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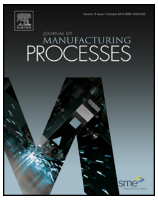

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Manufacturing of smart composites with hyperelastic property gradients and shape memory using fused deposition[☆]

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ABSTRACT

In this paper, we demonstrate two studies where fused-deposition modelling (FDM) is used to fabricate composites with (1) controlled hyperelastic property gradients and (2) shape-memory behaviour. In the first study, we first fabricate thermoplastic elastomer scaffolds consisting of freely suspending fibers through FDM and then encapsulate them with soft silicone elastomers. We first present our studies on how the scaffold geometry is correlated with the printing speed and flow rate. Next, through tensile testing, we demonstrate the capability of the method in generating structures with (1) different hyperelastic properties through scaffold design and printing parameter control and (2) controlled spatial gradients of such properties. In the second study, we use multi-material FDM to manufacture composite structures consisting of a thermoplastic elastomer shell and polycaprolactone (PCL) core. Owing to the lower melting point and higher room temperature modulus of the PCL, these composites exhibit shape memory behaviour if subjected to thermal cycling between the room temperature and the melting point of the PCL. We evaluate the geometry and temperature dependence of this behaviour. We also demonstrate the reprogrammability of the memorized shape by introducing a silicone encapsulation for the composites.

1. Introduction

Additive manufacturing (AM) is emerging as a key technology in manufacturing, owing to its unique capabilities such as manufacturing of functionally graded structures and geometrically complex lattice structures with light weight and high stiffness. Recently growing interest in AM led to the realization of these capabilities for a wide range of material systems including plastics, metals and ceramics [1]. Among these materials, plastics are by far the most established within the AM context as they have been widely used for rapid prototyping which constituted the initial application domain for these methods. Despite this strong background, AM with plastics has been mostly stagnant in the rapid prototyping domain since these processes couldn't provide a significant advantage over conventional methods due to limited range of applicable materials, and their generally slow nature. Recent emergence of novel technologies such as customized wearable biomedical devices, soft and flexible electronics and robotics renewed the interest in plastic AM as these technologies greatly benefit from multi-material structures with customizable geometries and well controlled gradients of part properties such as mechanical compliance [2]. As such, there is a growing need for research on AM with plastics involving novel material systems, their design and processing.

Commonly used AM methods for plastics include fused deposition modelling (FDM)[3], vat photopolymerization [4], powder bed fusion[5] and material jetting [6]. Each of these methods offer unique advantages and disadvantages. Among these, vat photopolymerization can be tuned to exhibit extremely high throughput [7]. Material jetting methods are highly versatile as they are capable of easily creating multi-material systems with continuous property gradients [8] or smart functionalities [9], [10]. The main drawbacks of vat photopolymerization and material jetting methods are (1) material limitations as they can only process photocurable polymers having specific ranges of precursor viscosity and surface tension[11], [12] which are mostly proprietary, and (2) the high cost of printers and materials. FDM distinguishes itself from the other methods as the most facile and low cost approach. The process is generally slow and manufactured parts exhibit high levels of anisotropy[13]. However, a number of properties render FDM as a highly flexible AM method. Firstly, it allows for processing of many different material systems including thermoplastic polymers blended with functional agents, such as metal particles [14], conductive carbon species including carbon nanotubes[15],

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graphene [16] etc. Secondly, it offers a straight forward multi-material configuration where dissimilar materials can be extruded from multiple different nozzles and can be composited in complex configurations.

These unique aspects of FDM have been attracting significant attention of both low-level users and researchers towards realizing novel functional products. There is substantial room for research on design and synthesis of new functional material systems that can be processed with FDM, understanding of material property-FDM process parameters-manufactured product properties for different materials of interest and new functional composites enabled by multi-material capabilities of FDM. A particularly unexplored area for FDM technologies is the manufacturing of smart composites possessing property gradation or responding to various stimuli. AM of such components through high-cost material jetting systems have been demonstrated[8], [9], [17]. Realization of such functionalities using the low-cost FDM method could substantially broaden the impact of AM in technologies including soft robotics, biomedical devices, flexible electronics etc, making such technologies available to a wider population.

In this paper, we present two case studies towards addressing this need. In these studies, we use a commercially available FDM printer and materials to realize (1) parts with spatially controllable hyperelastic property gradients and (2) composite structures with tunable shape memory behaviour. The first study involves manufacturing of multilayer grid-type scaffolds using thermoplastic elastomers and using them as reinforcement inside soft silicone rubbers to form composite elastomers as shown in Fig. 1(a). We can tune and spatially control the mechanical properties of such composites by controlling the scaffold design and fiber geometry. The parts generated this way possess the material properties that are required for stretchable, wearable electronic devices, where skin soft characteristics are sought after for sensor and actuator elements that conform to the body and low compliance is needed where such electronics are interfaced with rigid electronics. This method provides a substantially more facile alternative to other methods demonstrated in the literature [18–20]. In the second study, we print composites of the same thermoplastic elastomer and a low melting point polycaprolactone (PCL), a commonly used polymer for smart structures [21]. Here, produced part possesses shape memory behaviour, owing to the large melting point differential of the two polymers and elasticity of the elastomer. We demonstrate that this behaviour can be tuned by varying the volume percentage each material in core-shell configurations. We also show that by encapsulating such parts in silicone rubbers, reprogrammable smart structures can be obtained.

The rest of this paper is organized as follows: Firstly we introduce the materials and the equipment used in these studies. Secondly, focusing on the first study, (1) we present the results on thermoplastic elastomer scaffold fabrication and (2) we evaluate the mechanical properties of various hyperelastic composites fabricated using various scaffold designs and FDM parameters. Finally, we present the results on the second study where (1) we fabricate PCL-elastomer composites with varying material volume ratios and evaluate their shape memory behaviour, (2) we encapsulate the fabricated composites in silicone rubber to demonstrate the possibility of reprogramming their “memorized” default shape.

2. Materials and methods

2.1. Polymers used

The properties of the two commercially available thermoplastics used in these studies are listed in Table 1. The thermoplastic elastomer is a commercially available urethane-based FDM material (Recreus FilaFlex) which is one of the most flexible 3D printing materials with a room temperature tensile modulus of 48 MPa and an elongation at break of 700%. The PCL (3D4Makers PCL 99) has a melting point of 60 °C. Both materials are obtained in the filament form with 1.75 mm diameter.

The thermoplastic elastomer scaffolds and shape memory composites are encapsulated in platinum cured, thermoset silicone elastomers (Ecoflex 00-10, Slygard 184, respectively). Both silicone materials are prepared by mixing of the base polymer precursor with cross-linking agents followed by degassing and thermal curing around the 3D printed structures.

2.2. FDM-based printing

A commercially available 3D-printer (Felix Pro1) is used in this study. This printer is instrumented with a dual extruder with nozzles of 350 μm inner diameter. The parts are designed through CAD and Felix builder software is used to configure the prints, which allows the precise prescription of the flow rates during printing and using different materials for the contour and infill of the printed parts. The printer also features cooling fan which enables rapid solidification of the extruded materials and enables formation of free standing filamentary structures utilized during this study.

2.3. Characterization of the elastomer composites

The geometry of the 3D printed suspending FilaFlex filaments are characterized using a stereo microscope (FireFly GT700) where the diameter and the sag of the filaments are measured. The mechanical properties of the FilaFlex-Ecoflex composites are characterized by a tensile testing system (Mark-10 ESM 303). Dogbone type tensile specimens are prepared according to the ASTM D638-14 standard for rubbers. To this end, the printed scaffold were placed in an acrylic container with an outline of the dog-bone samples, having a depth equal to the desired sample thickness. The container is then filled with the liquid elastomer precursor encapsulating the scaffold. The cured composite was then demolded from the container.

Tensile testing is performed at a speed of 100 mm/min. The local strains on the specimens are measured through image processing using an open-source software tool (Ncorr). To this end, first, the test samples were painted using black and white spray paint to facilitate feature tracking-based image processing and then the entire testing process was recorded using a camera.

2.4. Activation and characterization of the shape-memory composites

The 3D printed FilaFlex-PCL and FilaFlex-PCL-Slygard 184 shape-memory composites are thermally activated in an oven (Thermo Scientific Lindberg/Blue Vacuum Oven). The shape measurements have been conducted with a vernier caliper (Mitutoyo 500-195-30).

To encapsulate the printed composites with PDMS, the composite was placed in a petri-dish which was then filled with the PDMS precursor (10:1 mixing of Part A and B). The polymer was cured at 70 °C for 5 h and then the encapsulated composite was cut out of the cured polymer using a knife.

3. Results

3.1. Elastomeric composites with tunable hyperelastic properties

The main idea behind this application is illustrated in Fig 1(a). Here, a thermoplastic elastomer (FilaFlex) scaffold is 3D printed through FDM and then encapsulated in a thermoset elastomer

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