4D printing of magnetoresponsive soft gripper and phenomenological approach for required magnetical actuation field

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Zitiervorschlag im APA Stil:

Kehret, D., Junk, S., Einloth, H., & Rapp, B. (2024). 4D printing of magnetoresponsive soft gripper and phenomenological approach for required magnetical actuation field. *The International Journal of Advanced Manufacturing Technology*, *135*(5-6), 2037–2049. https://doi.org/10.1007/s00170-024-14605-5

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The International Journal of Advanced Manufacturing Technology (2024) 135:2037–2049
https://doi.org/10.1007/s00170-024-14605-5

ORIGINAL ARTICLE

4D printing of magnetoresponsive soft gripper and phenomenological approach for required magnetical actuation field

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Received: 28 February 2024 / Accepted: 1 October 2024 / Published online: 15 October 2024

Abstract

4D printing is the next step in additive manufacturing. Magnetoresponsive materials facilitate the creation of gripping tools through 4D printing, allowing for structural changes in response to external stimuli. In this study, the structural change is manifested as motion, triggered by an external magnetic field. This technology offers significant advantages in medical and industrial applications, including the printing of life-like moving organ models for medical training and the development of actuators for use in explosive environments. Magnetoresponsive materials are programmed with a magnetic profile and actuated by an external magnetic field. A compound of strontium ferrite microparticles $SrFe_{12}O_{19}(\leq 20\mu m)$ and an elastic polymer (thermoplastic copolyester) with a Hardness of Shore D 40 was produced. A star-shaped body was programmed and actuated by two permanent magnets, each of $B_r = 1.29 - 1.32T$. As there is no analytical approach for calculating the required actuation flux density, one has been developed. The approach is verified experimentally by using a Hall probe. It is appropriate to set the field with a Helmholtz coil, despite the utilization of two permanent magnets. The use of a commercial fused filament fabrication printer for the processing of magnetoresponsive materials has been realized here for the first time. The main contributions are the short time constant (around $t_a = 0.1s$) for actuation and the repeatability (around $n = 200$ actuation cycles) of the motion. The feasibility of multiple diverse reprogramming is a step forward in 4D printing. Hence, the post-print programming and the inhomogeneity of the field limit the ease of the presented method.

Keywords Additive manufacturing · Magnetoresponsive material · Soft robot · 4D printing

1 Introduction

The novel research field of 4D printing implements the fourth dimension, i.e., time, into 3D printing. Bodies produced in this way exhibit, at different points in time, different structures and geometries. An input signal in the form of an external stimulus stimulates the printed body and its response is the desired output signal [1, 2]. This type of actuation allows, for example, manipulating a gripping tool without a constructive connection to the end effector. This advantage can be used in a wide range of fields, i.e., biomedical and sustainable engineering. The new application enables drug delivery and remotely controlled, minimally invasive operations, such as stent placement or biopsies with untethered grippers without the insertion of cables, tubes, and other handling technology [3–6]. Life-like moving organ models can be printed for medical training. Grippers for explosive environments can be realized, because electric servomotors, which can cause sparks, can be dispensed with.

Certain 4D printing processes produce a body at rest, which is later taught the required target structure. Teaching the desired structural change is called programming. The triggering of the target structure is called actuation. Suitable materials for 4D printing fulfill the criteria of printability and intelligence. Intelligence describes the ability to seemingly remember states and to assume them on the basis of a signal, as well as to return to the initial state in the absence of the signal. Printability refers to the formability of the material in the additive manufacturing process. Materials meeting these criteria are classified as smart materials. Magnetoresponsive

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materials are suitable for 4D printing [7]. They can be programmed with magnetic fields and later actuated.

In some studies, particle-filled silicones are processed using the direct ink writing (DIW) process [8–10]. Highly elastic bodies can be produced here. However, this process only allows flat 2D geometries. Lithography-based processes such as stereolithography (SLA) and digital light processing (DLP) enable small-scale objects to be produced from magnetically responsive material [1, 8–10]. The liquid monomer plastic processed causes the added magnetizable particles to sediment or chain together and thus prevent uniform distribution [11]. The fused filament fabrication (FFF) process is used to process filled viscoelastic thermoplastics [12]. The achievable size and low stiffness of the bodies are advantageous.

Soft robots are flexible robots that usually represent a material continuum [13]. In contrast, traditional industrial robots are typically made of rigid metallic materials with hard surfaces and exposed joints, which are driven electromechanically, hydraulically, pneumatically, or by combustion engines. Soft robots, due to their characteristics, allow a new level of human–machine interaction where the risk of injury or crushing is minimized. The field of collaborations between human and machines is called human-robot collaboration, and its importance is constantly increasing [14].

One form of actuation of soft robots is geometry change, i.e., motion and locomotion of the 4D printed part. Actuators made of smart materials can be actuated in different ways depending on the material and method. Actuation is possible by means of electro-, magneto-, temperature-, photoresponsivity, pressure sensitivity, and chemical responsivity [15]. Magnetoresponsive materials react to magnetic fields of different strengths. These responses are volume changes, deformations, or changes in position due to magnetic forces and torques. The actuators show a magnetization profile in which the magnetization direction and amplitude can vary.

In this way, soft grippers can be produced. These are soft robots that can perform gripping functions. The bionics of gripping by humans and animals is often imitated here. The majority of grippers available on the market are mostly two-finger grippers made of solid material, which do not meet the characteristic of soft robots. This challenge can be overcome with the use of smart materials [14].

Special challenges for the operational characteristics of soft grippers are speed, repeatability, control, and multiple diverse programming. The studies of Junk et al. on the application of smart materials in 4D printing observed long time constants and a lack of repeatability [16]. Magnetoresponsive materials show a fast targeted response when the external stimulus occurs and enable repeatability of the structural change as often as desired [12].

Kim et al. and Kricke et al. investigated a 4D printing process utilizing a magnetoresponsive polymer. In their processes, the body can be programmed on a single occasion during printing via a complex procedure [10, 11]. In the process of Kricke et al., the body can be stimulated irreversibly for a single time [11]. The new approach in this paper addresses the aforementioned two disadvantages by enabling reversibility and multiple diverse reprogramming. Furthermore, the technology employed is relatively straightforward, thus allowing the use of a conventional printer.

The system developed by Vito Cacucciolo et al. for categorizing soft grippers includes the three methods of gripping by actuation, gripping by controlled stiffness, and gripping by controlled adhesion. Gripping by actuation is gripping with a passive or active structure actuated by an external or integral actuator. Gripping by controlled stiffness is the utilization of tunable stiffness. Here, materials can vary their shape or stiffness due to external influences. Gripping by controlled adhesion is gripping by surface forces between gripper and object [17]. Given that the material under consideration cannot be unambiguously categorized within these established frameworks, it is necessary to expand the definition accordingly.

Magnetoresponsive soft grippers can be actuated particularly quickly, since the duration of the field buildup is the only influencing factor. An elastomer in the form of a rod with embedded permanent magnets can be stimulated to creep in a controlled manner in the field of a Helmholtz coil [18]. Ferromagnetic materials have the property that in an externally imposed magnetic field, they also receive a magnetic flux density inside, which remains to a certain extent after removal of the external field. The magnetic flux density that can be maximally preserved is called remanence B_R. The magnetic field strength that must be overcome to realign an already differently oriented ferromagnetic material is called coercivity H_C. Magnetoresponsive materials belong to the smart materials and possess the properties of deformability and magnetizability [8]. For this reason, a compound composed of the viscoelastic thermoplastic copolyester elastomer (TPC) and the intercalated ferromagnetic particles of strontium ferrite is used in this work. This compound material meets the requirements for 4D printing and can be produced and processed as granules as well as filaments [12]. Compounds produced in this way are called magnetoactive soft materials (MASM) [9]. The strontium ferrite exhibits soft magnetic behavior, which means relatively low coercivity H_C and relatively high remanence B_R. By introducing a magnetization profile into the printed body, its response to an external magnetic field can be determined. First, the body is produced in its rest position using the 3D printing process. The embedded particles are orientation-free at this point. The body is transformed into the target geometry using a die or a negative mold. If an external, parallel, magnetic field is now induced which exceeds the coercivity H_C of the strontium ferrite, the particles are polarized. From this point on, the soft gripper is programmed and the external magnetic field can be removed. The state of the orientation of the magnetic dipole moment before and after programming is shown in Fig. 2. The body can now be brought into its rest position and has an actuatable structure as intelligence. For actuation, the soft gripper is placed in a magnetic field below the coercivity H_C without any die or negative mold. In doing so, it assumes its target geometry and remains in this as long as the external field remains. If the field is removed, the body returns to its rest position. This process is reversible, and the programmed structure is maintained for a long time if the body is not exposed to strong external fields. The question is how to generate the external field to actuate the soft gripper. There are two possibilities for this. A magnetic field exists permanently in the surrounding area of a permanent magnet or can be generated with current-carrying coils. The use of permanent magnets is suitable because of the availability of highly remanent magnets. The effective field strength impacting a soft robot can be controlled by its distance towards the permanent magnet. The problem is that these fields are not very homogeneous due to the field lines running in closed paths. Better suited for this purpose are coil fields which generate parallel field lines along the coil axis. The Helmholtz coil design is particularly suitable, since a pair of coils is arranged in a row to create a penetrable and visible space in the center, which not only has homogeneous parallel field lines, but also a region of constant field strength H (see Fig. 11). The strength of fields generated in this way is technically limited. High field strengths H lead to high currents I which can cause a thermal overload. The advantages of the Helmholtz coil are the adjustability and the possibility of switching off.

This work deals with the design and fabrication of a magnetoresponsive soft gripper and a method for its programming and actuation as shown in Fig. 1. The subjects of repeatability, speed, multiple diverse reprogramming, and the use of a commonly available printer are of particular interest. Furthermore, a phenomenological approach for the analytical calculation of the necessary magnetic flux density is developed, which has to be generated by actuation using a Helmholtz coil. The analytical approach enables the

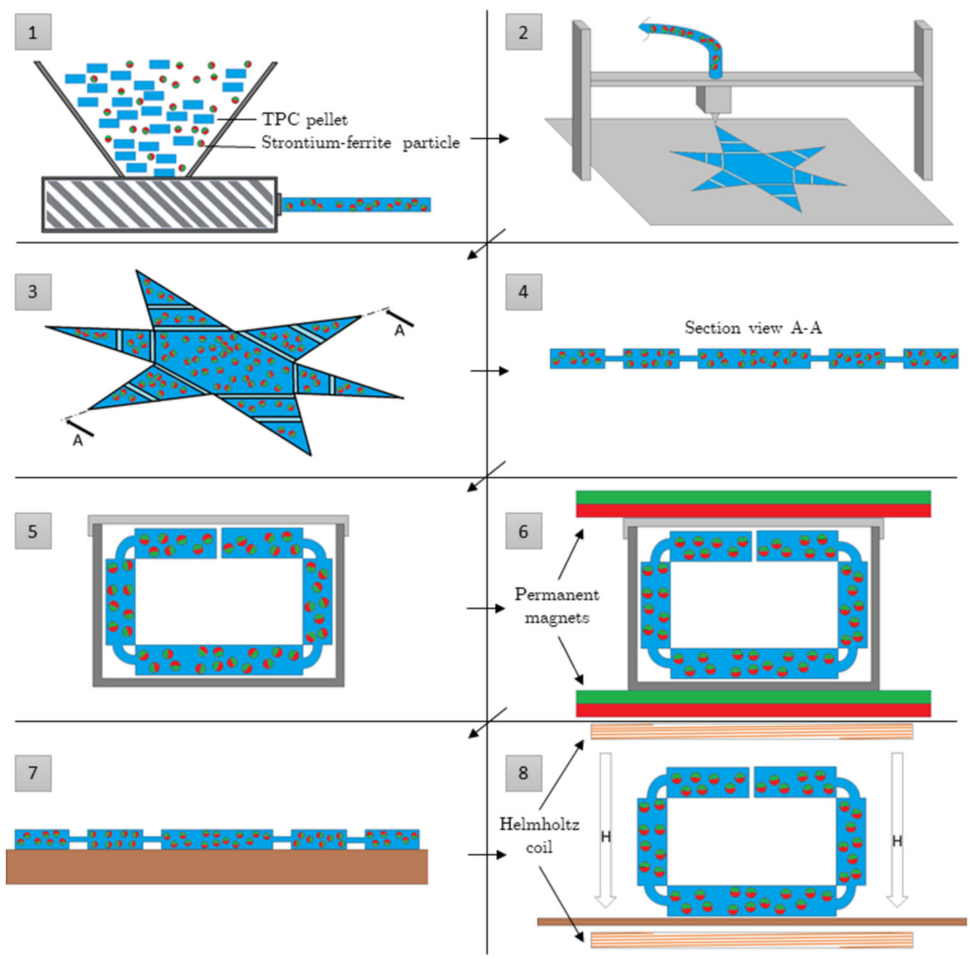


Fig. 1 **1** Compound extrusion of thermoplastic copolyester elastomer (TPC) pellets and strontium ferrite particles. **2** Printing by a fused deposition modeling printer (FDM). **3** Printed soft gripper (star gripper). **4** Section view of soft gripper with unorientated dipoles. **5** Die for forming actuation shape made out of polylactide (PLA). **6** Programming star gripper with two permanent disc magnets. **7** Star gripper at rest with orientated dipoles. **8** Actuated star gripper by magnetic field H generated by a Helmholtz coil

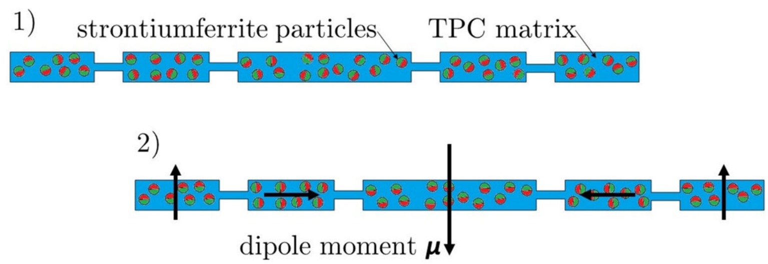


Fig. 2 **1** This section view accordingly to picture 4 in Fig. 1 shows a model of the compound of TPC and strontium ferrite particles with an anisotropic magnetical orientation before the programming. **2** Here, the oriented dipole moment μ after programming is shown. This stage of 4D printing refers to the picture 7 in Fig. 1

design of a Helmholtz coil configuration for this application. A measurement of the real actuation magnetic field will verify the approach. In addition, the impact of the magnetization process is analyzed (Fig. 2), and a revised system for categorizing soft grippers is proposed. Special attention is paid to the repeatability of the actuation and its time constant. All subjects close different knowledge gaps in 4D printing research.

2 Materials and methods

2.1 Components

Magnetoresponsive polymers must fulfill the properties of deformability and magnetizability. A compound material consisting of a polymer with viscoelastic properties and intercalated ferromagnetic particles is suitable for this purpose. Many works describe the use of polydimethylsiloxane (PDMS) and neodymium for the desired rheological properties [9, 10, 19]. However, in this work, the polymeric material thermoplastic copolyester elastomer (TPC) is used in conjunction with the ferromagnetic material strontium ferrite. These materials are readily available, easy to process, and inexpensive to purchase. The strontium ferrite exhibits soft magnetic behavior, which means relatively low coercivity Hc and relatively high remanence B_R. The polymer material used is the product FilaFlexible40 from the manufacturer Filatech. It has the necessary elasticity and can be printed in various processes. According to the data sheet, the melting temperature is $T = 160°C$ and the density at room temperature is $\rho_{Polymer} = 1.22\frac{g}{cm^3}$ [20]. The ferromagnetic material is the product MANIPERM magnetic powder from the manufacturer TRIDELTA Hartferrite. For production, iron oxide Fe_2O_3 and strontium carbonate $SrCO_3$ are mixed in a molar ratio of 5.7:1. Wetting with water produces granules under rotation. Calcination between $T_c = 1100°C$ and $T_c = 1320°C$ produces strontium hexaferrite $SrFe_{12}O_{19}$. A grain size of less than $d_g = 20\mu m$ is achieved by dry grinding in tubular vibration mills (see Fig. 3). The strontium ferrite powder has a remanence of $B_R = 168mT$, which manifests itself only by reaching the saturation flux density in the hysteresis (see Fig. 4). The polarization coercivity is $H_C = 170\frac{kA}{m}$. The density of the substance is $\rho_{solid} = 3.6\frac{g}{cm^3}$, although here, due to the powder form, there is a bulk density. It is $\rho_{bulk} = 1.15\frac{g}{cm^3}$ [21]. Further information is available in Tables 1 and 2.

2.2 Compound extrusion

The subsequent feedstock for 4D printing is in granular form. To obtain this, the particles and the polymer material are mixed and extruded in a Collin Teachline ZK 25T screw extruder which has two screws. In this process, based on the melting temperature of the polymer material, the first of five heating stages is set to a temperature of $T_1 = 190°C$ and $T_{2-5} = 220°C$ at the following ones. The mass ratio of 2:1 (strontium ferrite to TPC) is established. On an experimental basis, mass ratios of 3:1 and 7:3 are produced. Further studies

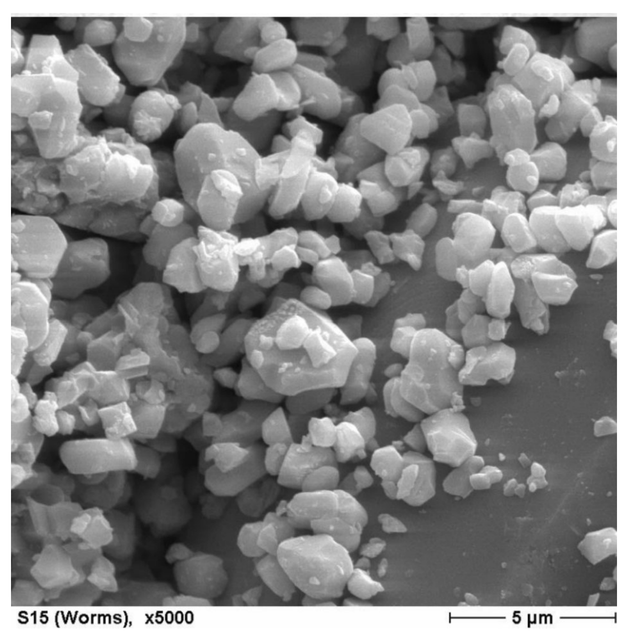


Fig. 3 Scanning electron microscopy (SEM) of strontium ferrite powder after calcination process. First iron oxide Fe_2O_3 and strontium carbonate $SrCO_3$ are mixed in a molar ratio of 5.7:1, granules are wetted, calcination takes place between $T_c = 1100°C$ and $T_c = 1320°C$, and dry grinding in tubular vibration mills is performed to produce strontium hexaferrite $SrFe_{12}O_{19}$ of a grain size of less than $d_g = 20\mu m$. With permission of Tridelta Hartferrite GmbH, Hermsdorf, Germany [21]

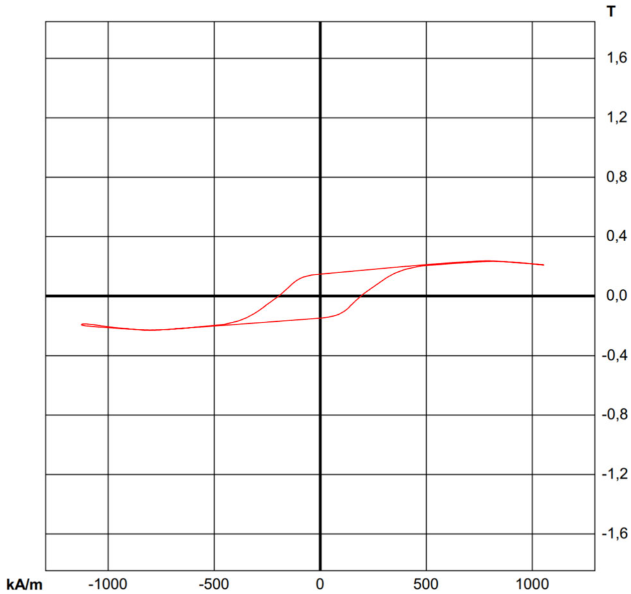


Fig. 4 Hysteresis of strontium ferrite powder. Measurement of isotropic pressed sample at a temperature of $T = 19.4°C$. Measured with a ECKEL Robograph 2. With permission of Tridelta Hartferrite GmbH, Hermsdorf, Germany [21]

can follow with this material. The investigations at the University of Offenburg showed that the 1:1 mixing ratio did not produce a satisfactory shape memory effect [12]. Varying the mass ratios is made possible by the use of two Coperion K-Tron feeders. They are mounted above the screw extruder and can gravimetrically add the two components. The feeder containing the particulates feeds $\dot{m}_{Particle} = 3.3\frac{kg}{h}$, while the one with the polymer material conveys $\dot{m}_{Polymer} = 1.65\frac{kg}{h}$. The layout is shown in Fig. 5. This process belongs to step 1 in Fig. 1.

2.3 Granulating the extrusion strand

The extruded strand is passed through a cooling tank where it solidifies. The strand is finally reduced to pellets in a Collin

Table 1 Strontium ferrite powder

Attribute	Unit	Value
Isotropic	–	Yes
Density	g/cm^3	3.6
Bulk density	g/cm^3	1.15
Specific surface	$cm^2/$	2500
Sieve analysis $> 71\mu m$	%	0
Sieve analysis $> 32\mu m$	%	15
Remanence B_R	mT	168
Flux density coercivity H_Cb	kA/m	102
Polarization coercivity H_Cj	kA/m	170
Magnetic energy product BH_{max}	kJ/m^3	4.3

Table 2 Thermoplastic copolyester elastomer

Attribute	Unit	Value
Density	g/cm^3	1.22
Hardness	D	40
Tensile strength	MPa	7.85
Elongation at break	%	> 400
Flexural strength	MPa	4.41
Heat deflection temperature HDT/B	$°C$	50
Melting temperature	$°C$	160

CSG171/1 pelletizer with an integrated milling wheel. The resulting granules are dried at a temperature of $T_{dry} = 40°C$ for 24 h in a drying oven.

2.4 Filament extrusion

For applications requiring a dimensionally stable filament, the granules are melted and extruded again in a further processing step, and then wound onto rolls. The extrusion strand from the first processing step is highly irregular in its diameter and surface finish. The diameter of the filament is specified by the 3D printer and must be $d_{filament} = 1.75 \pm 0.03mm$.

2.5 Printing method

The Josef Prusa company has been supplying reliable 3D printers that are widely used for many years [22]. With the choice to use the Prusa i3 MK3S+, a commercial printer is chosen which proofs the universal compatibility of the developed magnetoresponsive material. With respect to standards EN ISO/ASTM 52900 and DIN EN ISO 18064, the print job is referred to as MEX-TRB/Cp/TPC,$SrFe_{12}O_{19}$. In this process, polymeric plastic is processed. This is provided in wire form on spools and is referred to as filament. The geometries produced in the CAD program Catia V5 are transferred to the STL format to suit with the software Prusa Slicer. This data format is an optimization result of the geometry representation from CAD environments [23].

The printing of the soft gripper corresponds to the step 2 in Fig. 1. Here, the magnetoresponsive filament is deformed to the aimed shape. The printing parameters are a nozzle temperature of $T_{nozzle} = 245°C$ and a bed temperature of $T_{bed} = 60°C$. For more details, see Table 3.

2.6 Model-based analytical calculation of the required magnetic actuation field *H*

To achieve a certain torque τ on a magnetic plate, the required magnetic actuation field H has to be known. As a simplified model for the calculation of H, a square plate is assumed,

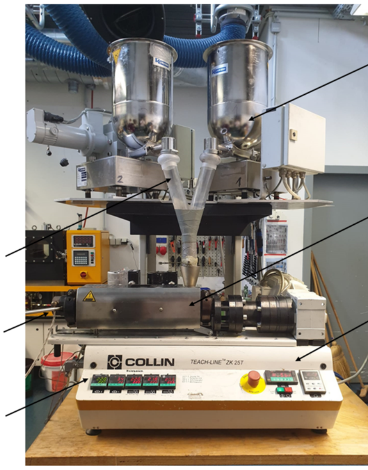


Fig. 5 Layout for fabrication of the compound of TPC and strontium ferrite. Two separate feeders containing the two components deliver gravimetrically the material into the screw extruder through a Y pipe. The mass ratio of strontium ferrite particles compared to TPC elastomer is 2:1. The screwextruder heats up the material by using five heaters in a row along the two screws. $T_1 = 190°C$ at the inlet and $T_{2-5} = 220°C$ at the following stages. The mixed compound leaves the extruder at the extrusion nozzle

which is programmed at an angle of $\varphi = 45°$ to the horizontal. The programming is done with the two neodymium disc magnets. The layout is displayed in Fig. 6. Now, the required magnetic flux density H of the actuating magnetic field, which causes the plate to be deflected from rest, needs to be determined. The torque M, which results from the gravitational force G and the lever arm h, must correspond to the magnetic torque τ in the equilibrium of forces at the erected plate.

$$M = Gh \tag{1}$$

$$M = mg\cos 45°\frac{l}{2} \tag{2}$$

The magnetic torque τ results from the magnetic dipole moment μ in interaction with an external magnetic field H, if the orientations to each other differ. To take this into account, the quantities are considered vectorially. The magnetic torque τ results from the cross product of the dipole moment μ with the external magnetic field H and is formally as follows:

$$\tau = \mu \times H \tag{3}$$

The dipole moment μ depends on the volume V, the permeability of the surrounding medium μ_0, and the impressed magnetic flux density B. Since V and μ_0 are constant and given, the only scalable parameter here is the inner flux density B.

$$\mu = \frac{V}{\mu_0}B \tag{4}$$

After exceeding the remanence B_R at programming, the inner flux density B is at most equal to the value of the remanence B_R. Inserted into the formula of the magnetic torque τ, it results trigonometrically expressed:

$$\tau = \frac{V}{\mu_0}BH\sin\theta \tag{5}$$

Here, θ corresponds to the angle between the vectors B and H. The remanence of the magnetoresponsive material is $B_R = 168mT$, the permeability of the ambient medium air is $\mu_0 = 1.26 * 10^-6T\frac{m}{A}$, and the angle between the vectors corresponds to $\theta = 45°$. With the absolute values of the model, the torque from gravity is $M = 0.325 * 10^-3Nm$. The formula (5) for τ can be converted to H to receive the aimed equation.

$$H = \tau\frac{\mu_0}{BV\sin\theta} \tag{6}$$

Table 3 Printing parameters

Parameter	Unit	Value
Layer thickness	mm	0.15
Nozzle temperature for first layer	$°C$	250
Nozzle temperature else	$°C$	245
Bed temperature for first layer	$°C$	70
Bed temperature else	$°C$	60
Printing speed	mm/s	20
Infill	$\%$	100
Nozzle diameter	mm	0.4

2.7 Programming and actuation

Two disc magnets made of neodymium are used to program the soft gripper as shown in step 6 in Fig. 1. They have a remanence $B_R = 1.29 - 1.32T$ and a coercivity of $H_C = 955\frac{kA}{m}$, which corresponds to the value $H_C = 1.2T$ in the unit Telsa. The soft gripper is placed in its actuation position in the programming box following step 5 in Fig. 1. The two disc magnets are placed under the box, respectively above the lid. The magnetic flux density $H = 349mT$ from the addition of both fields exceeds $H_C = 170\frac{kA}{m} = 214mT$ of the strontium ferrite, thus imprinting the magnetization profile in the

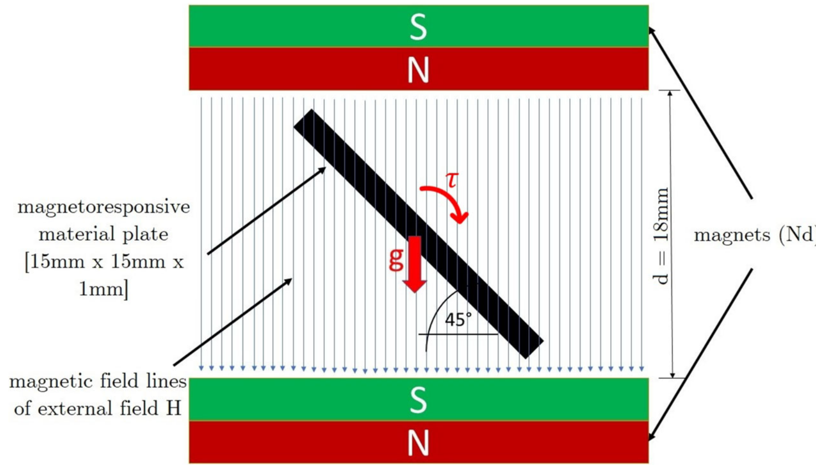


Fig. 6 Model for the calculation of the required external flux density H to generate the necessary magnetic moment τ. A magnetoresponsive plate is placed at an angle of $\varphi = 45°$ to horizontal. The plate is programmed by magnetizing the particles in the aimed orientation. Above and underneath the plate are permanent magnets made out of neodymium each with a remanence of $B_R = 1.29 - 1.32T$. The gravity g causes a torque M which must be repealed by the magnetic moment τ

material. The comparison between step 5 and step 6 in Fig. 1 visualizes the change of orientation of the dipoles inside the particles. To avoid causing further remagnetization, the magnets are moved away from the programmed body tangential to the field line direction. To do this, the magnet on the lid is first lifted. Then, the entire programming box is lifted off the lower magnet. As long as the effective fields of the two magnets stay fused, the field lines run parallel through the body to be programmed. Outside, the field lines run back on curved paths. This is due to the fact that only magnetic dipoles exist and field lines therefore always run on closed paths. Therefore, the undesirable effect occurs that the body to be programmed is briefly penetrated by inhomogeneous curved field lines while the magnets are removed. This can affect the intended magnetization profile. However, the magnetic characteristics of a permanent magnet always apply only directly on the surface of the magnet. They decrease rapidly with increasing distance normal to the surface. Thus, it is assumed that the magnetic flux density at the point where the body to be programmed is penetrated by curved field lines is below the coercivity H_C of the strontium ferrite. Thus, the intended magnetization profile should be preserved. In order to actuate the thus programmed body from rest, a parallel homogeneous external magnetic field H is again required, but its magnitude needs to be below the coercivity H_C of the strontium ferrite. The magnitude of the field strength H necessary to perform the desired motion can be calculated according to the analytical approach of this work.

2.8 Measurement of the magnetic actuation field

To verify the analytical approach, the specimen from the model calculation is printed and examined. The magnitude of the magnetic field H required for actuation is to be measured. The measuring tool to be used is a teslameter, which can measure the magnetic flux density by means of a tangential Hall probe. It is a device from the manufacturer Phywe.

3 Results and discussion

3.1 Shaping and printing of the soft gripper

An outstanding feature of magnetoresponsive soft grippers is their remote controllability. It opens the possibility to control movements in closed unstructured environments without a constructive haptic connection [12]. For example, this could be a minimally invasive surgery in a patient's body or an in vitro fertilization [3]. Among other things, this work answers the question of the sensible design of such a soft gripper. Bionically inspired, the soft gripper is based on the human hand. In order to reduce the complexity level, a point-symmetric shape with six gripper fingers was chosen. The geometry is shown in steps 3 and 4 in Fig. 1. The gripper fingers each have two joints and are tapered. They are connected by a hexagonal base. The pointed fingers can be used to grasp objects and remove tissue, as is necessary for biopsies. Other publications about magnetoresponsive actuators use flat designs of the bodies like sheets and foils [5, 24]. To obtain a foil-like body, the thickness was set to $d = 2mm$. The hinges have a thickness of only $s = 0.3mm$ to maintain deformability and lower the bending moment of resistance [12]. The span at rest is $l = 85mm$. This scaling was chosen for better visualization. The final gripper is shown in Fig. 7.

By choosing a flat primitive geometry, the full potential of additive manufacturing is not realized. Because of its shape, it is named star gripper. The use of a conventional filament printer for processing magnetoresponsive materials was implemented here for the first time. This enables research into magnetoresponsive 4D printing at a low cost

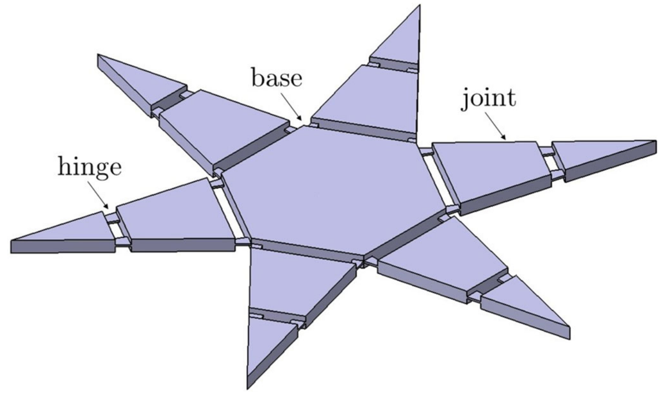


Fig. 7 The 3D model of the star gripper for the printing. The shape follows a foil-like body with six point-symmetric fingers and two hinges and joints each. Its thickness is $d = 2mm$, and the thickness of the hinges is $s = 0.3mm$

for the equipment. By dispensing with multi-material solutions, the printing speed is increased.

3.2 Influence of the magnetization procedure

A programmed soft gripper has a singular magnetic profile. In the actuation magnetic field, the dipoles aligned in the soft gripper strive to orient themselves parallel to the outer field lines H as shown in steps 7 and 8 in Fig. 1. In this theoretically idealized view, it is therefore only possible to achieve the programmed state at actuation at 100% of deflection. Stopping the movement at, for example, 50% of the deflection is not possible. Consequently, gripping an object with a different diameter requires new programming. In fact, in reality, gripping of larger objects is possible because parallelization of the field lines while actuation is not completely achieved due to a weaker magnetic flux. This finding is important because particularly fragile objects should not be deformed by the gripper, which could occur if the gripping force is too large. For programming the star gripper, a programming box was printed as a die from the material polylactide (PLA). This box is shown in Fig. 8. When inserted into the programming box, all joints of the star gripper are deformed by $\varphi = 90°$ and can thus be programmed by means of a magnetic flux density above the coercivity of the strontium ferrite $H_C = 213mT$.

Two methods can be considered for field generation: on the one hand, the use of permanent magnets; and on the other hand, the use of Helmholtz coils. Helmholtz coils are not very suitable for generating a field $H > H_C$, since they usually offer significantly lower magnetic flux densities and must carry very high electric currents to generate higher flux densities. This in turn leads to strong heating of the coils. For a magnetic flux density of $H = 320mT$, a pair of coils with $N = 200$ turns each and a current of $I = 80A$ is required. For this reason, programming using two permanent magnets made of neodymium with a remanence of $B_R = 1.29{-}1.32T$ was preferred. The two disc magnets were measured with respect to their magnetic flux densities by means of a teslameter. This showed that the magnitude H varies at different locations on the surface of the magnets. The inhomogeneity of the field of the permanent magnets leads to non-uniform programming. This disadvantage was tolerated because the advantage of the required magnetic flux density outweighed it. When lifting the permanent magnets after programming, care was taken to guide them away parallel to the field lines. Even if it comes to the fact that the common field of both magnets separates again and curved field lines arise again, it is without effect. In the intermediate space, the amount H with a relatively large distance to the magnet surface is below H_C.

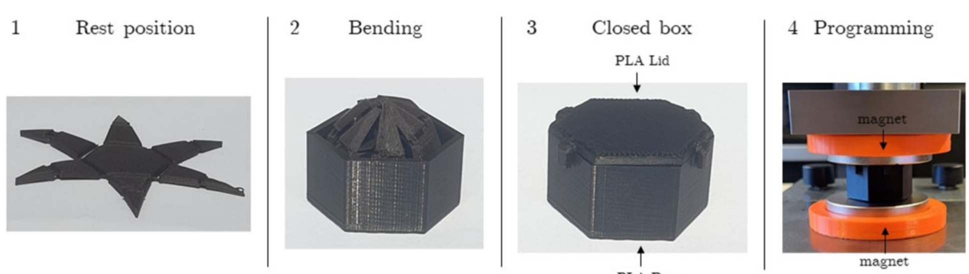


Fig. 8 For programming the star gripper (**1**), a programming box made out of PLA is printed (**2**). It keeps the gripper in its actuation position by bending all hinges by $\varphi = 90°$. A lid closes the box (**3**) and enables the application of two neodymium permanent magnets, each with a remanence of $B_R = 1.29 - 1.32T$ and a coercivity of $H_C = 1.2T$. The two magnets are applied above the lid and underneath the box (**4**)

Fig. 9 Without an external magnetic field H, the star gripper remains at rest. With the appearance of the external magnetic field H, the star gripper bends to actuation structure. Especially, picture **3** shows the uneven response caused by the inhomogeneity of the magnetization. **1** The external magnetic actuation field is $H = 10mT$, **2** $H = 22mT$, **3** $H = 31mT$, and **4** $H = 62mT$

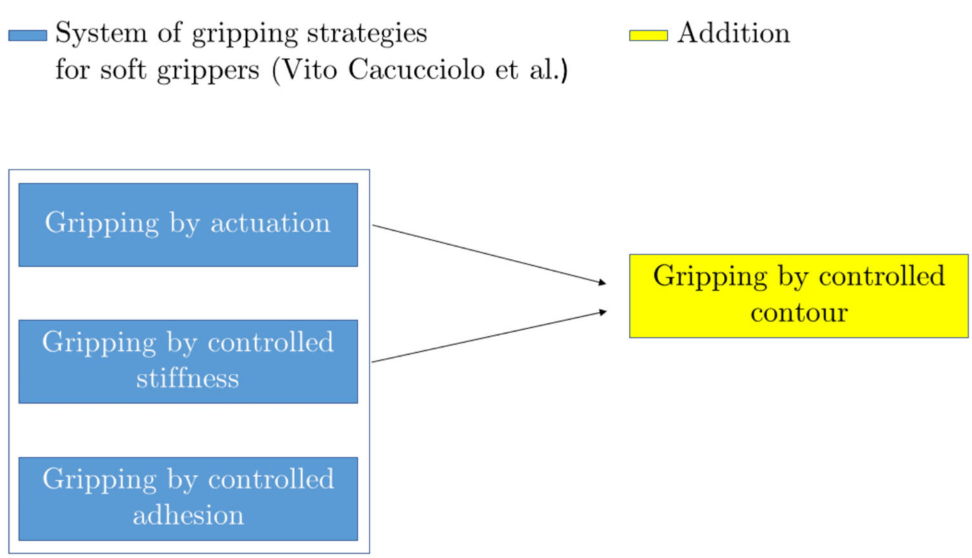
To actuate the star gripper from the rest position, a field below the coercivity H_C of the strontium ferrite must be established in order not to cause reprogramming. When using the permanent magnets, the effect can be used here that with increasing distance of the magnet surfaces, also, the magnitude H of the magnetic field decreases. However, the homogeneity and parallelism of the field lines also decrease. In the experiment, this effect manifested itself in the fact that the gripping fingers showed uneven responses to the magnetic actuation field. This influencing effect is shown in Fig. 9. Complete deflection of the gripper can be achieved with a magnetic actuation field of $H = 62mT$. Since smaller magnetic flux densities are required for actuation than for programming, the use of Helmholtz coils is an obvious choice here.

While the duration of the actuation depends on the built-up of the magnetic actuation field, the fastest tested actuation showed a time constant of around $t_a = 0.1s$. The repeatability is investigated and proofed by around $n = 200$ actuation cycles. It should be kept in mind that the repeatability is tested for its feasibility, not for its statistical limitations. Unspecific long-time tests showed a degradation of the deflection of the actuation, which is linked to the micro-material reversibility.

3.3 Definition of a new category of soft gripper

The magnetoresponsive material, processed by additive manufacturing and programmed in the magnetic field, has a certain elasticity or stiffness at rest. When an actuating magnetic field is applied, the particles align themselves in chains according to their dipole orientation and the body changes its shape. A certain shear force is required to break these chains. This further force indicates that the stiffness of the material has changed, and thus, the category gripping by controlled stiffness applies. When an external magnetic field is applied to a magnetoresponsive soft robot, its geometry changes due to the magnetic moment. This deformation of the body due to an external drive, in this case, the external magnetic field, suggests the category gripping by actuation. Vito Cacucciolo et al. themselves note that there is an overlap between the defined categories. For this reason, the category gripping by controlled contour is created in this paper. The reason is a controllable contour of a geometry. No distinction is made whether the contour change is due to the stiffness change or the external actuation of the structure. This addition is shown in Fig. 10.

Fig. 10 Categories of soft grippers published by Vito Cacucciolo et al. extended by the addition "gripping by controlled contour." This is a hybrid category between "gripping by actuation" and "gripping by controlled stiffness" with respect to the gripping by the controllable contour of a geometry



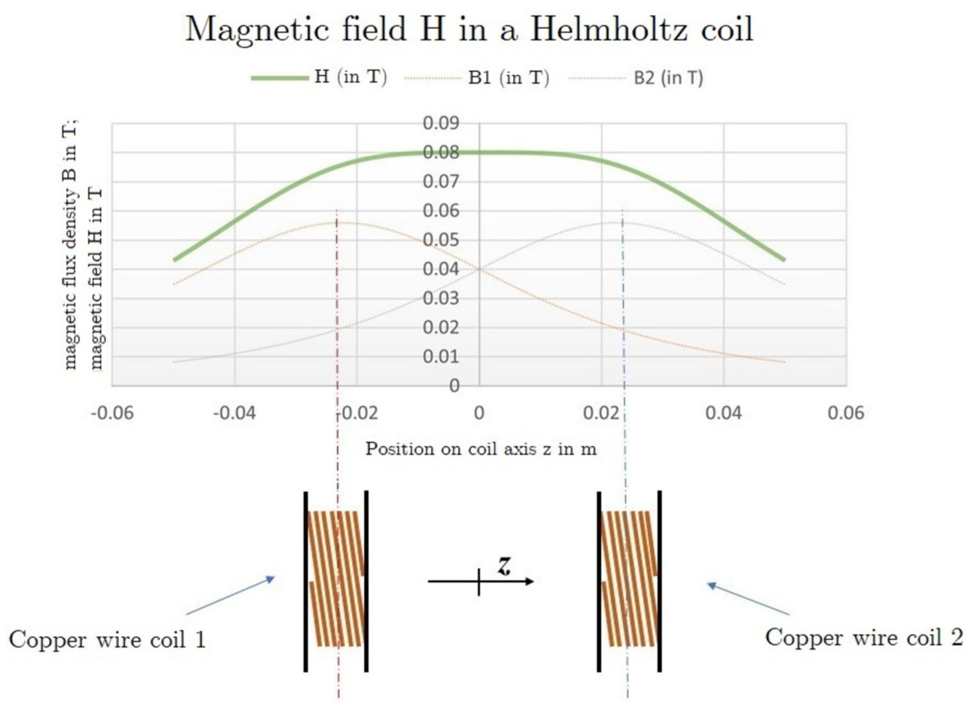


Fig. 11 Overlapping of the magnetic flux densities $B1$ and $B2$ generated by two coils (1 and 2) in a Helmholtz coil configuration with resulting magnetic field H. The green line shows the constant amplitude of H between $-0.01m < z < 0.01m$ where also a homogeneous field with parallel field lines is given

3.4 Design of the Helmholtz coil

The advantage of a Helmholtz coil is in particular that homogeneous, continuous fields are generated and that their magnitude H can be controlled as shown in Fig. 11. It is possible to create a desired magnetical torque τ in a layout with a magnetoresponsive part and a configuration of Helmholtz coils [25]. A single current-carrying coil generates a field with an extrema in the middle of the coil along the line of the imaginary axis. Outside the boundaries of that single coil, the field lines follow a curved path. If a second coil is applied along the same axis with a certain distance between each other, both fields become influenced. If the two coils are close enough, the intervening field lines are straightened and fused. As Fig. 11 visualizes, the resulting field between the coils is the sum of the single magnetic flux densities B of the two coils. In this manner, a greater amount is achievable by using two instead of one coil. The dimensions of the star gripper require a coil distance of $d = 45mm$. The diameter of the homogeneous field must correspond to $D = 90mm$, which corresponds to a radius of $R = 45mm$. The relation $d = R$ corresponds to an optimal utilization of a Helmholtz coil for a field which is continuous in magnitude H. With these dimensions, there is an available Helmholtz coil which can generate a maximum magnetic flux density of $H = 80mT$. It has $N = 98$ turns and can carry a current of $I = 40A$. To check whether this is sufficient for actuation, an approach was developed for the analytical calculation of the required magnetic field H of the Helmholtz coil.

The result of Eq. 6 with the model parameters, mentioned in Fig. 6, is the magnitude of the required external magnetic actuation field $H = 15.3mT$. In comparison with the maximum magnetic flux density of $H = 80mT$ of the previously mentioned Helmholtz coil, this confirms the possible use of the latter in the model case.

3.5 Additional verification of the calculation by means of a teslameter

By means of a teslameter with Hall probe, magnetic flux densities can be measured tangentially to the probe lance. In this way, it is checked whether the flux density for the actuation of the specimen calculated in the model can also be verified in the experiment. First, a specific flux density is set which places the specimen in the desired position. Then, the specimen is removed, and the Hall probe takes its place to obtain the most accurate result. Without taking into account the sources of error in the experimental setup, a magnetic flux density of $H = 17.0mT$ with a standard deviation of $s = 1.4$ can be measured as shown in Table 4. Thus, the magnitude of the analytical result $(15.3mT)$ is verified by a sample size of $n_s = 5$. The supplementary information

Table 4 Verification of calculation

Specimen	Calculation of H	Measurement of H
$15 \times 15 \times 1mm$	$15.3mT$	$17.0mT$ $(s = 1.4)$

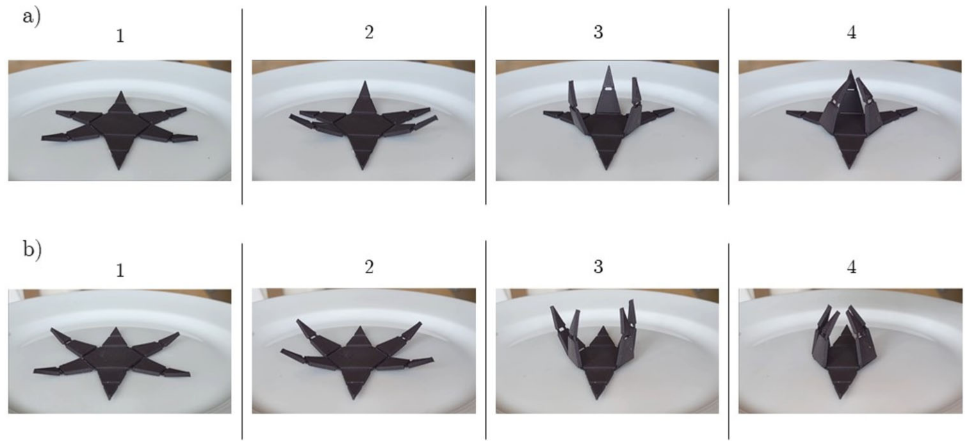


Fig. 12 The diversity of the programming of the star gripper is only limited by the maximum deformation of the gripper. The external magnetic actuation field H rises from (1) to (4). **a** Here are only three arms programmed to be closed while three arms stay at rest. **b** Here are four arms programmed to be closed while two arms stay at rest

provides a video of an actuation where the external field H is monitored in a different way than explained in the latter. It has to be known that the hall probe was attached under the platform. So the shown value differs lightly from the exact magnetical actuation field, but gives a good scope for the grippers' behavior.

In the future, this method will allow a theoretical design of soft gripper functions before manufacturing. The approach provides the basis of a simulation for the behavior of a soft gripper with respect to forces, moments, and kinematics during programming and actuation.

3.6 Multiple diverse reprogramming

The magnetoresponsive material developed for this study offers the potential for unlimited reprogramming. By shaping the body in a targeted manner and exceeding the coercivity with an external magnetic field, it is possible to reprogram the target geometry infinitely. The sole limiting factor is the maximum deformation of the printed body. Figure 12 illustrates the feasibility of multiple reprogramming of a single body. However, it must be taken into account that the reversibility of the actuation is decreasing with every reprogramming and further actuation.

Reprogramming is possible due to the ability of the body to polarize isotropically as a result of the coercivity H_C and remanence B_R of the strontium ferrite particles. When compared to the contributions of Kricke et al. and Kim et al., whose process does not allow reprogramming, this represents an evolutionary step in 4D printing [10, 11], even though the interactions of the boundary layer between particle and TPC need further investigation.

4 Conclusion

The aim of this work was to develop and manufacture a soft gripper from magnetoresponsive material and to derive an analytical approach for designing the magnetic field required for actuation. A particular focus was placed on the development of a straightforward process that would enable the repeatability and unlimited diverse reprogramming of the printed body. With the help of a 3D printer, the test specimens were printed using the process MEX-TRB/Cp/TPC,$SrFe_{12}O_{19}$. For the first time, a magnetoresponsive material could be processed using a commercial AM printer. This made it possible to research magnetoresponsive 4D printing quickly and cost-effectively.

The shape of the developed soft gripper is based on the bionics of the gripper for the very small insertion sites, such as those found in in vitro fertilization or minimally invasive operations. Here, an intervention can be carried out by remote manipulation by external magnetic fields. For this reason, a star-shaped point-symmetric flat body with two joints per gripping finger was printed. The gripper was deformed and programmed in an external magnetic field. A pair of two permanent magnets made of neodymium, which have a remanence B_R above the coercivity H_C of the magnetoresponsive material, is suitable for programming. For actuation, a controllable Helmholtz coil guarantees a homogeneous, parallel, and continuous field along a defined section. It can generate a sufficient magnetic flux density H, which is below the coercivity H_C of the magnetoresponsive material.

The actuation stimulus by means of a Helmholtz coil allows a specific control of the stimulus intensity, so the soft gripper can achieve even a partial movement. It turned out that a Helmholtz coil is unsuitable for programming because it must be disproportionately more powerful than for actuation due to the need to exceed the coercivity H_C of the magnetoresponsive material. The analytically determined necessary magnetic flux density for the magnetic torque was verified by a model in experimental studies. The calculated flux density ($15.3 mT$) and the measured one ($17.0 mT$) meet the expectations in terms of small deviations. In the future, this analytical approach will enable the design of the actuation magnetic field of a soft gripper before its fabrication. At the same time, the approach can initiate the development of soft robot simulation.

The soft gripper systematics of Vito Cacucciolo et al. do not allow a singular assignment of magnetoresponsive materials in 4D printing. They fulfill attributes from two of three groups. They can be actuated by stiffness changes as well as by an external drive. For this reason, the new category gripping by controlled contour is created here. The body thus exhibits a controllable contour. Thus, the new definition does not refer to the distinction of deformation by stiffness change or by external drive.

In the theoretical idealized view of programming and actuation of 4D printed soft grippers, it is only possible to execute the programmed deflection to 100%. Stopping the movement at any point in time is theoretically impossible. This is because there is only exactly one deformation where the field lines from the magnetic flux density of the body and those from the external actuation magnetic field are parallel. In reality, however, this is not the case, since external fields of different magnitudes can be used to reduce the bending of the gripping fingers. Parallelization of the field lines is not completely achieved.

The present work shows the strengths and limitations of the chosen method. The 4D printing of the developed material allows a fast actuation (around $t_a = 0.1s$) due to a short time constant and the ability to repeat the triggered response (around $n = 200$ cycles). An evolutionary milestone is the multiple and diverse reprogramming. The device allows large and small volume actuation, respectively. The need for post-print programming is an additional process step that limits the ease of 4D printing of magnetoresponsive materials. The programming with permanent magnets is problematic in terms of the inhomogeneity of the field.

For further research, the limitations of repeatability and a deep dive into reversible micro-material behavior are going to be investigated by measuring hysteresis loops and first-order reversal curves with a permagraph. Also, the design of the magnetical actuation field and the soft robot simulation will be intensified through the application of a Helmholtz coil. A magneto-mechanical material model is going to be developed and embedded in a simulation. The gradation of mechanical and magnetical properties will be investigated with a multi-material pellet printer by printing different mass ratios.

Practical applications could include soft robots and gripping tools in explosive environments, minimally invasive surgery, and life-like moving models of organs for medical training.

Supplementary information

A supplementary video file is provided showing the entire process of 4D printing a magnetoresponsive material. The video includes the design of the soft gripper using Catia V5 CAD software, its printing on a Prusa i3 MK3S+, forming using a printed PLA box, programming using a self constructed magnetisation device and the final actuation of the soft gripper. The actuation is monitored by measuring the magnetic flux density.

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1007/s00170-024-14605-5.

Acknowledgements The authors would like to thank the Freiburg Materials Research Center for technical support.

Author contribution Daniel Kehret, Stefan Junk, and Henning Einloth contributed to the study conception and design. Material preparation, data collection, and analysis were performed by Daniel Kehret. The first draft of the manuscript was written by Daniel Kehret, and all authors commented on previous versions of the manuscript. The review process was done by Daniel Kehret and supported by Bastian Rapp. All authors read and approved the final manuscript.

Funding Open Access funding enabled and organized by Projekt DEAL.

Declarations

Competing interests The authors declare no competing interests.

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