

Material characterization and precise finite element analysis of fiber reinforced thermoplastic composites for 4D printing[☆]



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ABSTRACT

Four-dimensional (4D) printing, a new technology emerged from additive manufacturing (3D printing), is widely known for its capability of programming post-fabrication shape-changing into artifacts. Fused deposition modeling (FDM)-based 4D printing, in particular, uses thermoplastics to produce artifacts and requires computational analysis to assist the design processes of complex geometries. However, these artifacts are weak against structural loads, and the design quality can be limited by less accurate material models and numerical simulations. To address these issues, this paper propounds a composite structure design made of two materials – polylactic acid (PLA) and carbon fiber reinforced PLA (CFPLA) – to increase the structural strength of 4D printed artifacts and a workflow composed of several physical experiments and series of dynamic mechanical analysis (DMA) to characterize materials. We apply this workflow to 3D printed samples fabricated with different printed parameters to accurately characterize the materials and implement a sequential finite element analysis (FEA) to achieve accurate simulations. The accuracy of deformation induced by the triggering process is both computationally and experimentally verified with several creative design examples and is measured to be at least 95%, with a confidence interval of (0.972, 0.985). We believe the presented workflow is essential to the combination of geometry, material mechanism and design, and has various potential applications.

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1. Introduction

Starting from the last century, fused deposition modeling (FDM) with additive manufacturing (3D printing) has become a widespread approach to build new structures [1,2]. Many types of thermoplastics, such as *polyether ether ketone* (PEEK) and *polyether ketone ketone* (PEKK), are frequently used in 3D printing technology for some high-end applications [3–5]. By feeding thermoplastic filament through the heated printing nozzle, the melted material is squeezed out and falls onto a horizontal low-temperature platform; specific printing paths along with printing parameters are designed and programmed to control the movement of the printing nozzle to form the desired 3D structure. As the deposited material accumulates, the structure is built layer by layer.

Based on the programmability of materials and printing paths, 3D printing changes the manufacturing process and provides an alternative approach for designers who prefer rapid structure modeling with customized materials and detailed distribution.

While the 3D printing industry has been introduced for both micron-length fibers and continuous fiber composites [6], printing self-assembled structures has not yet been fully adopted. The concept of self-assembly or the term *4D printing* [7], which uses shape memory materials (SMM) [1] and post-production actuation to combine 3D printing and time, is highlighted for its capability to fabricate adaptive structures [7–9] and could be traced back to the self-folding concept [10,11]. 4D printing produces artifacts with smart materials which enable shape-changing [12] in specific environment [10,13,14]. Traditional 3D printing is capable of creating various geometry by tweaking the geometry and printing parameters, while 4D printing allows users to control the final shape of an adaptive structure with programmable configuration.

Despite the possibility of designing programmable structures with 4D printing techniques, there exist several limitations. From the designer's perspective, there are two main limitations related to the 4D printing workflow: materials not being stiff enough and numerical simulations not being accurate enough. For materials

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usually used in 4D printing, thermoplastics is one of the most popular shape memory polymers (SMP), which is renowned for its high flexibility and material property transition under high temperature. However, many printable thermoplastics, including polylactic acid (PLA), have poor mechanical performance, which is not favorable in structural design. For numerical simulations, finite element analysis (FEA) is a commonly used technique to predict the deformation of the designed structure [15]. However, without high simulation precision, FEA results cannot truly help designers predict the structure's final configuration, and designers must keep testing until the final desired shape is achieved, which is extremely expensive and time-consuming [8,16]. To facilitate the 4D printing design process, both material and simulation aspects need upgrading: materials used in 4D printing need to be strengthened, and prediction from simulation needs to be more accurate to speed up the forward design process. In this paper, we provide solutions to both aforementioned issues.

Carbon-fiber PLA (CFPLA) is known for its high stiffness-to-weight ratio and is commonly used as a reinforcement material. PLA is soft and easy-to-deform when heated above the glass transition temperature (T_g), which enables PLA to be the most widely used thermoplastics in 3D printing. To maintain the merits of flexibility and easy-processing, meanwhile elevating its mechanical performance, we devise the use of CFPLA. With CFPLA, the designed structure can be strengthened along the direction of carbon fiber alignment; since the matrix material of CFPLA is still PLA, it can be easily 3D-printed with slightly higher extrusion temperature. In addition, based on 4D printing techniques, CFPLA can be easily combined with PLA to form bi-layer fiber-reinforced composite (FRC), which is relatively more difficult to adopt traditional fabrication approaches [6]. By programming the printing path, FRC can be conveniently and effectively designed and fabricated layerwise, and the accuracy of FRC design can also be improved.

Precise material characterization is important to ensure the accuracy of FEA. A lot of research has been conducted to characterize polymeric material properties [17–19], including 3D printing filament materials like PLA and CFPLA. Soares et al. [20] used an incompressible, isotropic neo-Hookean hyperelastic material to describe the mechanical response of stents. The static and dynamic loading effect on degradation of PLA stent fibers was studied to further define a hyperelastic incompressible material [21]. Khan et al. [22] combined compressible Ogden hyperelastic model and generalized the Maxwell model to obtain the linear viscoelastic behaviors of biodegradable polymers. A modified Eyring energy was utilized to define viscoplastic behavior of PLA [23]. Eswaran et al. [24] introduced anisotropy into the material modeling. A shape memory model was used to define thermal expansion coefficients for each printing layer [25].

In this paper, both PLA and CFPLA are tested based on repeated dynamic mechanical analysis (DMA), and specific models are subsequently proposed to characterize their material properties. Another cause of low FEA fidelity is the influence of residual stress and body force on 4D-printed samples during the triggering process. To improve the precision of simulation, we devise a new sequential FEA, which takes the influence of residual stress and body force into consideration, and minimizes the necessity of test-printing intermediate designs with an accurate prediction of the final shape from simulation results. With a high accuracy of the final configuration prediction, we propose a forward design process – or workflow – to summarize the iterative process from the material characterization to the FEA, and the design finalization and fabrication can be achieved by going through this proposed workflow. The main contributions of this paper can be summarized as follows:

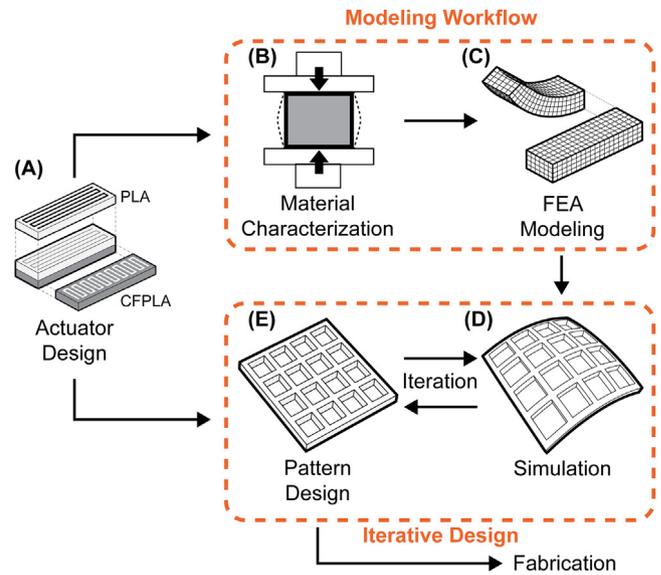


Fig. 1. The workflow of iterative 4D printing design. (A) shows the structure of a block with PLA as an actuator and CFPLA as constraint. (B) shows the process of material characterization based on DMA experiments. (C) refers to the FEA modeling on a bending unit. (D–E) show the iterative design process based on the simulation. This iterative process yields the finalized design, and leads to fabrication.

1. A novel workflow is proposed for forward design, with accurate material property characterization and precise FEA simulation. This workflow supports robust and accurate fabrication of the designed object through an iterative optimization process and accurate control of the final configuration.
2. The material properties of 3D printing polymers, including both PLA and CFPLA, are characterized in a precise way based on the DMA experiments. The characterization results are effectively incorporated into FEA with accurate mathematical models.
3. A sequential FEA is developed to achieve accurate simulation results, considering both the residual stress releasing and the body force creeping. We simulate these two processes in a sequence to precisely derive the final deformation of the fabricated product.

The layout of this paper is as follows. Section 2 overviews the workflow. Section 3 briefly introduces the geometric scope, structure design and fabricating parameter determination. Section 4 mainly discusses the physics behind material properties and precise material characterization of both PLA and CFPLA. Section 5 introduces simulation modeling and sequential FEA. Section 6 shows some simulations and design results derived from the proposed workflow. Section 7 draws conclusions and points out potential improvement and future directions.

2. Overview of the workflow

In this paper, a workflow is proposed to precisely simulate the self-morphing behavior of composite thermoplastic materials and structures. The proposed workflow consists of three major processes: material characterization, FEA modeling and simulation, and pattern design. As shown in Fig. 1, the workflow firstly characterizes the material property based on the DMA tests, and then creates FEA models and establishes material and boundary condition settings for Abaqus. In the end, the workflow simulates the final configuration to achieve the iterative forward design and fabricate the final product.

Material characterization. The material properties of PLA and CFPLA are characterized by using DMA experiments, which are conducted on strip samples under the temperature of 80 °C, aiming to obtain the stress–strain curves and derive the material’s hyperelasticity and viscoelasticity, respectively. The Mullins effect is taken into consideration to address material stress-softening [26].

FEA modeling and simulation. Based on the aforementioned material characterization results, the material definition is determined in each FEA model. The residual stress values of each actuator and constraint are obtained by matching the triggering experimental results with their corresponding simulation results. Then, a sequential simulation is implemented to include both the residual stress releasing and the body force creeping. In this way, the simulation precision can be ensured and maintained at a high level.

Pattern design. With precise simulation results, designs can be created and refined iteratively. The simulation results from intermediate geometry can be obtained from the aforementioned FEA sequences, providing information on the difference between the current design and desired final configuration. An iterative process of forward design is then implemented from the initial geometry to the finalized design.

3. Unit structure design and fabrication

The theorem behind the deformation of 4D printing structures and materials is the residual stress releasing in response to external stimulation. 3D printed PLA materials release embedded residual stress when treated with a temperature higher than T_g ; see Fig. 2(A). The releasing of residual stress will result in the shrinkage of PLA along the printing path direction.

In this paper, all the designed products consist of bending units. As shown in Fig. 2(B–C), each unit is a cuboid with two different blocks – the top actuator block and the bottom constraint block, and they form the *bi-layer unit* [27–29]. Each block has a different printing path direction: the top block has a longitudinal printing path direction while the bottom block has a lateral printing path direction. The bending unit is the most fundamental structural unit in this work. When a bending unit is placed into an 80 °C environment, the block with longitudinal printing paths will shrink more than the block with lateral printing paths, creating a difference in shrinkage and resulting in a bending behavior. The block with larger shrinkage is called an *actuator block*, and the layer with less shrinkage is called a *constraint block*.

Various structures and patterns are adopted in product design. In this study, the *grid structure* is implemented for our design workflow, and its pattern is formed by bending units. The distribution of bending units with different actuators and constraints determines the deformation and configuration of the grid structure. Fig. 2(D) shows that each bending unit is assigned with a different actuator block and a constraint block, connected with pure constraint blocks as joints to form a grid. By controlling the length of the actuator block on each bending unit, different bending curvatures can be achieved and further combined to form a specified 3D shape.

3.1. Three printing factors

3D printing is the basic method we use for material processing and structure fabrication. The brand of 3D printer is *Modix Big60*. To effectively control the deformation of the printed structure, filament materials and printing parameters are both tested. The distribution of actuator blocks and constraint blocks and the design of the structure should also be studied. In this paper,

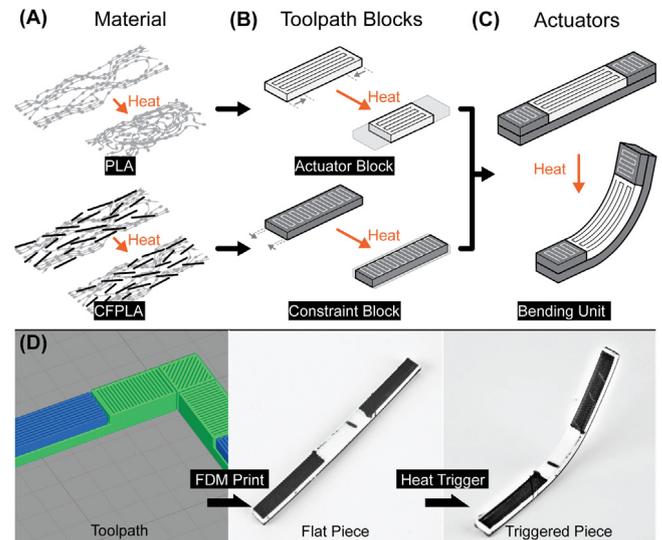


Fig. 2. Microscale view of PLA material and block structure with different functional components. (A) The polymer chain inside the 3D printed PLA structure. (B) Anisotropic blocks consisting of two layers with different printing path directions: the actuator blocks (white) and the constraint blocks (gray). (C) Bending unit with actuators (white) and constraints (gray). (D) The process from programmed printing path to fabricated flat piece and triggered bending unit.

three printing factors with dominating effects on morphing characteristics are studied: printing path orientation, printing layer thickness, and filament material property.

Printing path orientation determines the shrinking direction of the thermoplastic material. During the FDM process, residual stress is embedded in PLA along the printing path as the filament is being extruded from the nozzle. As a result, the printed material shrinks along the specified printing path. Using this specific shrinking behavior of PLA material, different bending units can be created for design, as shown in Fig. 2.

Printing layer thickness also affects the shrinkage ratio. Thinner layer thickness requires more printing layers during the FDM process, which means more residual stress is embedded into PLA structures. As a result, higher shrinkage ratio can be achieved by thinner layer thickness. By combining blocks with different layer thicknesses into one bending unit, different shrinkage ratios are introduced to achieve various bending performance.

Filament material property significantly changes for different shrinkage ratios. The PLA and CFPLA filaments do not exhibit the same shrinkage ratio after the FDM process, because the CFPLA material is embedded with chopped up carbon fibers along the printing direction. The chopped-up fibers greatly improve the stiffness along the printing direction and significantly undermine the shrinking performance of the block.

3.2. Determining optimal printing parameters

To characterize material properties for different printing parameters through DMA experiments, we studied three types of thermoplastic composites. All experimental samples are straight bending units with the dimension of 75 mm × 10 mm × 4 mm. Each sample consists of an actuator block at the top and a constraint block at the bottom. Actuator blocks are printed with a straight printing path that is along the sample’s longitude direction, and constraint blocks are printed with a lateral printing path. After the printing process is completed, samples are fixed at one end and placed horizontally in a water bath at 80 °C – which guarantees the homogeneous and stable heat transferring – to

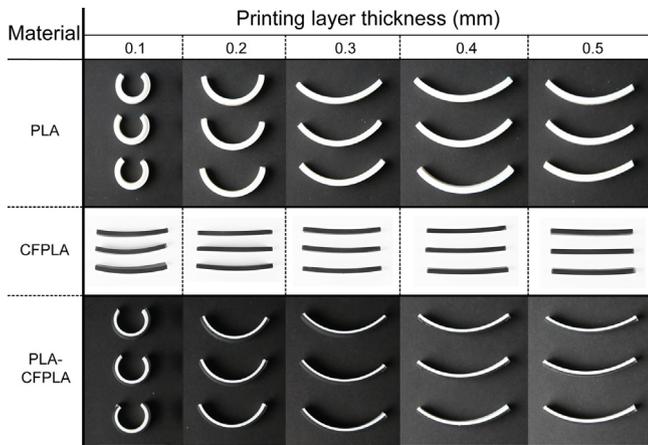


Fig. 3. Experiment results of three types of thermoplastic materials: PLA, CFPLA and PLA-CFPLA composite bi-layer blocks, with varying layer thicknesses from 0.1 mm to 0.5 mm. Results show that PLA bending units have the largest bending deformation while CFPLA bending units have the smallest bending deformation.

trigger the self-morphing process. In the experiments, three types of thermoplastic material designs were selected: PLA, CFPLA, and PLA-CFPLA combined bi-layer composite.

Experiments are also conducted to study the effects of the printing layer thickness on material bending behaviors. All experimental samples are printed with 0.6 mm nozzle, 3000 mm/min printing speed, but different layer thicknesses. 0.1 mm, 0.2 mm, 0.3 mm, 0.4 mm, and 0.5 mm are chosen as the printing layer thicknesses for three batches of sample blocks, categorized by different material types.

Fig. 3 presents the deformed shapes for experimental samples with varying layer thicknesses. We can observe that PLA samples with 0.1 mm printing layer thickness have the best bending performance among all PLA samples, while PLA samples with 0.5 mm printing layer thickness have inadequate bending performance. All CFPLA samples show minimal bending angles, indicating that the shrinkage ratio along the printing path direction of CFPLA is generally lower than that of PLA. Since CFPLA has limited bending performance, we choose the PLA-CFPLA composite bending units as the primary component in our design. This composite material is purposely designed to take advantage of the strength of CFPLA without sacrificing too much bending performance. Fig. 4(A) shows that composite samples have similar shrinkage ratio and bending angle as PLA samples.

Fig. 4(B) shows the stress-strain curve of PLA-CFPLA combined bending unit for five different printing layer thicknesses. Samples with printing layer thickness larger than 0.3 mm do not have a large enough bending angle; however, choosing a

printing layer thickness < 0.2 mm produces inconsistent results when dealing with layer thickness smaller than 0.2 mm. The Modix Big60 3D printer, designed for large-size 3D printing tasks, has a printing volume of 61 cm^3 and implements a unified bed leveling technique to actively adjust for small dents across the aluminum printing bed during printing. The design tolerance for printing bed dents is 0.2 mm, according to the manufacturer's instructions. As a result, the automatic z-height adjustment during printing leads to quality inconsistency if the printing layer thickness is smaller than 0.2 mm. Therefore, we choose the PLA-CFPLA combined bending units with 0.25 mm layer thickness for our workflow implementation.

3.3. Flexural test

Concerning the flexural test, three-point bending and four-point bending are conducted on bending units. The dimension of each unit sample for three-point bending test is $60 \text{ mm} \times 4 \text{ mm} \times 1.6 \text{ mm}$, and the effective length of the supporting span is 40 mm. The dimension of each unit sample for four-point bending test is $100 \text{ mm} \times 8 \text{ mm} \times 8 \text{ mm}$, and the loading span and the supporting span are 23.56 mm and 70.68 mm, respectively. Testing samples of PLA, CFPLA, and PLA-CFPLA composite are designed and tested with residual stress pre-released under room temperature to observe the corresponding flexural modulus and strength. The experimental setup of the three-point bending test is shown in Fig. 6(C). Fig. 4(C) shows the experimental result of the three-point bending in terms of flexural stiffness for three types of samples, and the corresponding four-point bending experimental result is listed in the supplementary material (Figure S3). From both experimental results, we can observe that although the CFPLA sample has the highest flexural stiffness, it is so brittle that it may break within relatively small effective deflection range; the PLA sample is capable of withstanding large deflection, but it has the lowest flexural stiffness among all three types of samples. By combining CFPLA and PLA into the bi-layer structure, the PLA-CFPLA composite bending unit can not only exhibit high flexural stiffness, but also withstand large deformation at the same time. Its strengthened mechanical properties are capable of expanding the application space of the designed structure under different loading cases.

4. Material characterization

Material characterization is important for precise simulation. Both PLA and CFPLA are characterized. The physics of PLA and CFPLA include their microstructure, anisotropic behavior and nonlinear and time-dependent material behavior. Based on the material physics, specific models are identified to facilitate FEA simulations.

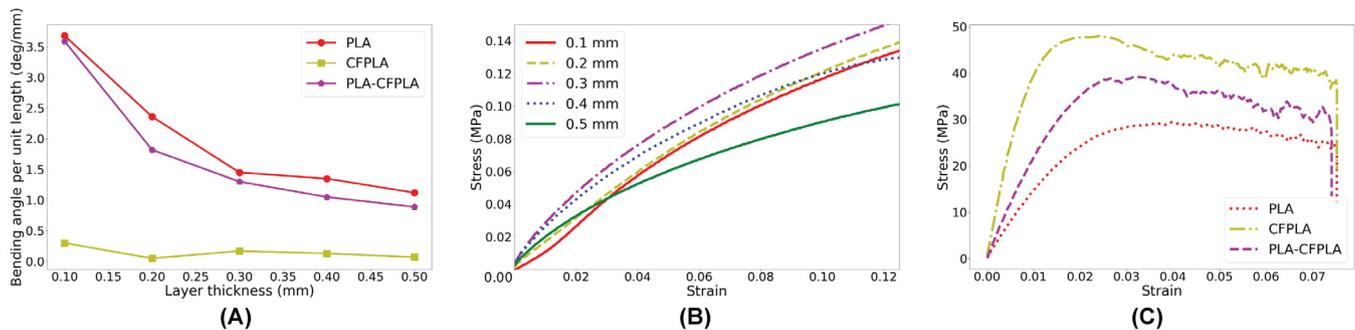


Fig. 4. The DMA experiment results for identifying the bending unit structure and the printing parameters. (A) Bending angle per unit length vs printing layer thickness for PLA, CFPLA and PLA-CFPLA composite. (B) Stress-strain curves of PLA-CFPLA composite for different printing layer thicknesses. Based on the experiments, 0.25 mm is chosen as the printing layer thickness. (C) Stress-strain curves with three different bending unit structures in the flexural test.

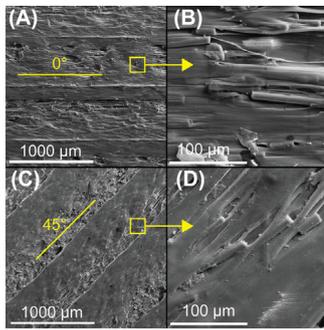


Fig. 5. SEM images of CFPLA. (A–B) show zoom-out and zoom-in pictures of the sample with the printing direction of 0° . (C–D) show zoom-out and zoom-in pictures of the sample with the printing direction of 45° .

4.1. Material physics

As a thermoplastic polymer, PLA is composed of numerous polymer chains [30]. These chains are often entangled together, forming a micro-scale network. During the printing process, as the temperature arises above the PLA's melting point, slippage at the network's entanglement links may occur, introducing viscoelastic properties to the printed material. In the case of 4D printing, when we reheat the material over its glass transition temperature, these viscoelastic properties will play a dominant role in the material's deformation behaviors. Therefore in addition to the commonly observed plastic behaviors, the viscoelasticity of PLA must be considered in establishing an accurate material model.

The polymer chain networks of PLA usually have random orientations, and the material can be considered to be isotropic. Yet, when the filament material is extruded from the nozzle during the printing process, the polymer chains are stretched along the printing direction, causing the material to become anisotropic. CFPLA also undergoes the same process during 3D printing, and fibers within the printed blocks are aligned, increasing the material's stiffness along the printing direction. Fig. 2(A) illustrates the detailed distribution and anisotropic material property of both PLA and CFPLA, and Fig. 5 further evidences the fiber-alignment and anisotropy of printed materials with scanned electron microscopy (SEM) images.

4.2. Material properties

Based on the material physics, PLA is expected to show both elastic and viscous behaviors. We adopted a hyperelastic constitutive model to describe the hyperelastic behaviors, and collect data by performing the uniaxial tensile test on both PLA and CFPLA with the RSA-G2 equipment. The PLA material is purchased from *Polymax*; the CFPLA material is purchased from *Proto-Pasta*, with the average length of carbon fibers less than $150 \mu\text{m}$. The following are the detailed testing procedures for each property of interest.

Thermal expansion and Poisson's ratio. Since the material and the structure are triggered in a high temperature, the material's thermal expansion rate (α_t) and the Poisson's ratio (μ) are considered in our workflow. These two parameters can be obtained by measuring the dimensional change of a 3D printed cubic sample before and after the uniaxial compression test. The cubic sample of size $5 \text{ mm} \times 5 \text{ mm} \times 5 \text{ mm}$ is fabricated and subjected to a compressing load perpendicular to the top and bottom surfaces (Fig. 6(A)). The obtained α_t and μ are shown as Table 1. Each result is with an error bound calculated from its confidence interval of measurement.

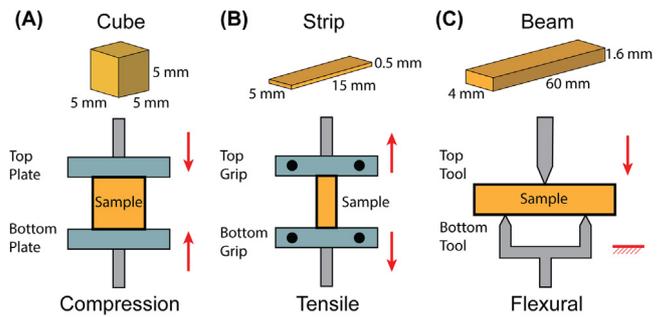


Fig. 6. DMA experiments setup. (A) refers to the DMA compression test with a cubic sample. (B) refers to the tensile test with a strip like sample. (C) refers to the three-point bending test with a bi-layer block sample.

Table 1

α_t and μ of PLA and CFPLA at 80°C .

Material	α_t ($1/^\circ\text{C}$)	μ
PLA	$(9.17 \pm 1.54) \times 10^{-4}$	0.419 ± 0.021
CFPLA	$(9.97 \pm 5.92) \times 10^{-5}$	0.359 ± 0.015

Hyperelasticity. The hyperelasticity data of the materials are obtained by performing uniaxial tensile tests on a strip sample of size $5 \text{ mm} \times 15 \text{ mm} \times 0.5 \text{ mm}$ (Fig. 6(B)). Each sample is heated to release its residual stress under 80°C . Since PLA can reach the rubbery state above T_g , stress-softening or the Mullins effect are taken into consideration. To capture this effect, four cycles of force loading and unloading process are applied to the samples with a speed of 0.005 mm/s and a 23% tensile strain limit. For CFPLA, due to its higher stiffness and smaller scale of deformation, the viscoelastic and plastic behaviors are insignificant during the triggering and deforming process. Therefore, we only consider the hyperelasticity for CFPLA by conducting the tensile test with a speed of 0.002 mm/s and a 3% tensile strain limit, while the Mullins effect and plasticity are not considered in the CFPLA material setup.

Plasticity. The plastic behavior of PLA is observed after cycles of force loading period. This behavior is described by identifying the anchor point, the intersection of the stress-strain curves and x -axis.

Viscoelasticity. We use the viscoelastic constitutive model to characterize the viscoelasticity of PLA. In the experimental setup (Fig. 6(B)), the equipment pre-strains the samples (0.0095) and applies oscillating loads (0.01 Hz to 100 Hz) to record the storage E' and the loss E'' modulus in the frequency domain to describe the frequency-dependent behaviors. Fig. 7 shows the modulus-frequency plot and the damping curve obtained for PLA actuator and constraint. The viscoelasticity experiment of CFPLA samples is also conducted, and the experimental result shows that the loss modulus to storage modulus ratio – which is also written as $\tan \delta$ – of CFPLA is always less than 1, which indicates that the viscoelastic behavior of CFPLA is not dominant. Detailed evidence based on the experimental result is listed in the supplementary material (Table S3, Figures S1 and S2).

5. FEA modeling and sequential simulations

To successfully simulate deformation of designed products, FEA processes with different boundary conditions and initial conditions need to be implemented in order. In this paper, we propose two kinds of sequential simulations: the sequential residual stress estimation and the sequential deformation simulation.

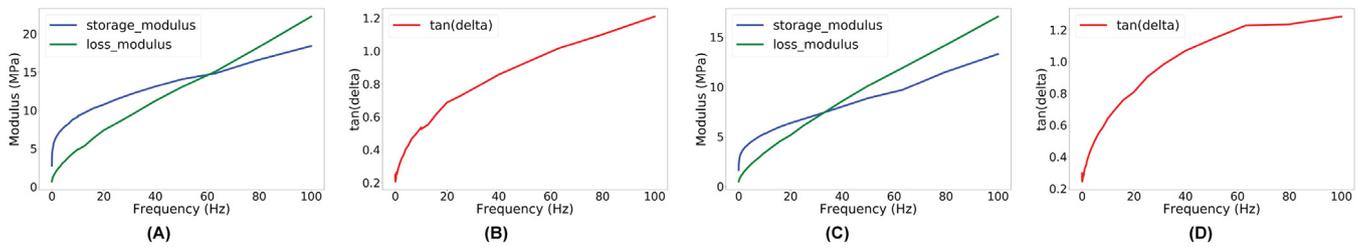


Fig. 7. Experimental results of the modulus-frequency plot and the damping curve for PLA actuator (A–B) and constraint (C–D).

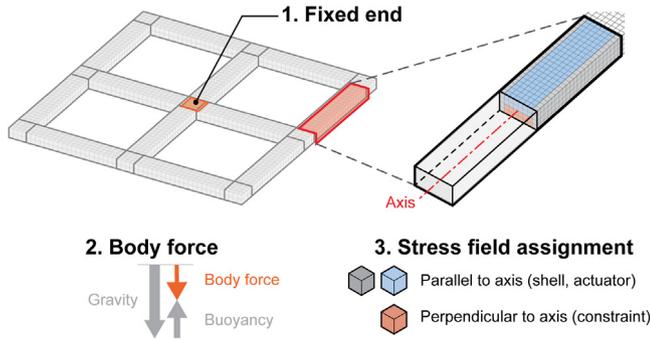


Fig. 8. Model discretization and boundary condition settings of a simple grid structure.

5.1. Sequential residual stress estimation

Since the deformation of a bending unit results from its embedded residual stress, identifying the initial residual stress is extremely important. Numerically identifying residual stress can be convenient and programmable by implementing the shooting method [31], which is a numerical method to obtain the initial residual stress and convert the boundary value problem to an initial value problem. Based on the shooting method, the process of residual stress estimation can be summarized in a sequence: (1) FEA modeling and material definition; (2) DMA experiment verification; and (3) residual stress and material property identification.

5.1.1. FEA modeling and material definition

FEA modeling includes meshing, boundary condition setting and material definition. As discussed in Section 3, the block and grid structure are the main structures studied in this paper, thus the geometry of all designed products can be easily discretized into hexahedral elements for FEA simulations in Abaqus. The boundary condition settings are consistent with the triggering experiment. Since the bending unit structure can be divided into actuator and constraint components, they are assigned with different material properties. The boundary condition settings are shown in Fig. 8, including the fixed region, the body force and the division of actuator and constraint components. For the material property settings, we consider hyperelasticity, plasticity and viscoelasticity based on the raw data obtained from the aforementioned DMA experiments.

In this paper, the material's *hyperelasticity definition* is assumed to be isotropic. To remove noise in the raw stress–strain data from experiments, a penalty spline based smoothing method [32,33] was implemented. The processed data for PLA is shown in Fig. 9. The four cycles of loading (printing) and unloading (releasing) processes are shown, and the main loading curve (the green dashed line) can be derived from the raw data. The material's hyperelasticity is merely determined by the unloading curve,

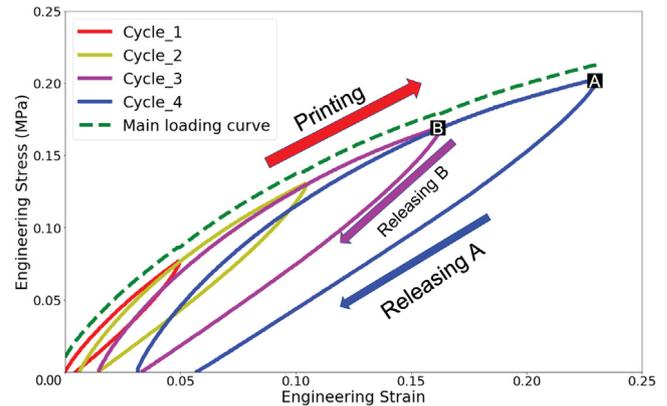


Fig. 9. The loading–unloading tensile test results for PLA. The stress–strain curves from four cycles of force loading and unloading clearly show the material's Mullins effect. The main loading curve can be obtained from the stress–strain curves marked as the green dashed line, which refers to the printing or residual stress embedding process. Different loading stresses (marked as A and B) yield different unloading stress–strain curves, which refer to the triggering or residual stress releasing process.

which is also relevant to the initial residual stress value. Different initial residual stresses may lead to different hyperelasticity definitions.

By using the aforementioned anchor points data, the material's *plasticity definition* is also assumed to be isotropic. The unrecoverable deformation of the corresponding material can be obtained from the anchor points, and the Mises plasticity with an associated flow rule was implemented to model the material's plastic deformation [34]. In Abaqus, the hyperelasticity and plasticity material properties are further combined. When only one set of data is available, the Marlow model is recommended [35]. The Marlow strain energy potential [36] is chosen to fit the main loading curve, and the unloading curve is described by the Marlow model plus the Ogden–Roxburgh model as the damage [37] and plasticity terms [38]. From Fig. 9, we can observe that different stress values on the main loading curve correspond to different unloading curves, which also verifies that the material property during the stress-releasing process is highly dependent on the initial residual stress value.

The *viscoelasticity definition* is obvious especially for PLA and is characterized by the storage modulus, the loss modulus, and the $\tan\delta$ derived from the DMA experiments (see Section 4). Based on the explanation in Section 4.2, the viscoelasticity is not the dominant mechanical property for CFPLA material, thus we decide not to include the viscoelastic behavior into CFPLA's material definition.

Discussion 5.1. The Marlow model assumes that the strain energy potential is a function of the first strain invariant and is independent of the second strain invariant. Eliminating the second strain invariant from the strain energy potential has several benefits, as discussed in [39]. In our case, by using this model, we can reduce

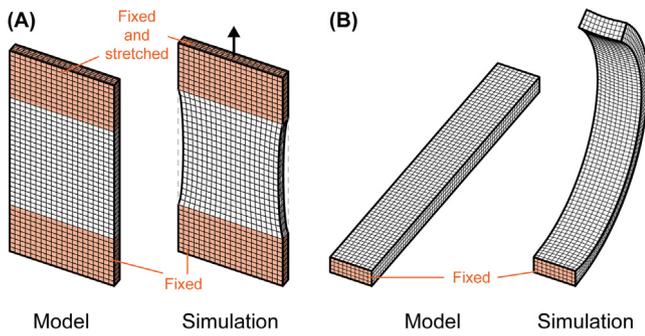


Fig. 10. The tensile testing samples for DMA experiments (A) and the bending unit samples (B). The colored area shows the applied boundary conditions during experiments. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

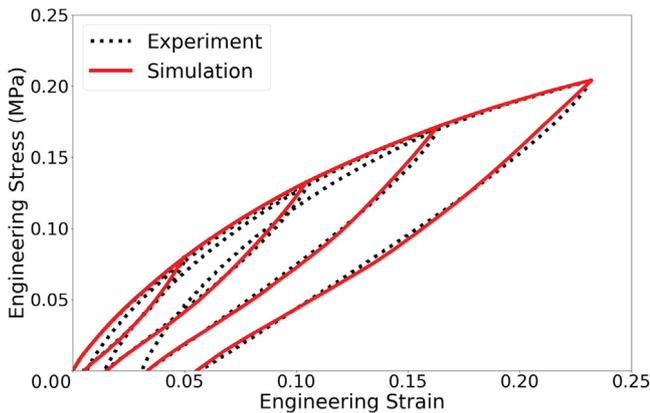


Fig. 11. Comparison between the simulation results and the tensile test data under cyclic loading conditions.

the number of experiments (the uniaxial tension test) required to describe the material behavior accurately. We can, therefore, avoid using complex experimental data from biaxial tension and planar tension for parameters calibration.

5.1.2. DMA experiment verification

After quantifying the material properties, a simple simulation model of the strip sample in DMA experiments was created and tested to check how the simulation results match the DMA experimental results. The dimensions of the strip sample are shown in Fig. 6(B), and the exact geometry is built for simulation. Fixing boundary conditions are set at the same position as in experiments, and the testing process can be simulated by applying a stretching load as shown in Fig. 10(A). The simulated stress-strain results can be subsequently derived and compared with the experimental results as shown in Fig. 11. We can observe that the simulation results match the experiment results pretty well.

5.1.3. Residual stress and material property identification

To precisely estimate the initial residual stress and finalize the material property definition, an iterative process is programmed to implement the shooting method. To show different bending curvatures for different beams, the actuator ratio, which is the ratio of the actuator block length to the total length of the bending unit, is controlled from 0 to 1. The bending units with different actuator ratios should have different bending curvatures, which may help precisely identify the initial residual stress value from the shooting method. The triggering experiment was conducted on three batches of bending units with different actuator ratios and the same dimension of 100 mm × 7.2 mm × 4 mm, and

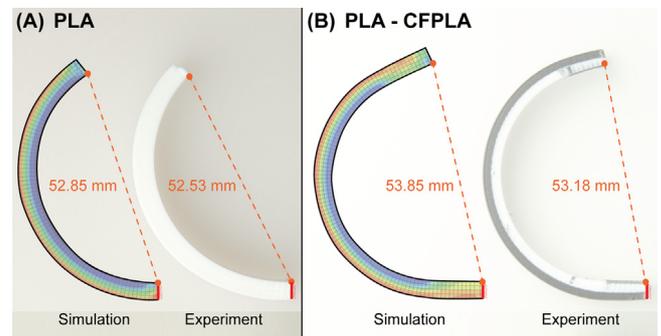


Fig. 12. Simulation and experimental results of a bending unit for PLA (A) and PLA-CFPLA composite (B).

the triggering environment is controlled at 80 °C. The distance between two selected points on two ends of the triggered block is measured to represent the bending curvature and describe the bending extent.

Meanwhile, the corresponding simulation model of the bending unit is built based on the aforementioned material property definition, and the boundary conditions are shown in Fig. 10(B). The initial residual stress for a particular bending unit is randomly set in Abaqus and the corresponding deformation of the block is derived after the simulation. Based on the deformation results, the difference of the measured distances between simulation and experiment can be obtained, and the initial residual stress value is subsequently adjusted to reduce this difference. Since the material's hyperelasticity is dependent on the initial residual stress value, the changing of the material property definition follows the changing of the initial residual stress. By iteratively conducting this adjustment, the initial residual stress value is identified, and the corresponding hyperelasticity definition is also finalized.

5.2. Sequential deformation simulation

Aside from accurate residual stress and material property identification, we propose a sequence of the deformation simulations, which can help improve the simulation accuracy especially for the cases with both the body force and the initial stress considered. Since the residual stress is identified as the initial condition for precise simulation, and we assume that the temperature during the triggering (deforming) process is a constant, the sequential deformation simulation is not time-dependent and temperature-dependent. Assume there is a model α to be simulated, the sequence includes (1) obtaining the initial stress value; (2) duplicating the model α to create a new model β , and simulating the model α with both the initial stress and the body force considered; (3) importing the deformation results from α after (2) into β as the initial state; and (4) removing the initial stress condition in β and simulating β with only the body force boundary condition.

To verify the aforementioned FEA sequences, we trigger two bending units with different material distributions and actuator ratios – one is a PLA bending unit with the actuator ratio of 1.0, and the other is a PLA-CFPLA bending unit with the actuator ratio of 0.75. We use the FEA sequences to simulate these two bending units with the same geometry and material settings as experiments (the FEA model and results of the PLA bending unit are shown in Fig. 10(B)). The experimental and simulation results are shown in Fig. 12. For the PLA and PLA-CFPLA bending units, the errors between experiment and simulation are 0.61% and 1.26%, respectively.

Discussion 5.2. The shooting method is used to convert the boundary value problem to an initial value problem, which can

simplify the problem solving process. In this paper, we assume that the residual stress is homogeneously distributed in the sample and is released linearly, and we use the distance between each specified point pair to measure the deformation. However, the actual deforming process of the 4D printed material is very complicated. For example, the rate of residual stress releasing is not a constant of time, and the distribution of the residual stress in each sample is not homogeneous. As such, further study is required to more precisely identify the stress and control the deformation of the material.

6. 4D printing of three creative designs

Three creative designs were completed using the proposed workflow to explore potential applications. Designers can use the workflow to create 2D grid structures in a CAD environment, and then simulate the grid deformation in the Abaqus software to visualize the final shape. With our workflow, the deformation of the grid structure can be altered toward the target shape by iteratively adjusting the initial 2D grid based on the intermediate simulation results. Numerous design iterations can be conducted without prototyping, and the accuracy of the simulation is verified by implementing the point-pair method – measuring the distance between any pair of two points in both experiment and simulation. The final design can be achieved with high efficiency at low cost, and the general accuracy of the simulation, which has a confidence interval of (0.972, 0.985), can greatly assist the forward design process.

The modular lamp cover is the first implementation example. The purpose of creating modular designs is to test the accuracy of our simulation tool and to allow for large scale implementation. The lamp cover is divided into three pieces: a 2×4 grid as the top section, a 2×2 grid as the bottom left section and another 2×2 grid as the bottom right section. Each sub-section design is constructed based on a 2×2 or 2×4 grid, and each grid consists of bending units and joints shown in Figs. 2 and 8. By combining bending units with different actuator blocks, the 2×2 or 2×4 grid structure can achieve specific edge shapes that match each other. Once the 2D grid structures are printed, they are placed in an 80-degree Celsius water bath to trigger the deformation. This approach drastically reduces the manufacturing time because all sub-sections can be printed flat without any support structure. With the workflow, designers can iterate designs for the best matches without printing and triggering each design, significantly reducing time and cost. Fig. 13(A) shows the design iterations completed with our proposed workflow. Fig. 13(B–C) show that our simulation tool can precisely simulate the actual deformation.

The FEA results for each lamp cover sub-section are shown in Fig. 14(A–B). Since both bottom sub-sections are perfectly symmetric, only one FEA result is needed for analysis. For each subsection, three different point pairs are chosen to quantify the accuracy of the simulation results. The distance between each point pair is measured in both simulation results and experimental results. Then the simulation error is determined by calculating the difference between experimental and simulation distances. Part of the distance results and the calculated errors are listed in Table 2. More measurement results are provided in the supplementary material (Table S4). Based on the aforementioned measurement results, the confidence interval of the accuracy is (0.968, 0.998).

The bottle holder is the second implementation example of our workflow. The purpose of designing a bottle holder is to enable people with disabilities to hold bottles without handles. The bottle holder is an evolved version of a 2×2 grid structure with an approximate dimension of 500 mm by 200 mm. By changing the construction of the 2D grid, complex and intersecting 3D structures like the bottle holder can be realized. With the

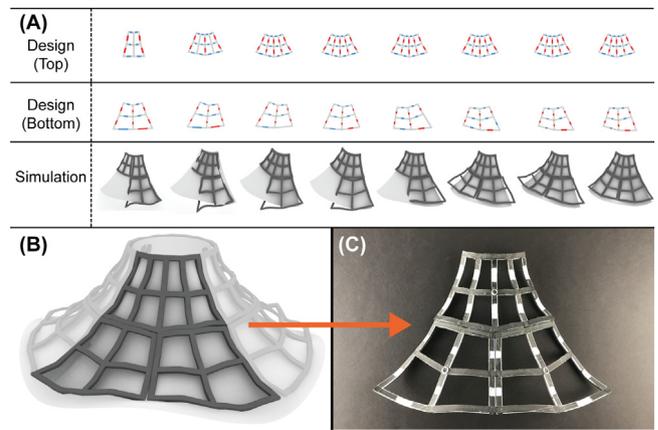


Fig. 13. Modular lamp cover design iterations. Results show the lamp cover pieces can fit the lamp quite well after eight iterations.

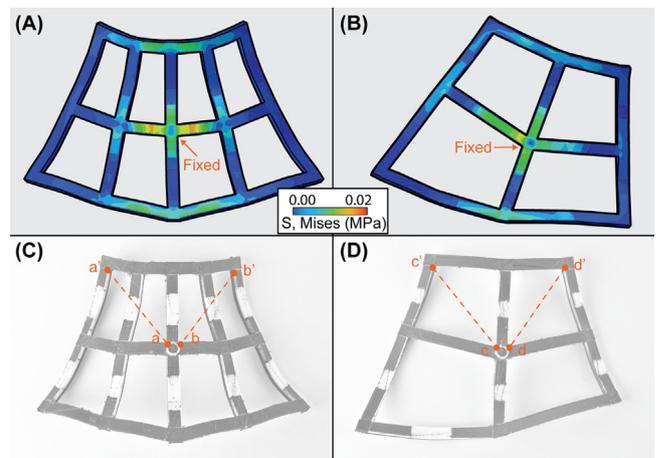


Fig. 14. Final configurations from simulation (A–B) and experiment (C–D), where a-a', b-b', c-c' and d-d' are four different point pairs. Simulation accuracy is quantified by comparing the distance between each point pair in simulation and the corresponding distance in experiment.

Table 2
Simulation-experiment error for the lamp cover.

Point pairs	Experiment (mm)	Simulation (mm)	Error (%)
a-a'	76.54	72.42	5.38
b-b'	78.16	78.50	0.44
c-c'	64.95	66.01	1.63
d-d'	65.06	61.93	4.81

conventional 3D printing strategy shown in Fig. 15, the bottle holder structure will require extensive support structures and extended printing time. With the proposed workflow, designers can iterate through different designs and achieve the best fit for a specific bottle with significantly faster speed. Once the design is finalized, FDM printed 2D grid structure is printed and placed in an 80 °C water bath. The residual stress embedded in the material and the gravity will deform the grid into the programmed bottle holder shape.

The deformation result is shown in Fig. 16(A–C). The error between simulation and experiment is also calculated based on distance measurements. Three point pairs on the bottle holder are labeled in Fig. 16(D). Table 3 shows part of the distance results for each point pair and the corresponding error. More measurement results are provided in the supplementary material

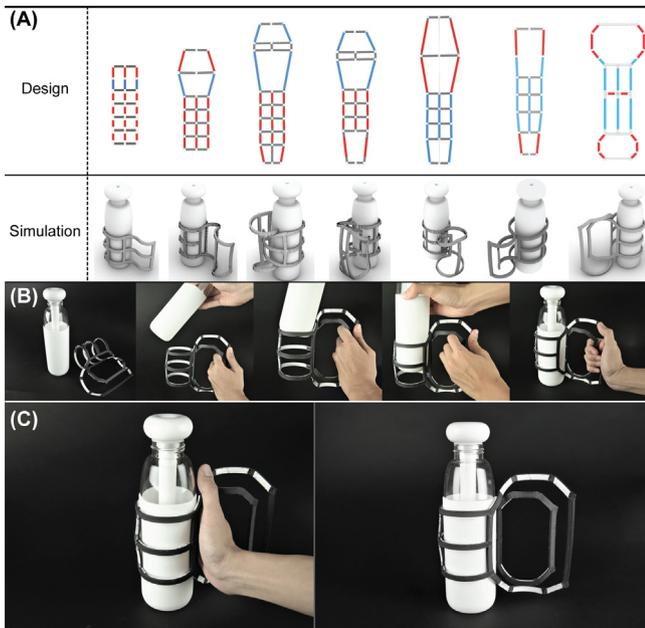


Fig. 15. Bottle holder design iteration. Results show the bottle holder can fit the bottle quite well after six iterations.

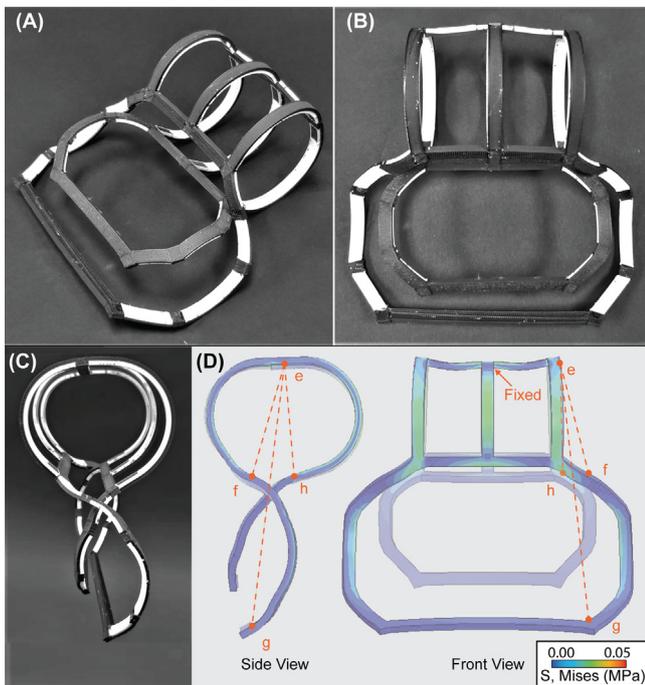


Fig. 16. Composite bottle holder. (A), (B) and (C) are different views of the final triggered result. (D) is the simulation result of the final configuration, where the labeled point pairs are used for the measurement and the accuracy calculation.

(Table S4). Based on the aforementioned measurement results, the confidence interval of the accuracy is (0.962, 0.986).

The shoe supporter is the third designed product. This application involves adapting sneakers for high-performance contexts such as hiking or climbing. This design is an exploration of how the composite material can interface with both the existing products and the human body. Employing the same strategy as the bottle holder, this design consists of two pieces – the top part and the bottom part. The design relies on connecting these two pieces

Table 3
Simulation-experiment error of the bottle holder.

Point pairs	Experiment (mm)	Simulation (mm)	Error (%)
e-f	71.35	70.68	0.94
e-g	161.45	160.31	0.43
e-h	67.58	64.98	3.85

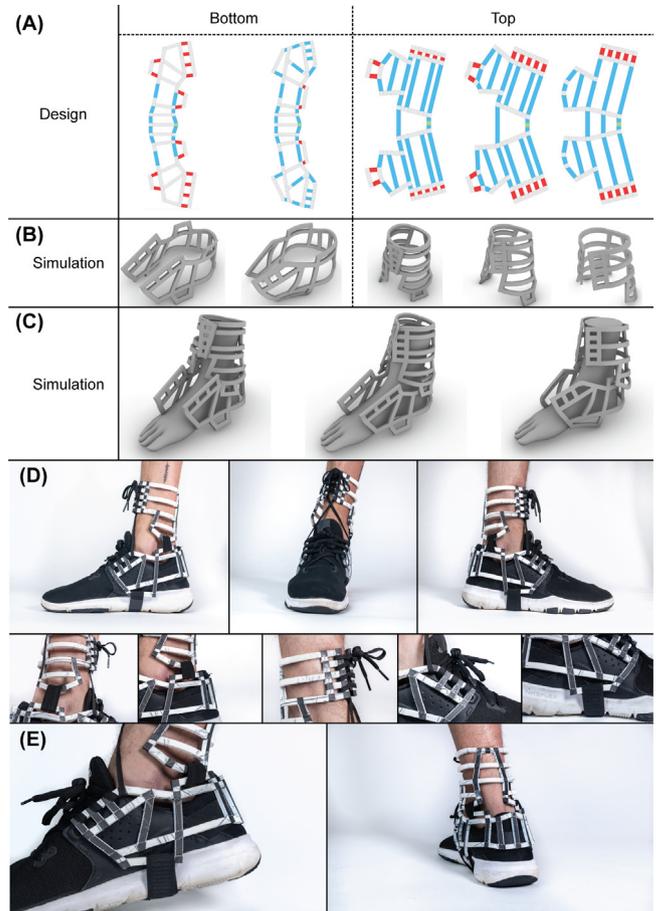


Fig. 17. The design iteration of the shoe supporter. The shape of the shoe supporter design is finalized to fit the shape of the foot by going through this iterative process.

attached to the sneaker and the ankle using elastic straps that anchor the bottom piece and strategically limit the movement of the top piece. In its active state, this design is meant to prevent injuries such as a rolled or sprained ankle, as well as to provide additional protection on the foot and the ankle in harsh conditions.

FEA results of both the top piece and the bottom piece are shown in Fig. 17. Experimental results of the finalized sample are shown in Fig. 18. The error is measured using the same strategy as the lamp cover and the bottle holder. Four point pairs are labeled in Fig. 18(B, E). Part of the measurement results and the calculated error are listed in Table 4. More measurement results are provided in the supplementary material (Table S4). Based on the aforementioned measurement results, the confidence interval of the accuracy is (0.969, 0.988).

7. Conclusion and future work

In this paper, we established a computational workflow that uses FEA to produce physically accurate results of the residual stress-induced morphing behaviors of mesh-like thermoplastic

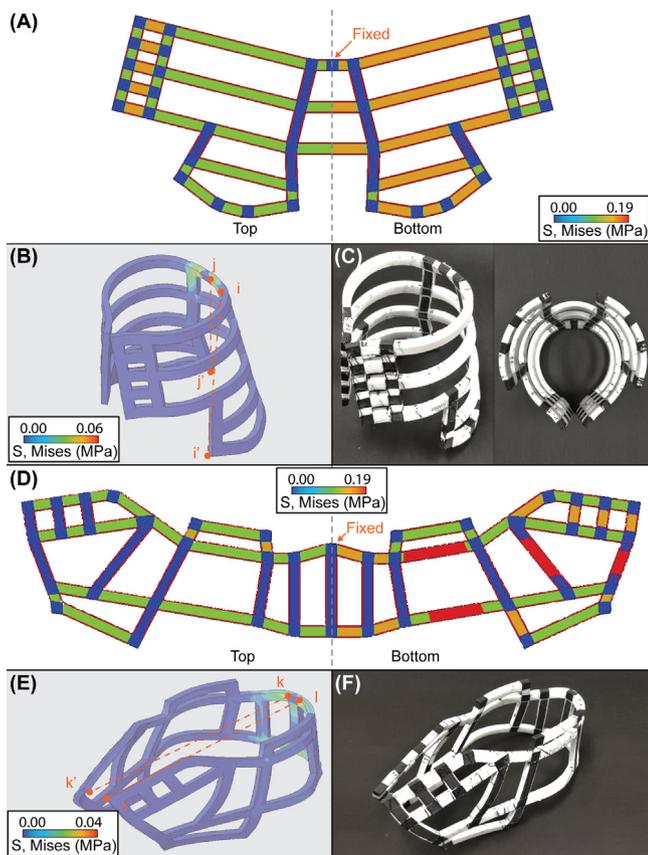


Fig. 18. The simulation and experimental results of the shoe supporter with the top piece (A–C) and the bottom piece (D–F). (A, D) show the initial condition from both the top view and the bottom view. (B, E) show the simulation result, where the labeled point pairs are used for the measurement and the accuracy calculation. (C, F) show the experimental results.

Table 4
Simulation-experiment error of the shoe support.

Point pairs	Experiment (mm)	Simulation (mm)	Error (%)
i-i'	89.74	92.25	2.80
j-j'	89.83	92.25	2.69
k-k'	160.57	157.02	2.21
l-l'	161.86	157.02	2.99

composite structures. To accurately conduct the simulation, experiments have been conducted to quantify the hyperelasticity and viscoelasticity of the thermoplastic component of FRC. These tests include uniaxial tensile and compression tests for the elastic component, DMA for the viscous component and uniaxial unloading–reloading with different strain levels for plastic deformation and the Mullins effect. This paper has also introduced the simulation sequence based on the triggering experiment and estimated the residual stress. The paper has validated the FEA modeling with an accurate matching between simulation and experiment for three creative designs.

One of the main limitations of the presented workflow is the high computational cost of FEA simulations, which makes it impossible for real-time, interactive design. Here we consider Graph Neural Network (GNN) as a potential model to speed up the simulation. It is worth noticing that the underlying topology of the grid structure can be abstracted to an undirected weight graph that is well compatible with GNN [40]. We are excited to look for appropriate abstraction and simplification by the aid of machine learning, and to explore a simulation method with both mechanical accuracy and computational efficiency.

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Appendix A. Supplementary data

Supplementary material related to this article can be found online at <https://doi.org/10.1016/j.cad.2020.102817>.

References

- [1] Ngo TD, Kashani A, Imbalzano G, Nguyen KTQ, Hui D. Additive manufacturing (3D printing): A review of materials, methods, applications and challenges. *Composites B* 2018;143:172–96.
- [2] Gao W, Zhang Y, Ramanujan D, Ramani K, Chen Y, Williams CB, et al. The status, challenges, and future of additive manufacturing in engineering. *Comput Aided Des* 2015;69:65–89.
- [3] Hu B, Duan X, Xing Z, Xu Z, Du C, Zhou H, et al. Improved design of fused deposition modeling equipment for 3D printing of high-performance PEEK parts. *Mech Mater* 2019;137:103139.
- [4] Singh R, Singh G, Singh J, Kumar R. Investigations for tensile, compressive and morphological properties of 3D printed functional prototypes of PLA-PEKK-HAP-CS. *J Thermoplast Compos Mater* 2019. <http://dx.doi.org/10.1177/0892705719870595>.
- [5] Roskies MG, Fang D, Abdallah M-N, Charbonneau AM, Cohen N, Jordan JO, et al. Three-dimensionally printed polyetheretherketone scaffolds with mesenchymal stem cells for the reconstruction of critical-sized mandibular defects. *Laryngoscope* 2017;127(11):E392–8.
- [6] Tian X, Liu T, Yang C, Wang Q, Li D. Interface and performance of 3D printed continuous carbon fiber reinforced PLA composites. *Composites A* 2016;88:198–205.
- [7] Tibbitts S. 4D Printing: multi-material shape change. *Archit Des* 2014;84(1):116–21.
- [8] Ge Q, Qi HJ, Dunn ML. Active materials by four-dimension printing. *Appl Phys Lett* 2013;103(13):131901.
- [9] Kwok TH, Wang CCL, Deng D, Zhang Y, Chen Y. Four-dimensional printing for freeform surfaces: design optimization of origami and kirigami structures. *J Mech Des* 2015;137(11):111413.
- [10] Deng D, Chen Y. Origami-based self-folding structure design and fabrication using projection based stereolithography. *J Mech Des* 2015;137(2):021701.
- [11] An B, Miyashita S, Tolley MT, Aukes DM, Meeker L, Demaine ED, et al. An end-to-end approach to making self-folded 3D surface shapes by uniform heating. In: *IEEE international conference on robotics and automation*. 2014. p. 1466–73.
- [12] Leung YS, Kwok TH, Li X, Yang Y, Wang CCL, Chen Y. Challenges and status on design and computation for emerging additive manufacturing technologies. *J Comput Inf Sci Eng* 2019;19(2):021013.
- [13] Davis D, Chen B, Dickey MD, Genzer J. Self-folding of thick polymer sheets using gradients of heat. *J Mech Robotics* 2016;8(3):031014.
- [14] Deng D, Yang Y, Chen Y, Lan X, Tice J. Accurately controlled sequential self-folding structures by polystyrene film. *Smart Mater Struct* 2017;26(8):085040.
- [15] Kwok TH, Chen Y. GDFE: Geometry-driven finite element for four-dimensional printing. *J Manuf Sci Eng* 2017;139(11):111006.
- [16] Bakarich SE, Gorkin R, Panhuis MH, Spinks GM. 4D Printing with mechanically robust, thermally actuating hydrogels. *Macromol Rapid Commun* 2015;36(12):1211–7.
- [17] Grijpma DW, Pennings AJ. (Co) polymers of L-lactide, 2. mechanical properties. *Macromol Chem Phys* 1994;195(5):1649–63.
- [18] Maharana T, Mohanty B, Negi YS. Melt–solid polycondensation of lactic acid and its biodegradability. *Prog Polym Sci* 2009;34(1):99–124.
- [19] Langer RR. Drug delivery and targeting. *Nature* 1998;392:5–10.

- [20] Soares JS, Moore JE, Rajagopal KR. Constitutive framework for biodegradable polymers with applications to biodegradable stents. *ASAIO J* 2008;54(3):295–301.
- [21] Hayman D, Bergerson C, Miller S, Moreno M, Moore JE. The effect of static and dynamic loading on degradation of PLLA stent fibers. *J Biomech Eng* 2014;136(8):081006.
- [22] Khan KA, El-Sayed T. A phenomenological constitutive model for the nonlinear viscoelastic responses of biodegradable polymers. *Acta Mech* 2013;224(2):287–305.
- [23] Söntjens SHM, Engels TAP, Smit TH, Govaert LE. Time-dependent failure of amorphous poly-D, L-lactide: Influence of molecular weight. *J Mech Behav Biomed Mater* 2012;13:69–77.
- [24] Eswaran SK, Kelley JA, Bergstrom JS, Giddings VL. Material modeling of polylactide. In: *SIMULIA customer conference*. Barcelona, Spain. May. 2011. p. 17–9.
- [25] Bodaghi M, Noroozi R, Zolfagharian A, Fotouhi M, Norouzi S. 4D Printing self-morphing structures. *Materials* 2019;12(8):1353.
- [26] Diani J, Fayolle B, Gilormini P. A review on the Mullins effect. *Eur Polym J* 2009;45(3):601–12.
- [27] Ionov L. Soft microorigami: Self-folding polymer films. *Soft Matter* 2011;7(15):6786–91.
- [28] Shim TS, Kim SH, Heo CJ, Jeon HC, Yang SM. Controlled origami folding of hydrogel bilayers with sustained reversibility for robust microcarriers. *Angew Chem Int Ed* 2012;51(6):1420–3.
- [29] Stoychev G, Turcaud S, Dunlop JWC, Ionov L. Hierarchical multi-step folding of polymer bilayers. *Adv Funct Mater* 2013;23(18):2295–300.
- [30] Jones DRH, Ashby MF. *Engineering materials 2: An introduction to microstructures, processing and design*. Elsevier; 2005.
- [31] Osborne MR. On shooting methods for boundary value problems. *J Math Anal Appl* 1969;27(2):417–33.
- [32] Eilers PHC, Marx BD. Flexible smoothing with B-splines and penalties. *Statist Sci* 1996;11(2):89–102.
- [33] Yu Y, Zhang YJ, Takizawa K, Tezduyar TE, Sasaki T. Anatomically realistic lumen motion representation in patient-specific space-time isogeometric flow analysis of coronary arteries with time-dependent medical-image data. *Comput Mech* 2019. <http://dx.doi.org/10.1007/s00466-019-01774-4>.
- [34] Lubliner J. *Plasticity theory*. Courier Corporation; 2008.
- [35] Systèmes D. *Simulia user assistance*. Dassault Systèmes 2019.
- [36] Marlow RS. A general first-invariant hyperelastic constitutive model. *Const Model Rubber* 2003;157–60.
- [37] Ogden RW, Roxburgh DG. A pseudo-elastic model for the mullins effect in filled rubber. *Proc R Soc Lond Ser A Math Phys Eng Sci* 1999;455(1988):2861–77.
- [38] Govindarajan SM, Hurtado JA, Mars WV. Simulation of Mullins effect and permanent set in filled elastomers using multiplicative decomposition. In: *Constitutive models for rubber - proceedings*, vol. 5, Balkema; 2008, p. 249.
- [39] Yeoh OH. Some forms of the strain energy function for rubber. *Rubber Chem Technol* 1993;66(5):754–71.
- [40] Zhou J, Cui G, Zhang Z, Yang C, Liu Z, Sun M. Graph neural networks: A review of methods and applications. 2018, [arXiv:1812.08434](https://arxiv.org/abs/1812.08434).