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ORIGINAL ARTICLE

Sustainable 4D printing of magneto-electroactive shape memory polymer composites

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Abstract
Typical techniques for creating synthetic morphing structures suffer from a compromise between quick shape change and geometric complexity. A novel approach is proposed for encoding numerous shapes and forms by magneto-electroactive shape memory polymer composite (SMPC) structures and integrating sustainability with 4D printing (4DP) technology. Electrically driven, remote controllability, and quick reaction are the features of these sustainable composite structures. Low-cost 4D-printed SMPC structures can be programmed remotely at high temperatures to achieve multi-stable shapes and can snap repeatedly between all programmed temporary and permanent configurations. This allows for multiple designs in a single structure without wasting material. The strategy is based on a knowledge of SMPC mechanics, magnetic response, and the manufacturing idea underlying fused deposition modelling (FDM). Iron-filled magnetic polylactic acid (MPLA) and carbon black-filled conductive PLA (CPLA) composite materials are investigated in terms of microstructure properties, composite interface, and mechanical properties. Characterisation studies are carried out to identify how to control the structure with a low magnetic field. The shape morphing of magneto-electroactive SMPC structures is studied. FDM is used to 4D print MPLA and CPLA adaptive structures with 1D/2D-to-2D/3D shapeshifting by the magnetic field. The benefits of switchable multi-stable structures are reducing material waste and effort/energy and increasing efficiency in sectors such as packaging.

Keywords 4D printing · 3D printing · Sustainability · Shape memory polymers · Composite materials · Fused deposition modelling

1 Introduction

A fast-developing new field of study known as 4D printing (4DP) technology focuses on the additive production of shape memory structures and smart materials [1, 2]. In 4DP, the structure is designed to self-assemble in the absence of traditional driving equipment [3, 4]. Creative concepts may be directly encoded into diverse structures via a modelling process, giving 4D-printed objects more design freedom [5, 6]. The benefits of 4DP include the ability of printed objects to change their form over time [7]. Products can be printed in a simple shape and 4DP allows them to achieve complex shapes without wasting material for the support structure. Hence, green design for complex products can be achieved easily. In reaction to one or more environmental stimuli, such as heat, light, water, and electrical and magnetic forces, 4D-printed structures can revert to their original shape [8]. Shape memory polymers (SMPs), shape memory polymer composites (SMPCs), and shape memory alloys (SMAs) have received increased interest in recent years because of their inexpensive cost, ease of production, and significant reversible deformation capabilities [9–11].

SMPs and SMPCs have been widely employed in industries because of their unique properties [12]. SMPs are stimuli-responsive materials that can be converted into a temporary shape when heated over the transition temperature. Under the same external force, the temporary form can be fixed during cooling [13, 14]. SMPs recover from the temporary shape to the original shape and complete one shape memory cycle when they are heated above the transition temperature. SMPCs are made up of SMPs and other functional materials, and they have better mechanical characteristics and respond quickly to external stimuli [15, 16]. 4DP is a highly specialised process in which varied printing

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equipment and procedures necessitate unique material property optimization [17, 18]. Fused deposition modelling (FDM) is a suitable 3D printing process to print low-cost products with high performance [19–21].

Stimuli-responsive techniques have a substantial influence on the utilisation of SMPCs in numerous sectors. Due to the Joule heating effect, electroactive SMPCs may regain their form and are less vulnerable to external impacts [22, 23]. Many approaches have been established to manufacture electroactive SMPCs [24]. Dong et al. [25] created electroactive PLA/carbon nanotube-based composites for different smart devices with remote control capabilities using the FDM technique. The researchers investigated how different SMPC-based complex structures formed memorised forms in 2D and 3D in response to electric stimulation. The results showed that increasing carbon nanotube content enhanced the thermal conductivity, electrical conductivity, and form recovery ratio of SMPC up to a point.

Mitkus et al. [26] described bi-layer SMP/conductive polylactic acid (CPLA) architectures, factors that influenced how they morphed, and approaches for improving both controllability and maximum potential. When various activation voltages were given to the structure, the results revealed a significant decrease in resistance. Wang et al. [27] developed a low-cost, electrically driven, reversible actuation and sensing method based on CPLA and FDM. Several applications of this newly designed actuator were shown to demonstrate the possibilities of electroactive polymers. Lee et al. [28] used CPLA to print four actuators at different printing rates that were actuated by Joule heating, resulting in increasing bending as the printing speed rose. The structures were activated by gradually heating them from 30 to 80 °C while allowing electrical current to pass through them.

Moreover, magnetic fields may easily and safely permeate SMPCs, providing a safe and effective actuation approach [29, 30]. Magneto-active materials, which are primarily created by integrating magnetic particles or discrete magnets into PLA material, have a high application potential for soft sensors and actuators due to the benefits of magnetic field control [31]. SMPCs can be programmed and stimulated remotely to achieve the desired shape. For example, Zhao et al. [32] developed a customised tracheal scaffold that restored its form in 35 s under a 30 kHz alternating magnetic field. Furthermore, magneto-responsive materials are promising for occlusion devices in congenital cardiac disorders due to biodegradability, shape memory effect (SME), remote controllability, and quick reaction.

Zhang et al. [33] showed a variety of shape memory capabilities of 4D-printed PLA/Fe_3O_4 composite actuators. The shape recovery mechanism was studied at a certain temperature and magnetic field. A magnetic field of 27.5 kHz was used to actuate a bone tissue-shaped structure printed with PLA/Fe_3O_4 composite filaments containing 15% Fe_3O_4. According to the findings, 4D-printed magnetic structures offered a high potential for functioning in biological and medical applications. Riley et al. [34] represented a substantial development in the programming of various permanent forms in SMP architectures, allowing for quick, reversible shape changes without reprogramming. They used a combination of PLA and magnetic PLA (MPLA) for the remote activation of snap-through via magnetic fields.

4DP can be used to eliminate issues in printing procedures such as staircase defects and support structure [35, 36]. Reduction of material in the printing procedure decreases the production cost and printing time accordingly. Shape-morphing structures, which may alter their forms from one state to another, can be achieved using the 4DP technique [37, 38]. A common case is morphing from an initial flat 2D shape to a 3D goal shape [39]. The structure can be printed in a 1D shape and then converted into the required 2D shape. The same procedure from 2 to 3D can be achieved using this technique [40]. This technique helps to reduce material wastage by eliminating the support structure in 3D and complex shapes. Changing the geometry of a single structure into various shapes helps to decrease the material usage in the printing procedure. Hence, developing sustainable smart structures leads to saving costs and energy consumption.

Morphing structures in engineering applications have the potential to greatly increase design efficiency and customization across various sectors [41, 42]. Remote reconfiguration allows dynamic structures to react to external stimuli without the requirement for traditional sensor and actuation systems for different applications [43, 44]. Morphing utilising pre-strain domains allows for rapid, pre-programmed form change in response to external stimuli. Typical techniques for creating synthetic morphing structures suffer from a compromise between quick shape change and geometric complexity. Also, 3D printing of a structure with complex shapes is difficult, time-consuming, and material wastage is high. The magneto-electroactive SMPCs can solve these issues due to the discussed capabilities in terms of a fast response and shape changing. This technique eliminates the hot water programming which affects the mechanical properties of PLA materials.

Despite the various advantages that electroactive and magneto-responsive polymers bring in 4DP, a combination of low-cost magneto-electroactive 4DP using FDM technology has not been explored yet. Also, a magnetic field is always required to activate the magnetic structure and hold them in the proper position [45, 46]. It is also yet to be determined how effectively magneto-electroactive 4DP could work in terms of sustainability and green design. The present study is the first of its kind to integrate CPLA and MPLA components to 4D print low-cost sustainable bi-stable, reversible, light composite structures using the FDM process. A composite adaptive structure of CPLA and MPLA is developed, which is driven by permanent magnets and operates at a low magnetic field and Joule heating approach. The technique proposed in this paper helps to eliminate the magnetic field after actuation due to the combination of stimuli.

This research introduces a novel smart composite structure that can be programmed remotely. The primary aims of this study are to emphasise several significant green design aspects, provide a conceptual design for sustainability, and thoroughly detail the 4DP technique utilised to create shape-memory structures and shape morphing. The model and solution approaches are expected to be effective in developing light and green 4D-printed magneto-electroactive structures with excellent stability. Shape morphing of 4D-printed structures is evaluated to investigate their capabilities in different applications. While prior research on this topic focused on chemistry-based techniques, 4D printing is employed to regulate the structure's topology. Figure 1 depicts the shape memory effect and remote programming, and how this research work is organised. CPLA's and MPLA's mechanical characteristics and microstructure features are investigated. Additionally, details about the electroactive material and shape-changing effect of 4D-printed CPLA and MPLA are provided. The created actuator's application is then suitably evaluated.

2 Materials and methods

2.1 Materials

A range of thermoplastic filaments that currently exist and may incorporate functional additive particles can be employed in the application of FDM 4DP to construct items with a variety of material characteristics and multifunctional features. These properties may be customised across the print by purposely changing the print at precise points. In this work, electrically CPLA (Proto-Pasta, Proto Plant, USA) and iron-filled MPLA (Proto-Pasta, Proto Plant, USA) are employed. This is because PLA is commercially available and has a noticeable SME when compared to other well-known SMPCs. CPLA is made up of electrically conductive carbon black (around 21 wt%) and PLA. Therefore, CPLA is an excellent choice for low-voltage applications such as circuits, touch sensors, and using prints to interact with

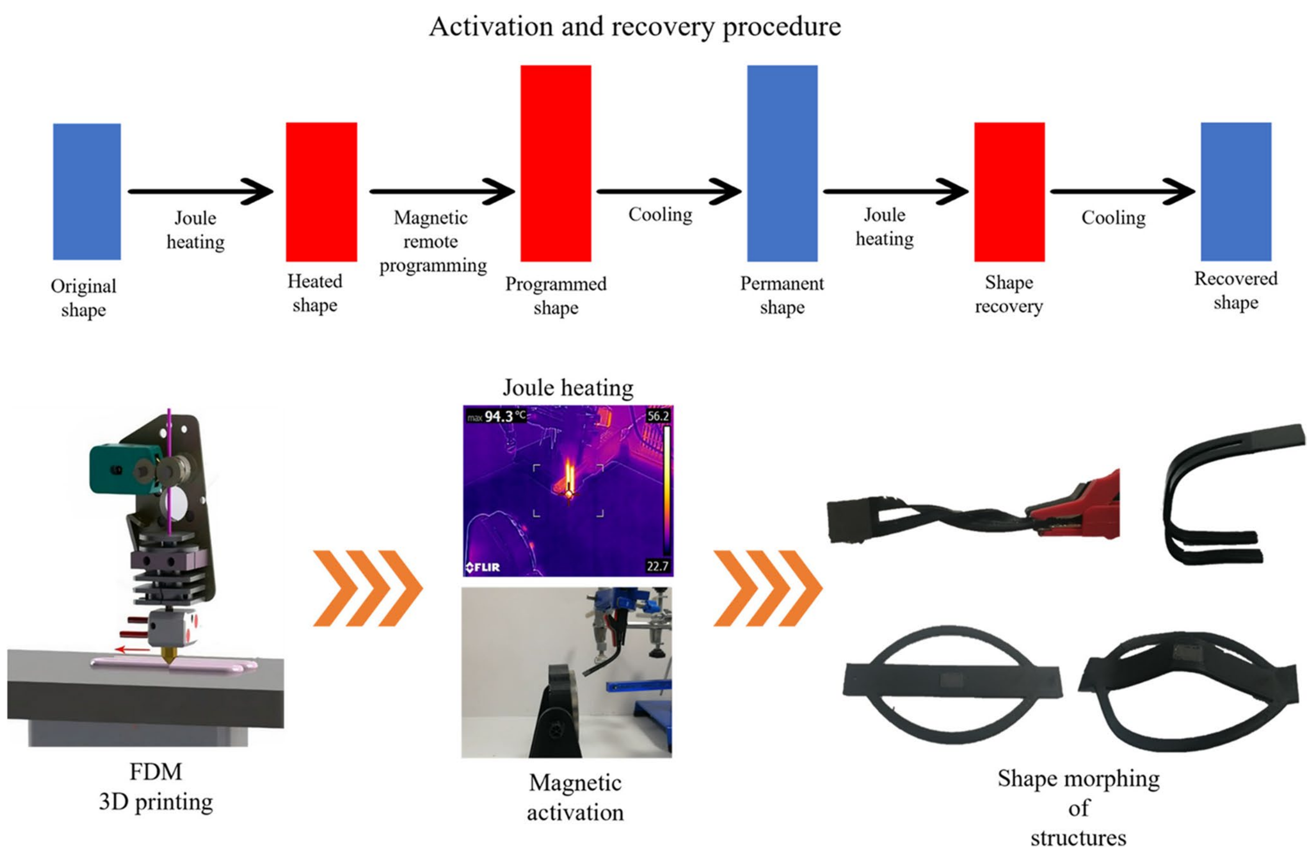

Fig. 1 A schematic of the remote programming and proposed 4DP method. The structure is heated through Joule heating. The heated shape is programmed via the magnetic field. The smart structure is cooled down in an activated form. The actuator is activated via Joule heating to recover its original shape

touch displays. Also, MPLA contains metal powder with 250 microns particle size with PLA-based resin. Young's modulus is reported to decrease by up to 98.6% when heated from room temperature to 80 °C [34, 47].

The magnetic characteristics, mechanical properties, and deformation of materials are used to examine this design space. Scanning electron microscopy (SEM) using a JSM-7100F LV FEG SEM (JEOL, Tokyo, Japan) is used to examine the microstructure and properties of CPLA and MPLA printed samples. Surface elemental analysis with energy dispersive spectroscopy (EDS) (Oxford Instruments, UK) is used for the semi-quantitative determination of chemical composition. The analysis is conducted on bonding between the 3D-printed CPLA and MPLA to evaluate the particles' distribution.

2.2 Design and 3D printing

A combination of CPLA and MPLA materials is employed as rapid and stable actuators, as well as 3D objects with morphing capabilities via remote control. The usage of 4D-printed construction is designed to offer the thinnest and most two-way actuator conceivable. The FDM technique can be used to produce smart structures with the capability of shape changing and shape recovery [48–50]. All the structures in this study are created using an open-source customised FDM-type 3D printer with two 0.4 mm nozzles. Figure 2 depicts the 4D-printed structure and its design with 1 mm thickness which is developed by SolidWorks software. The structure's design is centred on the phase transitions of CPLA and MPLA. The chosen layouts improve heat diffusion throughout the structures. The conductivity and resistance characteristics of CPLA are described previously [51, 52]. Slic3r software is used to slice the computer-aided design (CAD) file and converts it to.gcode files. To guarantee smooth nozzle extrusion, the printing parameters are described in Table 1. The concentric filling strategy is employed to ensure internal continuity. The models are merged to print both MPLA and CPLA material.

Magnetic flux density on the surface of the samples in the out-of-plane direction is used to analyse the magnetic characteristics of the 4D-printed composite structure. It is critical to maintain the actuator in position without altering its rigidity or weight. A structure using CPLA and MPLA is developed to eliminate the magnetic field and achieve a stable design after activation. In particular, the method is

Table 1 3D printing parameters of CPLA and MPLA

Parameters	Value
Layer height (mm)	0.2
Infill density (%)	100
Infill pattern	Concentric
Extruder temperature (°C)	210
Bed temperature (°C)	60
Material flow (%)	100
Printing speed (mm/s)	70

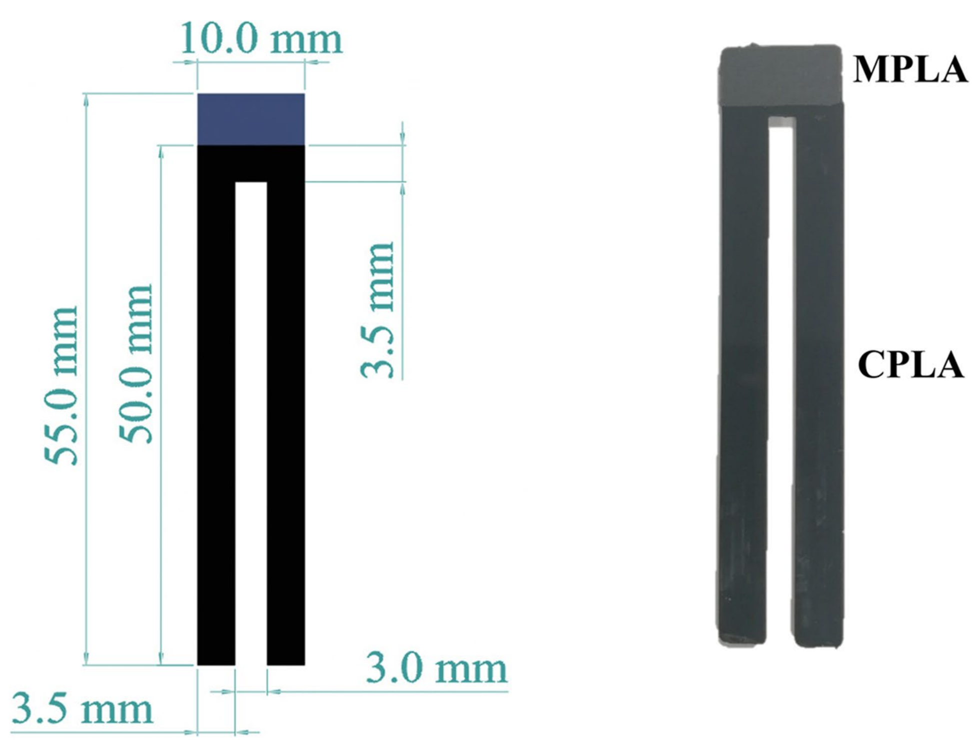

Fig. 2 The details of 3D design and 4D-printed structure

ideally suited for the creation of actuators with two-way behaviour and shape recovery capabilities. The applied magnetic field may be controlled by adjusting the distance between the magnet and the 4D-printed actuator. By activating the 4D-printed structure using Joule heating and providing a modest magnetic field, the actuators are triggered accordingly.

2.3 Electrical measurements

Printed CPLA on the power cables should provide a mechanically reliable connection at room temperature. For most applications, a readily breakable connection is also desired. The required electricity is supplied by the DC power source (RS Components Ltd, UK). The voltage for experimental circuits without extra electrical protection is limited to 120 VDC. Crocodile clamps and wires on exposed terminals are commonly used in labs for semi-permanent or temporary connections. A digital multimeter (Keithley Instruments, USA) is used to measure the current entering the CPLA structure during activation and Keithley KickStart software (Keithley Instruments, USA) reads the data. An infrared camera from FLIR (Teledyne FLIR LLC, USA) is used to assess the form memory capacity of the 4D-printed structures. The applied voltage is increased from 60 to 120 V to ensure that the structure is heated over its glass transition temperature (T_g). The wires are connected, one specimen at a time is positioned in the test stand, and the ensuing test procedure is used (see Fig. 3). A test is used to investigate SMEs of 4D-printed specimens. The shape-fixing method comprises applying an external force to the specimen using a permanent magnet. The deformations state above T_g and maintain that magnetic force after the power is turned off until the specimen cools and hardens. A video camera captures the shape recovery behaviour, which is then quantified using metrics such as recovery time.

2.4 Magnetic field

The deflection of the 4D-printed composite structure is tested in the presence of an external lateral magnetic field from the permanent magnet to analyse the actuator's responsiveness. To analyse the actuator's behaviour, the magnetic field strength between the actuator and the permanent

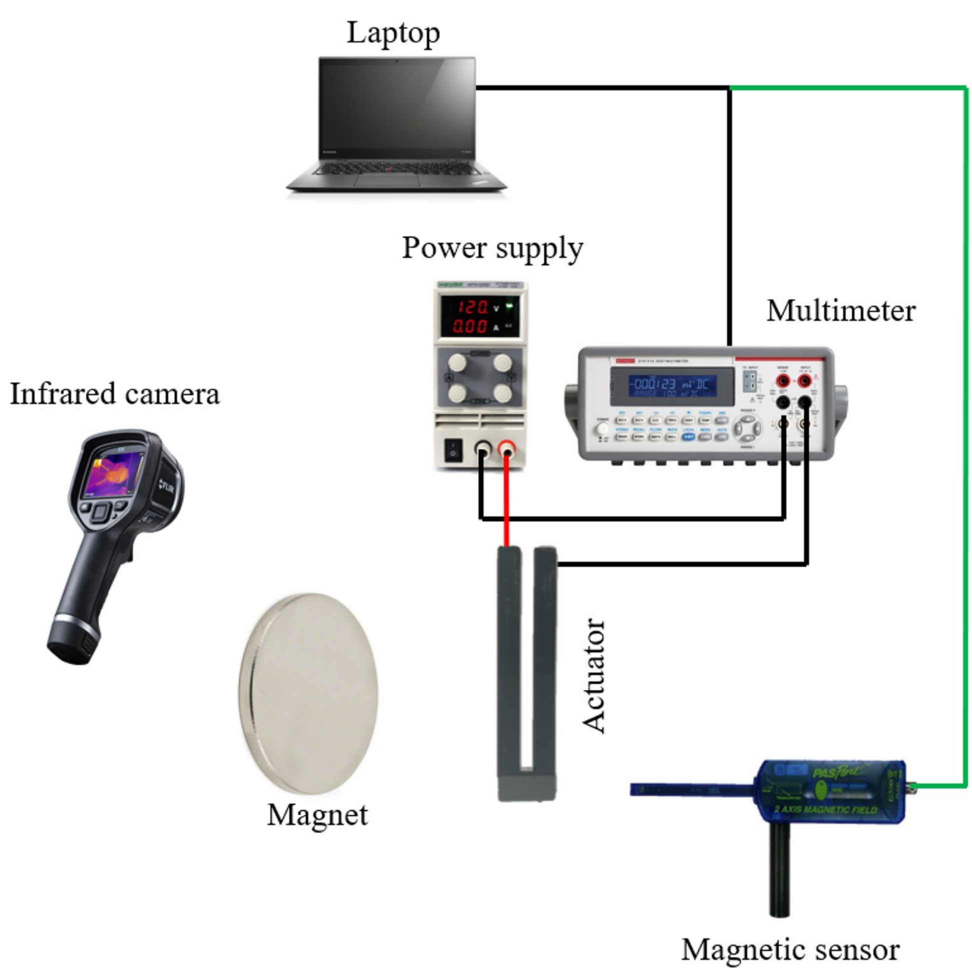

Fig. 3 A diagram of the data recording system and activating the actuator

magnet is measured. When the composite actuators are exposed to an external magnetic field and Joule heating activation, the experiment is carried out, see Fig. 3. A Pasco magnetic field sensor (PASCO, UK) with a probe with a precision of 0.01 G and a frequency of 10 Hz is utilised to measure the magnetic field. Pasco Capstone (PASCO, UK) is used to measure the magnetic strength of the permanent magnet. Pasco software records the motion trajectories of soft actuators to determine the deflection.

2.5 Dynamic mechanical analysis of materials

At room temperature, most thermoplastics like acrylonitrile butadiene styrene (ABS), polycarbonate (PC), polyethylene terephthalate glycol (PETG), polyetheretherketone (PEEK), and PLA are hard and brittle, but they become soft above glass transition temperature states. Storage modulus and T_g are the two most critical thermodynamic parameters that contribute to the SME of SMPCs and should be examined to determine the electro-induced shape memory behaviour of 4D-printed pure PLA, CPLA, and MPLA. PerkinElmer's dynamic mechanical analyser (DMA) 8000 (PerkinElmer, USA) is used to measure the storage modulus of CPLA, PLA and MPLA. The DMA test samples are manufactured at a speed of 70 mm/s, with model dimensions of 40 mm in length, 10 mm in width, and 1 mm in thickness. The selected test frequency is 1 Hz, which corresponds to the gradual change in features caused by temperature. The DMA test sequence increases from 30 to 100 °C at a rate of 2 °C/min.

3 Results and discussion

3.1 Material properties

Figure 4a displays the DMA storage modulus and tan δ values for CPLA, MPLA, and PLA. The addition of carbon black and iron particles increases the T_g of the composites. Therefore, the T_g of CPLA and MPLA is near to 75 °C and 65 °C, respectively. The activation temperature must be beyond the T_g to stimulate the structure. It happens because carbon and iron act as heterogeneous nucleation sites to raise the crystallisation temperature of the PLA matrix.

Carbon and iron greatly increase the stiffness of the PLA matrix, resulting in CPLA and MPLA storage moduli that are higher than pure PLA as shown in Fig. 4a. The chain flow causes the materials to lose stiffness and rapidly reduces their storage modulus, transforming them into a rubbery, viscous liquid. In contrast, the loss modulus of the material increases as molecular friction between its polymer chains grows. When it is close to the T_g phase, the molecular activities decrease and less energy is wasted, causing the loss modulus to decrease.

SEM is used to study the microstructure of CPLA and MPLA and demonstrate the distribution of carbon and iron particles. Also, the boundary condition between these two 3D-printed materials is investigated accordingly. SEM images of the 3D-printed CPLA and MPLA structure show that carbon and iron particles are uniformly spread throughout the PLA matrix, which is critical for the conductivity and magnetic properties, see Fig. 4b and c.

EDS test is employed to investigate boundary conditions and the interface of materials. As shown in Fig. 4d, the iron particles are visible in the 3D-printed MPLA structure, and there are no iron particles from the border to the CPLA sections. Few iron particles are visible in the border section due to the layer interaction. However, no iron particles are found in the CPLA structure and the small blue dots in the CPLA structure are noise. The tests reveal that the iron and carbon black particles are distributed throughout the structure which is very important in activating 4D-printed bi-stable actuators.

3.2 Magneto-electroactive composite structures

The initial research looks at the heating capabilities of 4D-printed composite structure traces as well as the voltage required for fast activation and heating. The needed voltage to trigger the structures is determined using this measurement. As illustrated in Fig. 5a, three structures are linked and heated using voltages of 60 V, 90 V, and 120 V. An infrared camera monitors temperature changes while the building is heated and cooled. Figure 5b, c, and d show the maximum temperature rise at various voltages. Using 120 V, the structure is heated to reach a stable temperature of 104 °C throughout the sample in 26 s. However, 100 °C T_g is not reached when 60 V is used. The data reveal that increasing the temperature leads to a decrease in current over time.

Resistance variations as a function of temperature are recorded using 120 V as illustrated in Fig. 5e. Data demonstrates that rising temperature and voltage result in greater resistance. Also, Fig. 5f shows the required time to achieve a stable temperature using different input voltages. The CPLA structures attain a temperature plateau once the voltage is applied, and an equilibrium is created between the energy supplied by Joule heating and the energy lost to the environment. The specimen is heated more quickly, resulting in a lower Young's modulus by 120 V. It should be noted the required voltage depends on the structure's design. Therefore, stimulating a 4D-printed structure with lower input voltage is discussed further.

Factors influencing printed magneto-electroactive structures are more complicated than those influencing electroactive structures. Because of its 1 mm thickness, the structure has five identical single layers. In theory, an overall energization may be obtained by connecting electrodes at

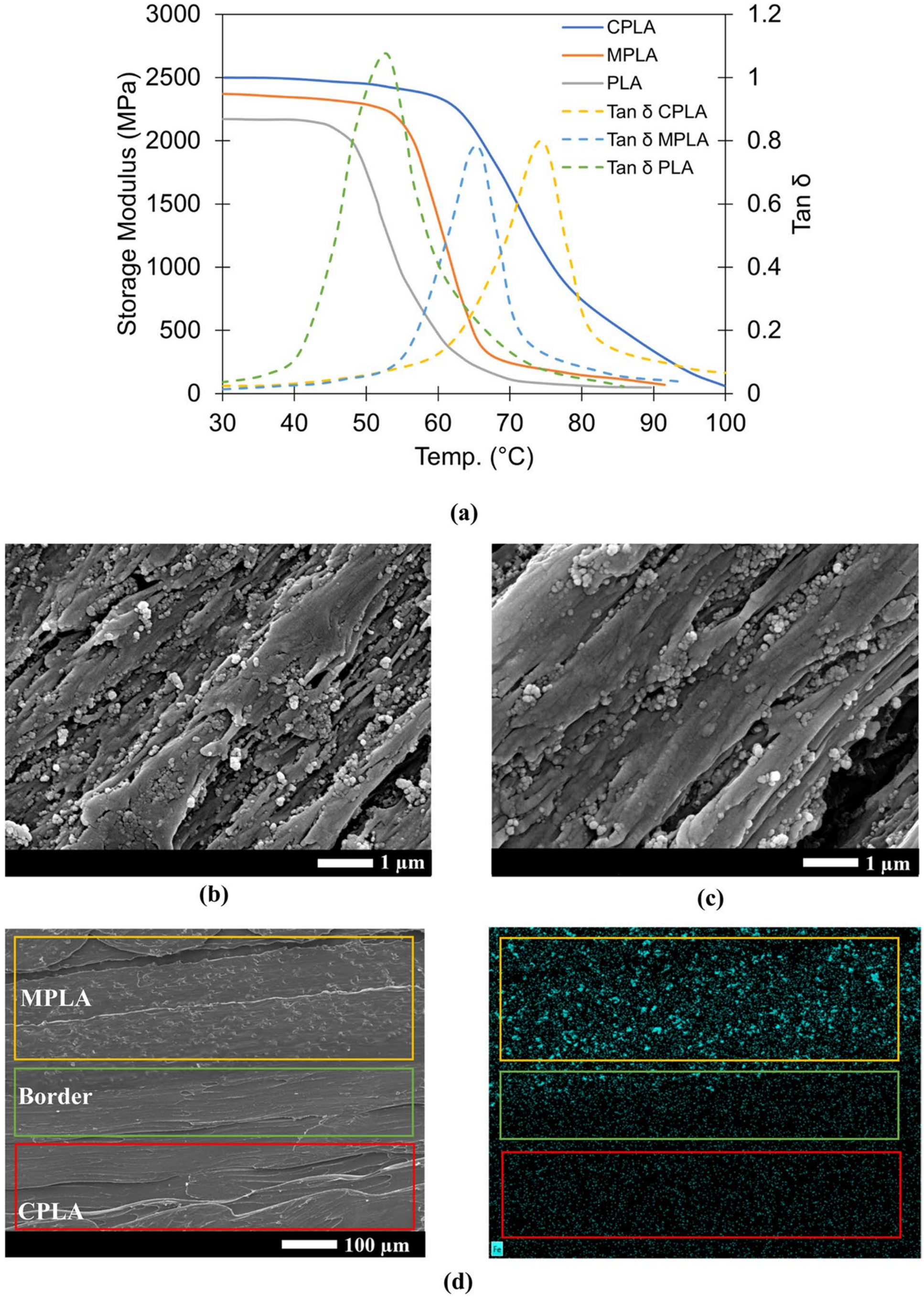

Fig. 4 **a** DMA measurements of materials. SEM image of 3D-printed **b** CPLA and **c** MPLA materials. **d** SEM and EDS images of the boundary condition and interface of 3D-printed CPLA and MPLA for iron particles. No iron particles can be seen in the CPLA area. (Yellow box is MPLA, the green box is the border of MPLA and CPLA, and the red box is CPLA)

the margins of the two long legs of the U-shaped structure. However, the current largely goes through the interior of the structure with the shortest distance, and the temperature rises in other portions mostly due to heat diffusion from this location. As a result, the heat conductivity of the construction along the direction perpendicular to the two long legs is lower than predicted. Meanwhile, because of the structure's narrow breadth, this problem is resolved.

Because of the naturally occurring concentration of iron particles in MPLA material, the actuator's end is activated by a low magnetic field. Actuation of the 4D-printed composite actuator is already attainable with an external magnetic field. The magnetic field creates a substantial force. Thus, clamping of one side of the actuator is required as seen in Fig. 6a. The technique for activating the actuator begins with heating it up. The current flows through the actuator and rapidly heats the 4D-printed actuator. As seen in Fig. 6b, the storage modulus of the structure drops, and the magnet attracts the actuator. The actuator bent to align with the magnetic field. The power is switched off,

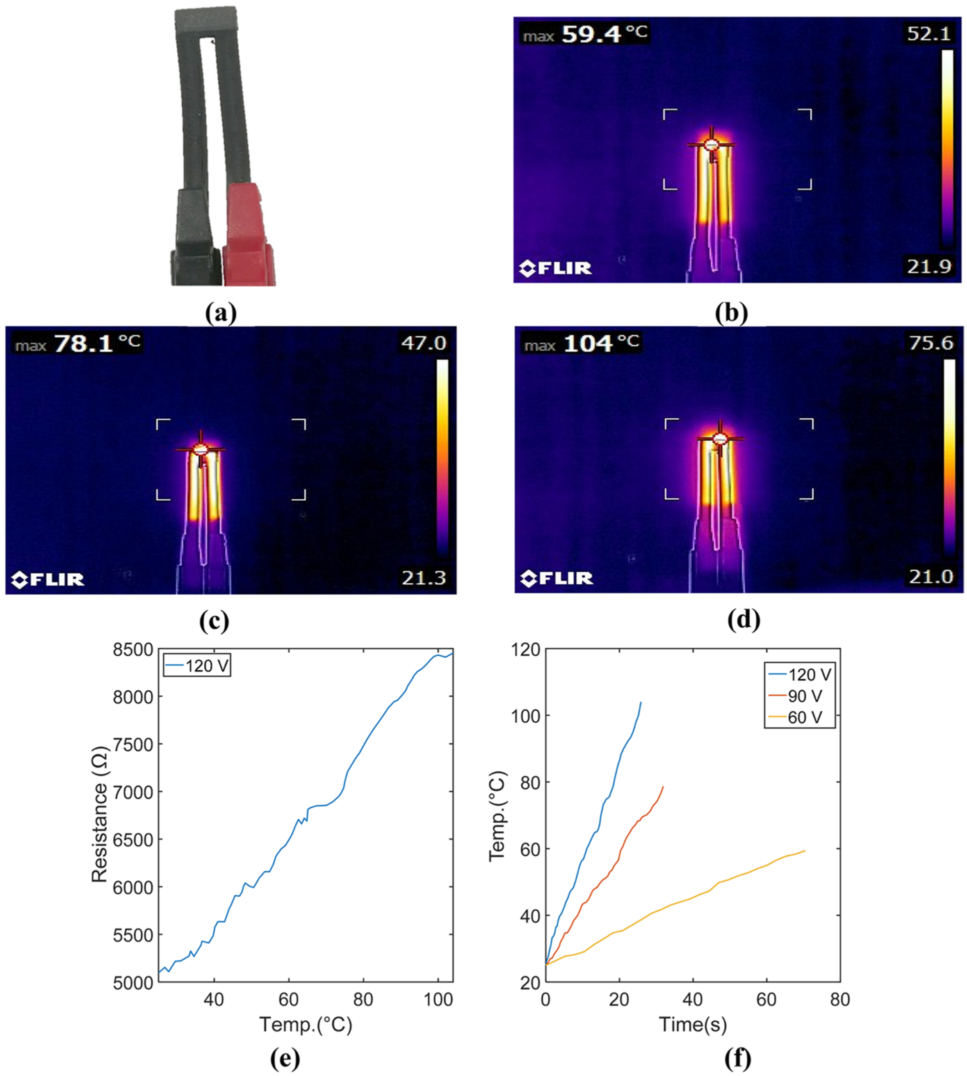

Fig. 5 **A** The crocodile clippers clamp the 4D-printed structure. Maximum heat distribution of 4D-printed actuator using **b** 60 V, **c** 90 V, and **d** 120 V. **e** The resistance varies as the temperature rises. **f** Stable temperature versus time using various input voltages

and the actuator is allowed to cool to room temperature. The actuator stays bent, and the magnetic field is removed correspondingly (see Fig. 6c). This is because of the SMPC structure's strength and rigidity. The adaptive structure becomes stable in a bending condition without any external stimuli. Joule heating returns the composite actuator to its original shape. The heated bi-stable 4D-printed structure restores to its original shape, as seen in Fig. 6d. The shape recovery is found to be around 100% of the original shape.

The composite actuator's reaction to an external magnetic field reflects the magnetic and mechanical qualities measured. In the presence of a magnetic field, MPLA exhibits exceptional magneto-responsiveness because of its high compliance and magnetism. The magnetic field strength is also measured during the procedure. The axial strength of the magnetic field grows as the distance between the magnets decreases. The attraction axial strength versus bending angle is shown in Fig. 6e. Also, Fig. 6f shows the changes in magnetic strength at different distances over time. The results imply that the bi-stable composite actuator can be programmed and controlled remotely. This finding suggests that the 4D-printed composite structure is particularly effective in improving bi-stability and shape programming. Furthermore, depending on the needs and magnetic field intensity, this concept may be used in a variety of shapes and forms.

3.3 Shape morphing and sustainability

Integrating magneto-electroactive SMPCs can achieve multi-stable structures and many shapes and forms into a single structure. When heated above T_g, these multi-stable structures are triggered to switch between numerous shape configurations by a permanent magnet. They become rigid and stable when cooled below T_g. This activation is repeated using Joule heating and a magnetic field. Quick, reversible shape changes, and complicated geometries with regionally customised deflections can be achieved by combining

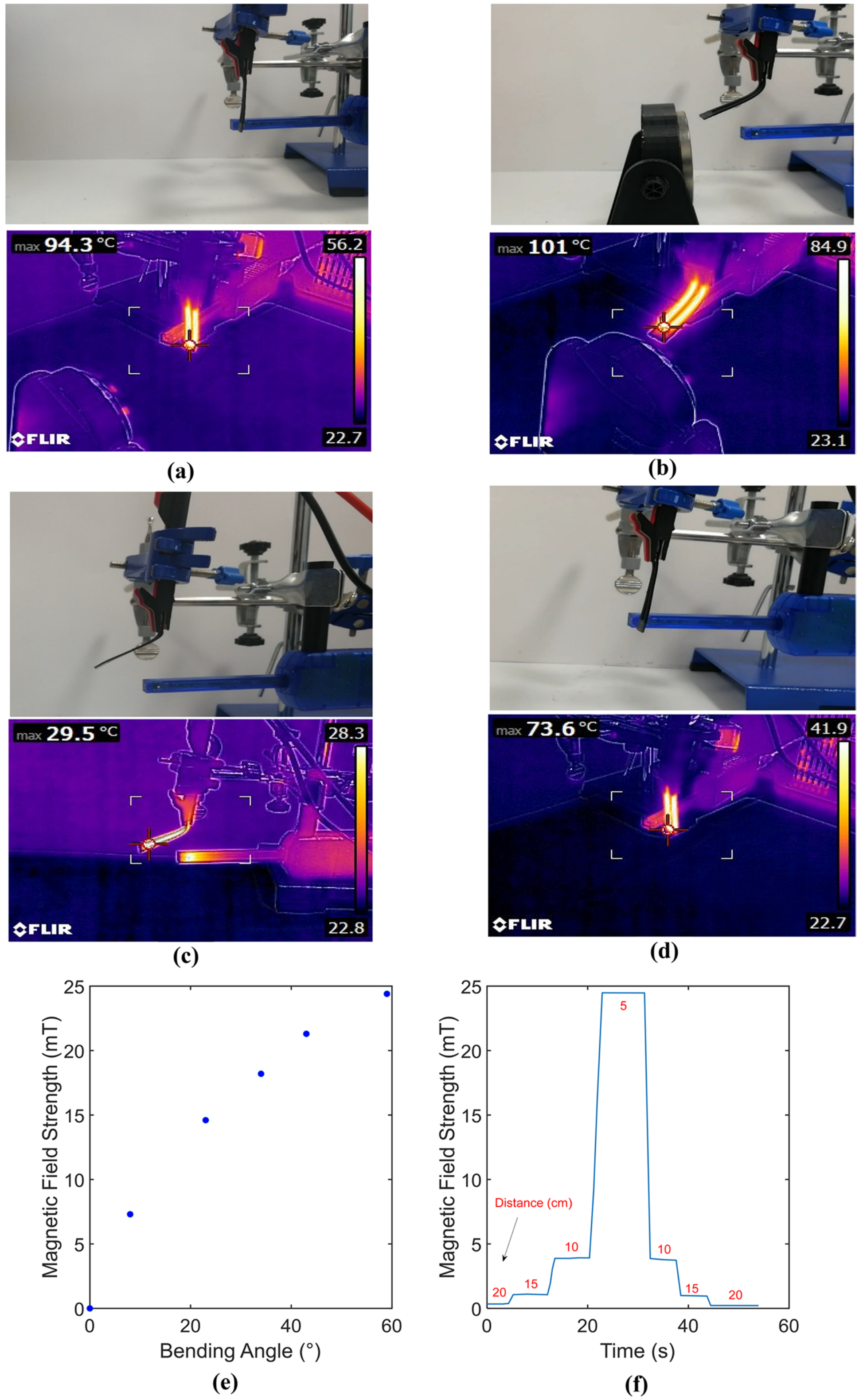

Fig. 6 **A** Heating the 4D-printed actuator in the vertical position. **b** Magnetic attraction to achieve maximum temperature and bending angle. **c** Cooling the composite actuator and removing the permanent magnet. **d** Joule heating returns the composite actuator to its original form after 26 s with 100% shape recovery. **e** Deflection of the actuators at various magnetic field strengths. **f** Magnetic strength versus time at different distances

4D-printed MPLA and CPLA. It should be noted that various designs of simple sheets can be developed and the sizes and dimensions of structures can be varied.

Structures with the same voltage at both ends are designed to have the same resistance and thermal conductivity to guarantee that the entire structure has the same heating efficiency. Thus, each area linked to the power source should have the same length. Various heating efficiencies are achieved due to the layer-by-layer forming properties in FDM. The previous design is determined to show the shape morphing of the structure in the presence of the magnetic field. The temperature set for programming is 80 °C.

The stable shapes of the structure are depicted in Fig. 7a. The structure is frozen in arbitrary shapes, such as folded and twisted shapes using a permanent magnet. As soon as the electrical current passes the structure, it goes back to its initial shape. This is more efficient than traditional robotic devices like pneumatic grippers, which require continual external input to retain a grasp on an object. Also, self-coiling and self-folding can be achieved easily. Another benefit of this technique is that multiple shapes are driven in a single sample.

A flat 2D beam structure is created to investigate the SME of magneto-electroactive SMPCs, as illustrated in Fig. 7b. Two sides of the beam are clamped and connected to the DC power supply. The beam is heated up to 80 °C using 60 V. Lower voltages can activate the structure due to better heat diffusion. A permanent magnet is used on top of the beam to attract the centre of the beam. The beam is snapped to its second shape using a permanent magnet to create a magnetic field that quickly drags the magnetic part up. The structure is heated and programmed until the final shape is achieved. Then, the structure is cooled down and remains in this stable shape as long as required.

Moreover, a flat 2D rectangular structure is designed accordingly (see Fig. 7c). Four sides of the structure are connected to the power supply with 120 V to have equal

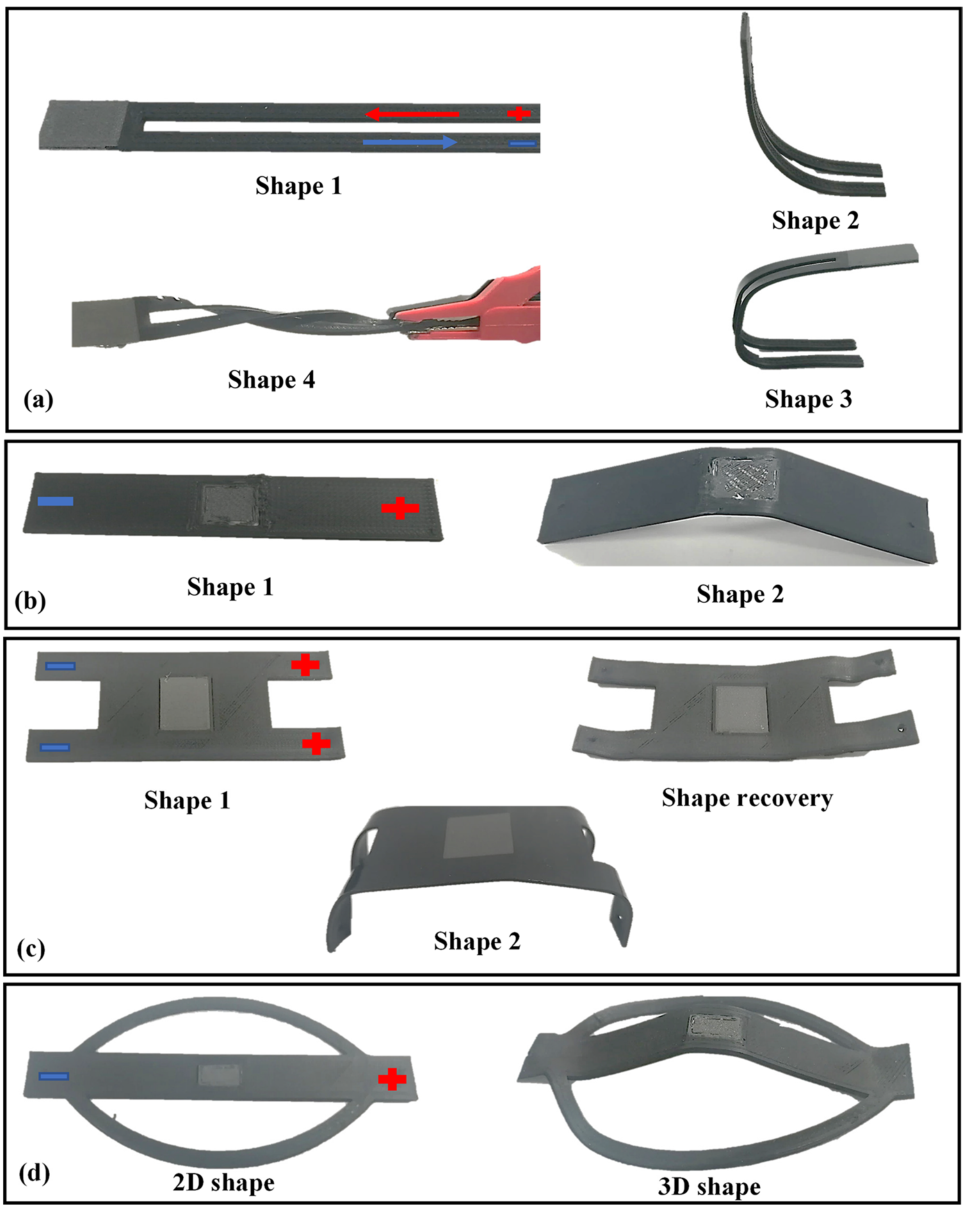

Fig. 7 **A** Different shapes of a 2D U-shape with CPLA material (black) MPLA (gray). **b** Transferring 1D beam shape to 2D shape using a permanent magnet and 60 V power supply. **c** Converting a 2D rectangular shape into a 3D structure and 93% shape recovery using Joule heating after magnetic remote programming. **d** Programming a 2D pyramid into a 3D structure

heat distribution. Two sides of the structure are clamped, and a permanent magnet is used to trigger the middle of the structure. The sample's shape fixity is around 100%. However, the shape recovery percentage is lower due to the high force from the magnetic field. Also, the capability of this method is converting 2D to 3D shapes.

A 2D pyramid shape is designed and printed. The structure is stimulated using the same technique. A final 3D shape is achieved as shown in Fig. 7d. The structure is cooled, drastically increasing stiffness and remains in the achieved shape. As a result, it is locked without the requirement for continual external energy or actuation. The structure goes back to its initial form when it is reheated. The multi-stability in conjunction with the locking offered by the switchable temporary and permanent shapes allows for low-energy, remotely operated actuators.

The main benefit of this technique is reducing material waste and energy in the 3D printing process and achieving different shapes in a single sample. Printing 3D twisted or folded structures needs too much effort/energy/time and material support. Hence, using this method can reduce material waste and energy/effort, and complex 3D shapes can be achieved easily. We develop the structures with PLA in Fig. 8 using the 3D printing procedure to show the benefit of our technique in green and sustainable design. A comparison between 4D-printed adaptive structures and 3D-printed structures is conducted. The first benefit of this technique is reducing material consumption and energy. 3D printing of different shapes consumes more materials during the procedure.

Figure 8 shows the support structures used in a few examples of 3D-printed samples. The weight of printed samples is more due to the support usage. It would be possible to reduce support structures in these shapes by changing the angle of printing. However, the surface quality and staircase defect become more in other orientations. Also, there is a need for support structures in other orientations for complex structures in the FDM process. Meanwhile, the structures can be printed in a flat orientation with high quality and good layer binding using 4DP and shape morphing techniques. This decreases the printing time and energy consumption accordingly. Hence, excessive material is used to build the structures. Also, material wastage would be high if the structures become large and massive. On the other hand, the surface quality of some areas in 3D printing becomes poor. The complex areas and shapes in which the printer does not use support show poor surface quality and integrity. As an example, the machine does not use support to build the rectangular shape in Fig. 8. The legs of the structure have poor surface quality. Meanwhile, these issues can be minimised or eliminated using the 4D printing of magneto-electroactive materials. The structures can be printed in a sheet shape and the final shape can be achieved using this technique without sacrificing material and surface quality.

The benefits of this concept point to tremendous implementation possibilities for engineering applications. The concepts can be used for a smart adaptable structure. This

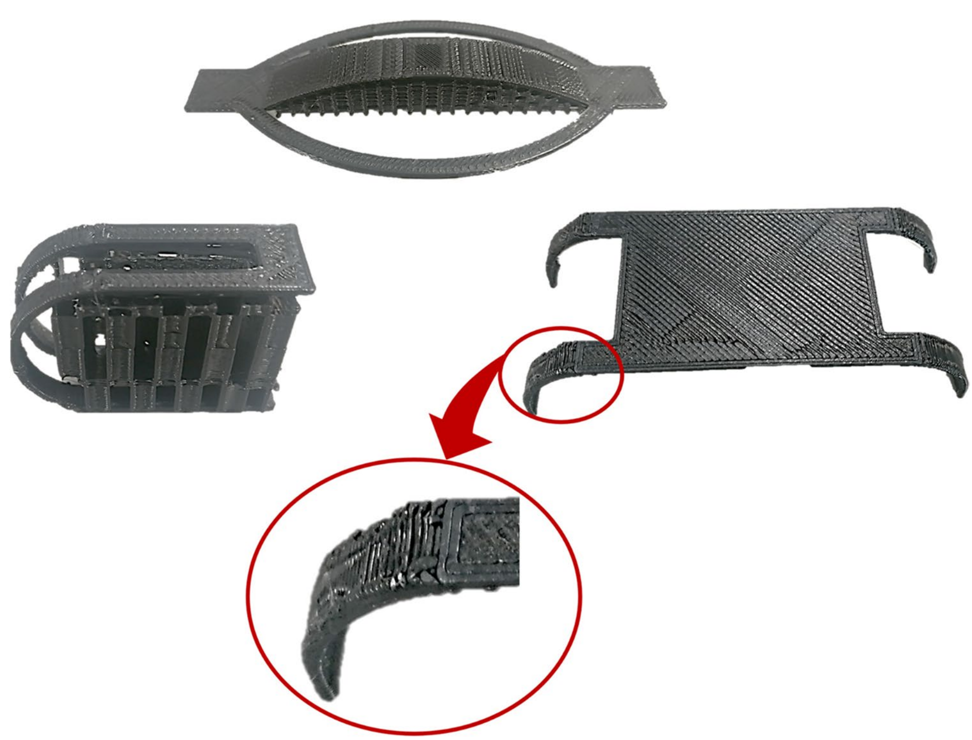

Fig. 8 3D printing of permanent shape structures with support

method can be used as a robotic gripper that can grasp an object without constant external input or sensing mechanisms. Additionally, this technique creates new opportunities for customised packaging and smart building applications. This technique is used in the packaging industry due to the reduction in storage space. The structure can be printed in a flat 2D shape and activated afterwards to achieve a customised shape. Since the structure can be locked in the required shape, this method can be used as a hook-shaped locker after activating without external stimuli as well.

In the design process of 4D-printed magneto-electroactive structures, in addition to enhancing printing accuracy, it is vital to verify that branches involved in deformation have the same internal printing structure. The combination of magnetic material with electroactive material allows for quick, reversible, and remotely induced form changing of 4D-printed thermoplastic objects. Printing CPLA and MPLA to illustrate the capability of remote actuation with magnetic fields has been discussed. A concept for smart and adaptable structures with multi-stable shapes at low cost has been developed. The proposed method increases efficiency and reduces material wastage. Green design can be implemented using this technique to eliminate 3D printing limitations. Finally, the proposed method can be used in different applications based on requirements.

4 Conclusion

The current work proposed a low-cost straightforward approach for creating magneto-electroactive shape memory structures utilising FDM 4D printing. As the fundamental core structure, 4D-printed CPLA and MPLA materials were investigated. The 4D-printed structure's joule heating and magnetic response were evaluated accordingly. Various input voltages were used to determine the structure's electrical, thermal, and electroactive shape memory properties. Remote control and programming of the composite actuator were also established. The shape recovery technique was provided for a given temperature and magnetic field. A small magnetic field was used to modulate the actuator. The unique composite actuator was investigated in terms of microstructure, composite interface, shape recovery, and dual stimuli.

The 4D-printed composite actuator was determined to be fast enough to revert to its previous shape when powered by a 120 V power supply. The composite actuator achieved a maximum bending angle of 59° for attraction with low external magnetic fields. The capabilities of the actuator in terms of shape morphing and being bi-stable were investigated. Shape morphing was studied to show the benefits of using this method to achieve various 2D/3D shapes from a single 1D/2D structure. It was demonstrated experimentally that 4DP composite actuators have tremendous promise in shape morphing and sustainability by reducing material waste and energy/effort. This study is anticipated to enhance the state-of-the-art 4DP and unleash the potential in green design and development of controlled functional structures with multi-stable and shape recovery properties.

Author contribution The authors contributed to the conceptual design, methodology, investigation, formal analysis and validation. Material preparation, data collection, and analysis were performed by Mohammadreza Lalegani Dezaki. The first draft of the manuscript was written by Mohammadreza Lalegani Dezaki, and Mahdi Bodaghi revised and commented on all versions of the manuscript. The authors read and approved the final manuscript.

Declarations

Competing interests The authors declare no competing interests.

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