% (mass fraction) were printed using high hardness (90 A) TPU. These fibers were able to maintain a straight orientation when heated and extruded, and we found that NdFeB content had little impact on the wire drawing pattern. Moreover, Fig. 2c shows that the nozzle plugging rate of the mixed material with an NdFeB content of 40% (mass fraction) was too high and is, therefore, unsuitable for actual printing. Figure 2c shows that qualified rates after 50 samples with NdFeB contents of 10%, 20%, 30%, and 40% (mass fraction) were printed. Figure S3 (Supplementary Information) shows unqualified samples with an NdFeB content of 40% (mass fraction). Thus, it is clear that the ideal NdFeB content for 3D printing ranges from 10% to 30% (mass fraction).

Tensile tests were then carried out to analyze the effect of doping NdFeB content on the mechanical performance of ferromagnetic fibers. To do so, standard tensile samples with a thickness of 1 mm with NdFeB contents of 10%, 20%, 30%, and 40% (mass fraction) were printed using 90-A-TPU-based ferromagnetic fibers. The resulting stress–strain curves

(Fig. 2d) from tensile tests demonstrated that the amount of NdFeB had a significant impact on the mechanical performance of different ferromagnetic fibers. Moreover, increased NdFeB content was found to weaken the elastic deformation properties of fibers, while low TPU content caused samples to have poor mechanical performance when the NdFeB content reached 40% (mass fraction).

Figure 2e shows the recycling process of the ferromagnetic fiber material. It is possible to re-melt and re-extrude ferromagnetic fibers for extrusion molding since TPU readily melts and has a good re-molding ability. Accordingly, we cut ferromagnetic fibers into slivers using a dicing cutter; these slivers were then added to an extruder and re-extruded to reform ferromagnetic fibers. The mechanical performance of standard samples printed from recycled ferromagnetic fibers showed an almost identical mechanical performance as original fibers. Moreover, this continued to be true even after five recycling cycles (Fig. 2f), thereby demonstrating that the proposed fibers are highly recyclable and can be used to create sustainable soft robots.

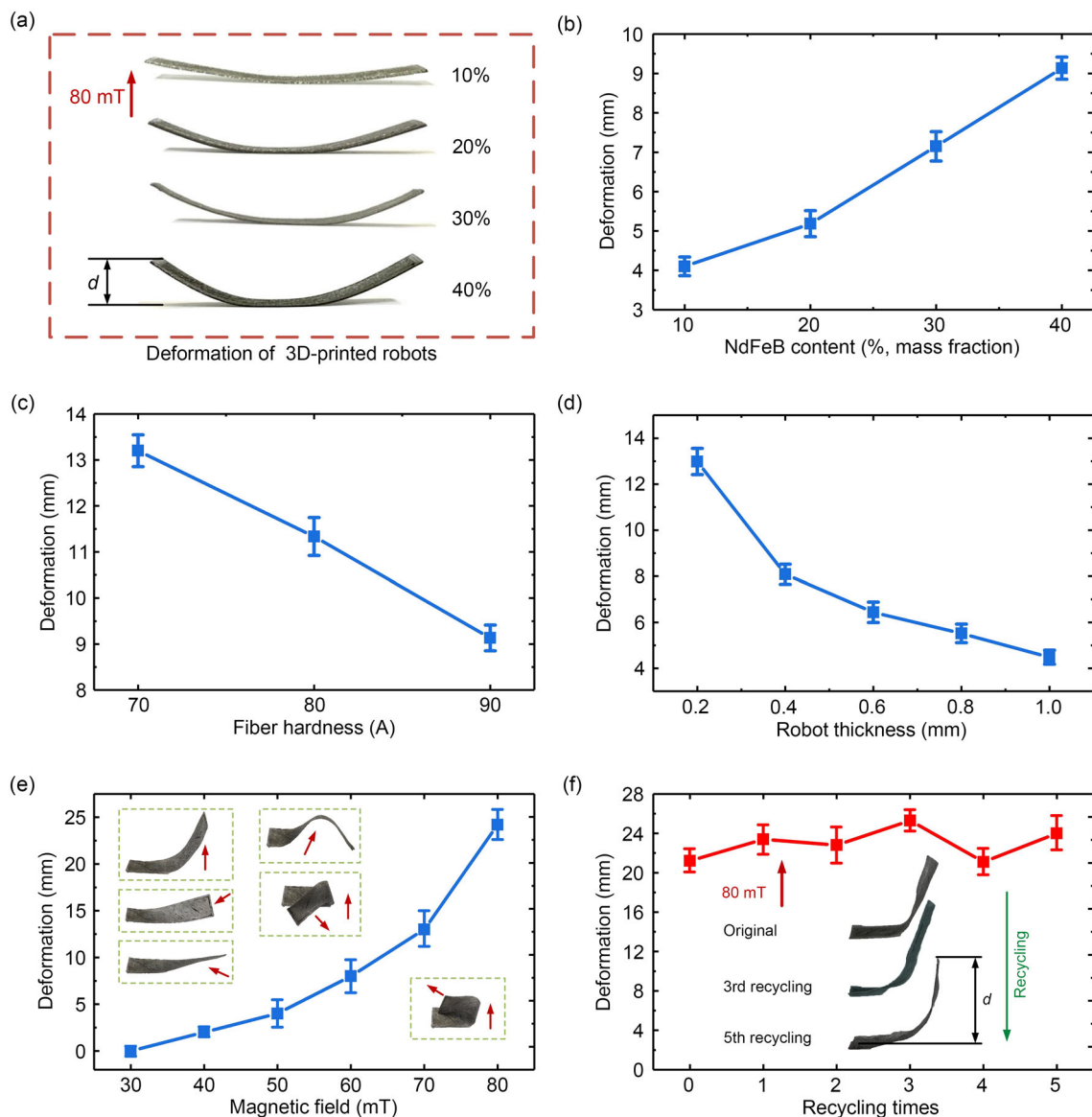


Fig. 3 Characterization of magnetic soft robots. **a** Deformation of 3D-printed soft robots. **b** Deformation of robots with different NdFeB contents. **c** Deformation of robots with varying fiber hardness. **d** Deformation of robots with varying robots thickness. **e** Deformation of robots

in different magnetic field sizes and directions. **f** Recycling performance of 3D-printed robots. Data are expressed as mean±standard deviation ($n=9$)

Printable and recyclable magnetic soft robots

Next, we evaluated the use of fibers in creating printable soft robots. Here, soft robots were created using a desktop FDM 3D printer, and a magnetization machine was then used to complete robot magnetization by magnetizing NdFeB particles within the printed material. This magnetization causes the robot to experience magnetic moments when placed in a magnetic field, thereby causing pre-programmable deformation. Next, to optimize the FDM 3D printing of ferromagnetic fibers, we improved the nozzle of the printer and installed

a thin tube with an inner diameter of 1.85 mm between the drive mechanism and the heating block. In general, soft robot deformation is influenced by the properties of the ferromagnetic fibers. Thus, to determine the impact of NdFeB content on robot deformation when a magnetic field was applied, four soft robots with NdFeB contents of 10%, 20%, 30%, and 40% (mass fraction) were printed using ferromagnetic fibers. Each of these robots had dimensions of 10 mm×50 mm and a thickness of 0.5 mm. Figure 3a shows the deformation of 3D-printed soft robots in an 80-mT magnetic field. We found that higher percentages of NdFeB particles were associated

with better magnetic actuation performance as well as a larger degree of soft robot deformation in response to the magnetic field (Figs. 3a and 3b). The hardness of the ferromagnetic fibers also affected the deformation of soft robots. We tested soft robots printed on fibers with substrates of 70 A, 80 A, and 90 A TPU hardness, and found that the smaller the fiber hardness, the larger the robot deformation (Fig. 3c). In addition, we investigated how the thickness of the robot affected its ability to deform. As thickness increased, the robot stiffened and lost its capacity to deform; thus, we concluded that the thinner the robot, the larger the potential degree of robot deformation (Fig. 3d). Figure 3e shows robot deformations in different magnetic field strength and directions. Robot deformation increased with increasing magnetic field strength according to changes in the direction of the magnetic field.

The aforementioned soft robots are recyclable due to the high degree of recyclability of the ferromagnetic fiber material. Thus, once their tasks have been completed, these robots can be recycled by cutting them into slivers and re-extruding them into new fibers using an extruder. Robot deformation after five cycles of cycling is shown in Fig. 3f. Here, we observe that the robots exhibit deformation performances that are similar to those of the original robot in each new cycle. This demonstrates the ability of the soft robot to recover motion and deformation quickly even after several cycles of recycling.

To demonstrate that ferromagnetic fibers can be used for the direct printing of various magnetic soft robots, 2D soft robots with different magnetization patterns and different shapes were printed. These printed robots can perform various deformations in a magnetic field within 0.5 s after being magnetized by pre-deformation in a magnetizer (Figs. 4a–4f and Movie S4 in Supplementary Information). The 2D soft robots can be used to design biomimetic robots, large deformation displays, and moving objects in magnetic fields. This technique for shaping and controlling soft robots allows them to quickly change from a simple 2D planar structure to a complex 3D shape that is fully reversible and untethered. In addition, magnetic domain programming techniques can achieve complexity, selectivity, and flexibility in controlling soft robot deformation. Finally, soft robots can achieve shape programmability via demagnetization and re-magnetization using different magnetization profiles.

Our fiber-to-robot approach can be further extended to complex 3D soft robots. The high printability of fiber-based materials makes it very simple to print 3D structures. Different 3D soft robots programmed with magnetic domains are capable of exhibiting complex shape changes when subjected to an applied magnetic field, including collapse, elongation, contraction, and aggregation (Figs. 4g–4j and Movie S5 in Supplementary Information), thereby demonstrating that ferromagnetic fibers offer a viable pathway for printing complex

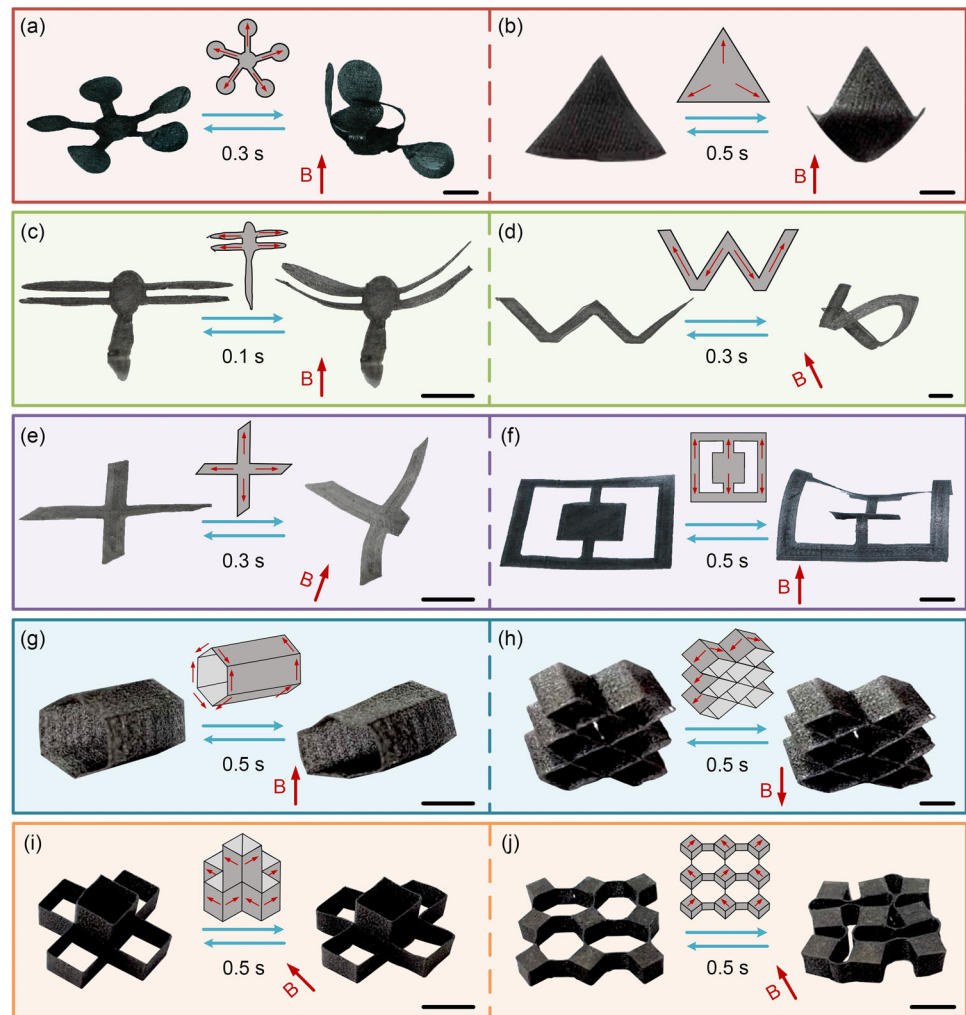
and sustainable 3D soft robots that can be used for metamaterial design and object manipulation.

Applications of sustainable soft robots

Robots controlled by magnetic fields can be operated wirelessly, while 3D printing enables the creation of personalized robots; combining these technologies further allows for various functions. For example, a lattice-shaped soft robot can be used as a miniature lifting frame due to its ability to contract in the vertical direction under a magnetic field. Here, we show how a lattice-shaped soft robot can lift and lower a small bead (Fig. 5a and Movie S6 in Supplementary Information). Here, the weight of the small bead is about 0.3 g. When a magnetic field is applied, the robot lowers the height of the small bead by 3 mm within 0.5 s. When the magnetic field is removed, the robot quickly extends upward to restore its original shape, and the small bead returns to its original location. In addition, we printed a magnetic soft gripper; this consisted of a 2D soft gripper that can be transformed into a 3D soft gripper when a magnetic field is applied (Fig. 5b). This property allows it to grasp and release a variety of objects. Figure 5c depicts the gripper's maximal horizontal diameter during magnetic field manipulation, which reflects the gripper's degree of actuation. The gripper nearly completely lacks deformation when the magnetic field strength is less than 40 mT, which is defined as a non-driven region. In contrast, the gripper's diameter steadily reduces as magnetic field intensity rises to between 40 and 65 mT, indicating that the degree of gripper drive gradually increased, which is defined as a partly-driven region. The gripper is fully actuated and had a 26.82 mm diameter when the magnetic field intensity was greater than 65 mT. Figure 5d and Movie S7 (Supplementary Information) show the gripper's process used for grasping a ball. After conducting several experiments, we found that the gripper can grip balls with diameters ranging from 8 to 34 mm, and the gripper's maximum load capacity is 2.25 g, i.e., 2.4 times its own weight. In addition, the flexibility of the gripper allows the gripper to adapt to different object shapes without causing damage to objects (Fig. 5e and Movie S7 in Supplementary Information), thereby demonstrating the wide application prospects of the gripper.

In general, soft robots made from PDMS or Ecoflex substrates coupled with a magnetic powder cannot be reshaped once they have been created, which limits their further use. Here, we propose a different model using fibers and robots that are fully recyclable, allowing them to be repurposed using extruders and printers after completing tasks. For example, lattice-shaped soft robots and soft grippers can be chopped into slivers by a dicing cutter and then further processed into fibers by an extruder. New soft robots with new shapes, new patterns of re-programmable magnetization, and new functionalities can then be fabricated by printers, thereby

Fig. 4 Various 3D-printed magnetic soft robots. **a–f** Deformation of a 2D soft robot under applied magnetic fields. **g–j** Deformation of a 3D soft robot under applied magnetic fields. The magnetic field is about 80 mT. All scale bars are 10 mm

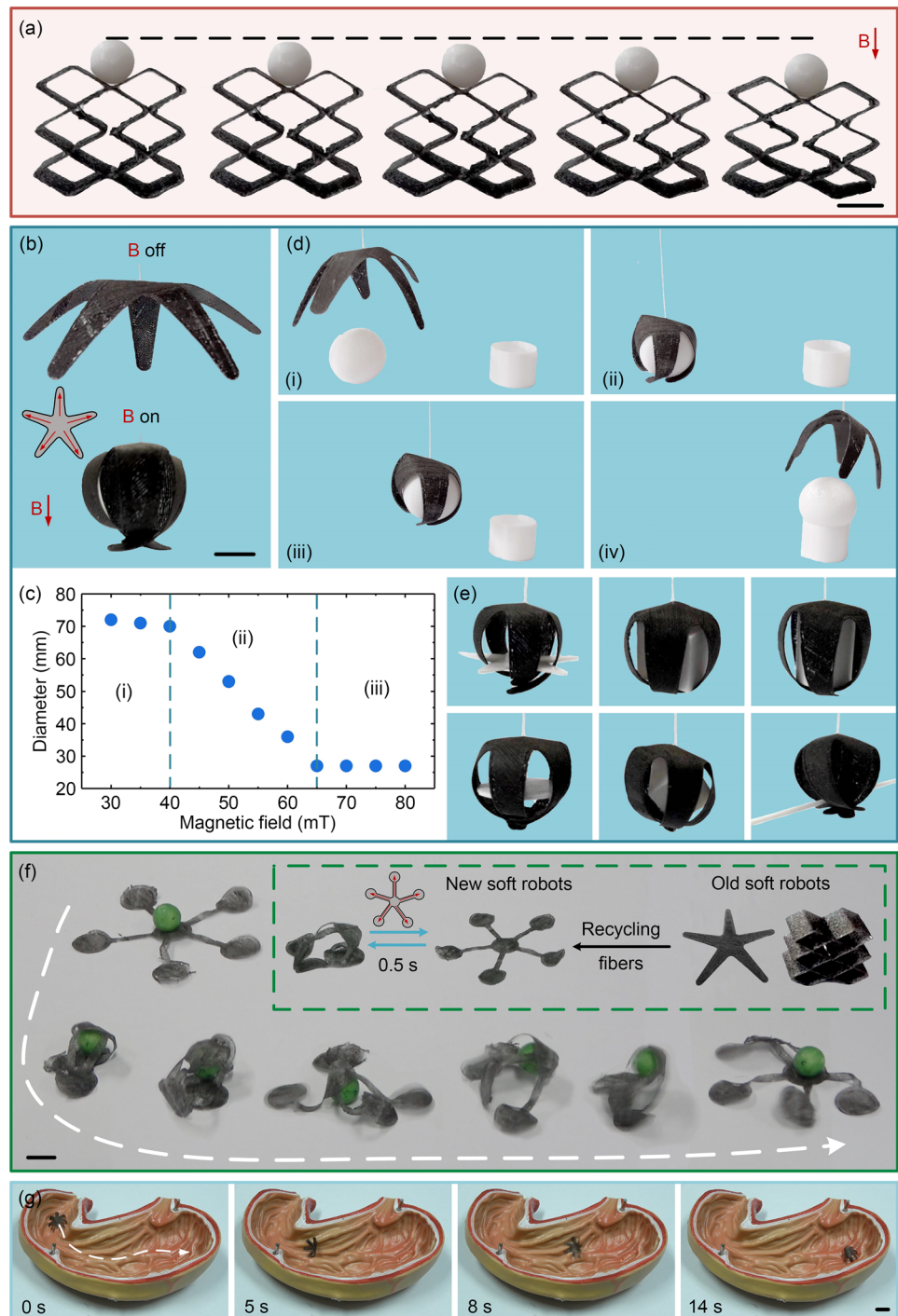


offering potential use cases beyond the fixed shapes and functionalities of conventional soft robots with highly targeted applications. The new robot can deform within 0.5 s, as illustrated in Fig. 5f and Movie S8 (Supplementary Information), and its actuation ability is nearly identical to the old robot. Furthermore, the ability of the new robot to carry objects (about 0.1 g) in a magnetic field and move quickly (Fig. 5f and Movie S8 in Supplementary Information) suggests that magnetic soft robots may one day carry and release medications in the medical field. Moreover, the high degree of recyclability may be important for resource conservation and environmental protection. To further exhibit the application prospects of this approach for medical applications, we printed a soft microrobot (size: 17 mm × 17 mm × 0.2 mm). This robot can be wirelessly controlled using an external magnetic field and can move about 15 cm in 14 s in a human stomach model (Fig. 5g and Movie S9 in Supplementary Information).

Discussion

In this study, we suggested using ferromagnetic fibers to print soft robots using FDM 3D printers. We used TPU/NdFeB hybrid particles to produce ferromagnetic fibers that were highly recyclable and sustainable. Once these ferromagnetic fibers were created, their printability, mechanical performance, and recycling performance were verified via a series of experiments. Ferromagnetic fibers can be produced using an extruder, a tried-and-true industrial technology that is suitable for commercial desktop 3D printers. Moreover, 2D- and 3D-printed soft robots, unlike magnetic soft robots that use cross-linked PDMS or Ecoflex as a base material [30–32, 41], can be directly recycled in one cycle [42] and reprinted into new robots once their tasks are completed. We experimentally verified that the new robots show almost no differences in actuation ability related to unrecycled robots, and were able to adopt new functions to be utilized in new application scenarios, thereby demonstrating a high degree of sustainability. In addition, we chose to use FDM printing, which

Fig. 5 Applications of sustainable soft robots. **a** A lattice-shaped soft robot is used to lift and lower a small bead. **b** A magnetic soft gripper can create a grasping motion in response to the application of a magnetic field. **c** The gripper's maximal horizontal diameter during magnetic field manipulation. **d** The gripper's process for grasping a ball. **e** The gripper is used to grasp different object shapes. **f** Lattice-shaped soft robots and soft grippers are processed and reprinted to create a new robot. The new robot can move objects in a magnetic field. **g** The locomotion of a magnetic soft robot in a human stomach model. All scale bars are 10 mm



has notable benefits over DIW [36–39], including simplicity, convenience of use, low/no particle deposition, and FDM printing can be used to print complicated structures, and low cost. Using this approach, we were able to readily print soft robots of diverse structures, including complex 3D structures. These untethered soft robots were able to operate wirelessly and adopt a wide range of uses including lifting, gripping,

and moving objects. In addition, we demonstrated locomotion of a magnetic soft robot in a human stomach model to show the promise of this application for medical applications. Finally, combining 3D printing with machine vision [43–45] is another important future research direction. Overall, our fully recyclable ferromagnetic fibers described here permitted the printing and reprinting of sustainable soft robots

while also significantly reducing material waste, suggesting an important new sustainability strategy for soft robotics.

Methods

Materials and fabrication

The matrix materials used to produce ferromagnetic fibers were chosen from three TPU particles with different harnesses (i.e., 70 A, 80 A, and 90 A, China Dongguan Guangyuan Plastic Co., Ltd.), whose main parameters are shown in Table S1 (Supplementary Information). The main parameters of the NdFeB particles (38 μm in diameter, China Guangdong Guangzhou New Nord Co., Ltd.) used as the magnetic filler for the ferromagnetic fibers are shown in Table S2 (Supplementary Information). A single-screw extruder (Wellzoom model C) that comprises a main machine, a water cooler, a pulling machine, and a reeling machine was used to extrude ferromagnetic fibers. This apparatus is shown in Fig. S1 and Movie S1 (Supplementary Information).

Criteria for the selection of extruder parameters

At low temperatures, the TPU particles did not melt sufficiently in the extruder, resulting in difficulties in fiber extrusion. However, when the extruder's temperature was too high, TPU particles softened excessively, which caused TPU to adhere to the extrusion mold and fail to plasticize. This, in turn, prevents the formation of fibers of a fixed diameter size. After multiple temperature adjustment experiments, we found that the temperature control I parameter should be set to about 205 °C and temperature control II to about 200 °C. These temperatures not only provided sufficient heat for the melting and mixing of TPU/NdFeB particles, but also ensured adequate reshaping performance of all materials.

Next, we optimized the extrusion speed, pulling speed, and other fiber production parameters. When the extrusion speed remained constant, an increase in pulling speed would lead to a decrease in fiber diameter. However, if the extrusion speed was high, the extruded fiber would accumulate in the water cooler tank due to the lack of pulling force and could not form a fiber. If the pulling speed was too high, it would cause the extruded fiber to become excessively fine or cause breaks. When the pulling speed remained constant, an increase in extrusion speed would lead to an increase in fiber diameter. We found that pulling materials at a speed slightly lower than the extrusion speed was effective; this could prevent necking or expansion of materials. In addition, when the extrusion speed was high, the consistency of fiber extrusion was inadequate and operation was difficult; however, when

the extrusion speed was too low, the efficiency of fiber fabrication was too low. Thus, taking all of these factors into consideration, we chose to set the extrusion speed to about 9.5 r/min and the pulling speed to about 30 r/min, and this was able to stabilize the extrusion of fibers with a diameter of about 1.75 mm.

Magnetization of soft robots

A mold was used to keep soft robots in place during pre-deformation. Next, a magnetizing machine was used to magnetize the robot. When the robot was to be recycled, it was demagnetized at a temperature of about 100 °C before being sliced into pieces.

Actuation of soft robots

The magnetic field generator consisted of three perpendicular Helmholtz coils, each made of AWG22 (0.67-mm-diameter enameled copper wire) wound on an aluminum alloy sheet coil holder. This device was able to generate magnetic fields in three directions (i.e., along the x , y , and z axes) and could be used to remotely actuate magnetic soft robots.

Supplementary Information The online version contains supplementary material available at <https://doi.org/10.1007/s42242-024-00303-4>.

Acknowledgements This work was funded by the International Cooperation Program of the Natural Science Foundation of China (No. 52261135542) and Zhejiang Provincial Natural Science Foundation of China (No. LD22E050002). VVG and EYK are grateful to the Russian Science Foundation (No. 23-43-00057) for financial support.

Author contributions WT, JZ, and EYK proposed and supervised the project. WT designed the research. WT, YDG, and ZYD conducted the experiments. WT, YDG, and VVG analyzed the data. DH provided the magnetic devices and magnetization method. WT wrote the manuscript. WT, EYK, and JZ revised the manuscript. All authors participated in discussions of the research and revisions of the manuscript.

Declarations

Conflict of interest JZ is an associate editor and DH is an academic editor for *Bio-Design and Manufacturing*. They were not involved in the editorial review or the decision to publish this article. The authors declare that they have no conflict of interest.

Ethical approval This study does not contain any studies with human or animal subjects performed by any of the authors.

Data availability All data are available from the corresponding authors upon reasonable request.

References

- Awasthi AK, Li J, Koh L et al (2019) Circular economy and electronic waste. *Nat Electron* 2:86–89. <https://doi.org/10.1038/s41928-019-0225-2>
- Shittu OS, Williams ID, Shaw PJ (2021) Global E-waste management: can WEEE make a difference? A review of e-waste trends, legislation, contemporary issues and future challenges. *Waste Manag* 120:549–563. <https://doi.org/10.1016/j.wasman.2020.10.016>
- Peng P, Shehabi A (2023) Regional economic potential for recycling consumer waste electronics in the United States. *Nat Sustain* 6(1):93–102. <https://doi.org/10.1038/s41893-022-00983-9>
- Tembhare SP, Bhanvase BA, Barai DP et al (2022) E-waste recycling practices: a review on environmental concerns, remediation and technological developments with a focus on printed circuit boards. *Environ Dev Sustain* 24(7):8965–9047. <https://doi.org/10.1007/s10668-021-01819-w>
- Hartmann F, Baumgartner M, Kaltenbrunner M (2021) Becoming sustainable, the new frontier in soft robotics. *Adv Mater* 33(19):2004413. <https://doi.org/10.1002/adma.202004413>
- Titirici M, Baird SG, Sparks TD et al (2022) The sustainable materials roadmap. *J Phys Mater* 5(3):032001. <https://doi.org/10.1088/2515-7639/ac4ee5>
- Mohanty AK, Vivekanandhan S, Pin JM et al (2018) Composites from renewable and sustainable resources: challenges and innovations. *Science* 362(6414):536–542. <https://doi.org/10.1126/science.aat9072>
- Xu ZP, Wu MR, Gao WW et al (2022) A sustainable single-component “Silk nacre.” *Sci Adv* 8(19):eabo0946. <https://doi.org/10.1126/sciadv.abo0946>
- Bai XJ, Shang JZ, Luo ZR et al (2022) Development of amphibious biomimetic robots. *J Zhejiang Univ-SCI A (Appl Phys & Eng)* 23(3):157–187. <https://doi.org/10.1631/jzus.A2100137>
- Wang JR, Xi YX, Ji C et al (2022) A biomimetic robot crawling bidirectionally with load inspired by rock-climbing fish. *J Zhejiang Univ-SCI A (Appl Phys & Eng)* 23(1):14–26. <https://doi.org/10.1631/jzus.A2100280>
- Jin YB, Cheng SW, Yuan YY et al (2022) Anthropomorphic hand based on twisted-string-driven da Vinci’s mechanism for approaching human dexterity and power of grasp. *J Zhejiang Univ-SCI A (Appl Phys & Eng)* 23(10):771–782. <https://doi.org/10.1631/jzus.A2200216>
- Tang W, Zhang C, Zhong YD et al (2021) Customizing a self-healing soft pump for robot. *Nat Commun* 12(1):2247. <https://doi.org/10.1038/s41467-021-22391-x>
- Tang W, Lin YQ, Zhang C et al (2021) Self-contained soft electrofluidic actuators. *Sci Adv* 7(34):eabf8080. <https://doi.org/10.1126/sciadv.abf8080>
- Zhong YD, Tang W, Zhang C et al (2022) Programmable thermochromic soft actuators with “two dimensional” bilayer architectures for soft robotics. *Nano Energy* 102:107741. <https://doi.org/10.1016/j.nanoen.2022.107741>
- Zhong YD, Tang W, Xu HX et al (2023) Phase-transforming mechanical metamaterials with dynamically controllable shape-locking performance. *Natl Sci Rev* 10(9):nwad192. <https://doi.org/10.1093/nsr/nwad192>
- Tang W, Zhong YD, Xu HX et al (2023) Self-protection soft fluidic robots with rapid large-area self-healing capabilities. *Nat Commun* 14(1):6430. <https://doi.org/10.1038/s41467-023-42214-5>
- Wang D, Zhao BW, Li XL et al (2023) Dexterous electrical-driven soft robots with reconfigurable chiral-lattice foot design. *Nat Commun* 14(1):5067. <https://doi.org/10.1038/s41467-023-40626-x>
- Zhu PA, Tang W, Jiao ZD et al (2023) Liquid manipulator with printed electrode patterns for soft robotic systems. *Adv Mater Technol* 8(18):2300308. <https://doi.org/10.1002/admt.202300308>
- Fu L, Zhao WQ, Ma JY et al (2022) A humidity-powered soft robot with fast rolling locomotion. *Research* 2022:9832901. <https://doi.org/10.34133/2022/9832901>
- Zhang C, Zhu PA, Lin YQ et al (2021) Fluid-driven artificial muscles: bio-design, manufacturing, sensing, control, and applications. *Bio-Des Manuf* 4(5):123–145. <https://doi.org/10.1007/s42242-020-00099-z>
- Ma SQ, Jiang ZY, Wang M et al (2021) 4D printing of PLA/PCL shape memory composites with controllable sequential deformation. *Bio-Des Manuf* 4(4):867–878. <https://doi.org/10.1007/s42242-021-00151-6>
- Kim Y, Zhao XH (2022) Magnetic soft materials and robots. *Chem Rev* 122(5):5317–5364. <https://doi.org/10.1021/acs.chemrev.1c00481>
- Dong Y, Wang L, Zhang ZF et al (2022) Endoscope-assisted magnetic helical micromachine delivery for biofilm eradication in tympanostomy tube. *Sci Adv* 8(40):abq8573. <https://doi.org/10.1126/sciadv.abq8573>
- Zhou C, Yang YZ, Wang JX et al (2021) Ferromagnetic soft catheter robots for minimally invasive bioprinting. *Nat Commun* 12(1):5072. <https://doi.org/10.1038/s41467-021-25386-w>
- Yi SZ, Wang L, Chen ZP et al (2022) High-throughput fabrication of soft magneto-origami machines. *Nat Commun* 13(1):4177. <https://doi.org/10.1038/s41467-022-31900-5>
- Wang CX, Wu YD, Dong XG et al (2023) In situ sensing physiological properties of biological tissues using wireless miniature soft robots. *Sci Adv* 9(23):eadg3988. <https://doi.org/10.1126/sciadv.adg3988>
- Jin ZBY, He CF, Fu JZ et al (2022) Balancing the customization and standardization: exploration and layout surrounding the regulation of the growing field of 3D-printed medical devices in China. *Bio-Des Manuf* 5(3):580–606. <https://doi.org/10.1007/s42242-022-00187-2>
- Mohammadi M, Zolfagharian A, Bodaghi M et al (2022) 4D printing of soft orthoses for tremor suppression. *Bio-Des Manuf* 5(1):786–807. <https://doi.org/10.1007/s42242-022-00199-y>
- Hou YH, Wang WG, Bartolo P (2022) Application of additively manufactured 3D scaffolds for bone cancer treatment: a review. *Bio-Des Manuf* 5(3):556–579. <https://doi.org/10.1007/s42242-022-00182-7>
- Chung HJ, Parsons AM, Zheng LL (2020) Magnetically controlled soft robotics utilizing elastomers and gels in actuation: a review. *Adv Intell Syst* 3(3):2000186. <https://doi.org/10.1002/aisy.202000186>
- Bira N, Dhagat P, Davidson JR (2020) A review of magnetic elastomers and their role in soft robotics. *Front Robot AI* 7:588391. <https://doi.org/10.3389/frobt.2020.588391>
- Peng LL, Zhang YX, Wang J et al (2022) Slug-inspired magnetic soft millirobot fully integrated with triboelectric nanogenerator for on-board sensing and self-powered charging. *Nano Energy* 99:107367. <https://doi.org/10.1016/j.nanoen.2022.107367>
- Chen GS, Ma B, Zhang J et al (2023) Reprogrammable magnetic soft robots based on low melting alloys. *Adv Intell Syst* 5(10):2300173. <https://doi.org/10.1002/aisy.202300173>
- Zhao R, Dai HD, Yao HC (2022) Liquid-metal magnetic soft robot with reprogrammable magnetization and stiffness. *IEEE Robot Autom Lett* 7(2):4535–4541. <https://doi.org/10.1109/LRA.2022.3151164>
- Chen Y, Zhao X, Li Y et al (2021) Light- and magnetic-responsive synergy controlled reconfiguration of polymer nanocomposites with shape memory assisted self-healing performance for soft robotics. *J Mater Chem C* 9(16):5515–5527. <https://doi.org/10.1039/D1TC00468A>

36. Zhang LW, Zhao S, Zhou XZ et al (2023) A magnetic-driven multi-motion robot with position/orientation sensing capability. *Research* 6(33):0177. <https://doi.org/10.34133/research.0177>
37. Ma CP, Wu S, Ze QJ et al (2021) Magnetic multimaterial printing for multimodal shape transformation with tunable properties and shiftable mechanical behaviors. *ACS Appl Mater Interfaces* 13(11):12639–12648. <https://doi.org/10.1021/acsami.0c13863>
38. Yan CY, Zhang XQ, Ji ZY et al (2021) 3D-printed electromagnetic actuator for bionic swimming robot. *J Mater Eng Perform* 30(9):6579–6587. <https://doi.org/10.1007/s11665-021-05918-7>
39. Roh S, Okello LB, Golbasi N et al (2019) 3D-printed silicone soft architectures with programmed magneto-capillary reconfiguration. *Adv Mater Technol* 4(4):1800528. <https://doi.org/10.1002/admt.201800528>
40. Zhao DK, Xu HQ, Yin J et al (2022) Inkjet 3D bioprinting for tissue engineering and pharmaceuticals. *J Zhejiang Univ-SCI A (Appl Phys & Eng)* 23(12):955–973. <https://doi.org/10.1631/jzus.A2200569>
41. Kim Y, Yuk H, Zhao RK et al (2018) Printing ferromagnetic domains for untethered fast-transforming soft materials. *Nature* 558(7709):274–279. <https://doi.org/10.1038/s41586-018-0185-0>
42. Abdel-Bary EM (2003) *Handbook of Plastic Films*. Smithers Rapra Publishing. <https://chemtec.org/products/978-1-85957-338-9>
43. Shi XK, Wang Q, Wang C et al (2022) An AI-based curling game system for winter Olympics. *Research* 2022:9805054. <https://doi.org/10.34133/2022/9805054>
44. Qin KC, Tang W, Zhong YD et al (2023) An aerial–aquatic robot with tunable tilting motors capable of multimode motion. *Adv Intell Syst* 5(11):2300193. <https://doi.org/10.1002/aisy.202300193>
45. Buchner TJK, Rogler S, Weirich S et al (2023) Vision-controlled jetting for composite systems and robots. *Nature* 623(7987):522–530. <https://doi.org/10.1038/s41586-023-06684-3>

Springer Nature or its licensor (e.g. a society or other partner) holds exclusive rights to this article under a publishing agreement with the author(s) or other rightsholder(s); author self-archiving of the accepted manuscript version of this article is solely governed by the terms of such publishing agreement and applicable law.