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3-D printed composites with ultrasonically arranged complex microstructure

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Abstract

This paper demonstrates the efficacy of implementing ultrasonic manipulation within a modified form of stereolithographic 3D printing to form complex microstructures in printed components. Currently 3D printed components are limited both in terms of structural performance and specialised functionality. This study aims to demonstrate a novel method for 3D printing composite materials, by arranging microparticles suspended within a photocurable resin. The resin is selectively cured by a 3-axis gantry-mounted 405nm laser. Ultrasonic forces are used to arrange the microfibres into predetermined patterns within the resin, with unidirectional microfibre alignment and a hexagonal lattice structure demonstrated. An example of dynamic microstructure variation within a single print layer is also presented.

Introduction

With the rapid development of various affordable and approachable 3-dimensional printing techniques, there is an increasing drive towards producing high quality components with material properties comparable to those of traditional manufacturing techniques. Due to its low waste, rapid prototyping and ease of use, 3D printing is a beneficial future alternative for conventional manufacturing techniques. 3D printing processes enable multiple design iterations to be completed in a day, where in the past this may have taken weeks or months employing traditional subtractive manufacturing methodologies. More advanced recent technologies (e.g selective laser sintering and plastic jet printing) enable the production of both prototypes and final components, with a wide variety of potential applications (e.g prosthetic limbs, biomedical implants, aeroplane components).^{15,18} An increasing number of solutions for multimaterial printing have become available, allowing for additional functionality in 3D printed structures.^{7,13} The most prevalent form of 3D printing currently is fused filament fabrication (FFF), which builds components layer by layer by extruding a thermoplastic filament through a heated nozzle. Fused filament fabrication is cheaply implemented, reliable and is compatible with a wide range of materials.

As with the majority of 3D printing techniques, however, the structural properties of FFF manufactured components are relatively low, and as such FFF is not appropriate for high performance applications.¹ It has been shown that introducing nano or microscale particles/fibres into the thermoplastic feedstock is a viable method for improving the mechanical performance of the printed polymer and can provide additional functionality.^{4,23} In order to use polymeric solutions in high performance applications, the typical approach is to use continuous fibres (glass, Kevlar or carbon) to improve the mechanical performance by maximising the fibre-matrix stress transfer. The use of continuous fibres in 3D printing constrains the tool path used, as the fibres must be printed along the primary stress path for optimal performance, and this in turn adversely affects the quality of the finished component. A viable alternative is the use of short discontinuous fibres, which have most notably been employed in FFF printing by including the fibres within the feedstock filament.^{4,23} In this process the direction of the fibres axis follows the toolpath closely due to shear flow at the extruder nozzle. This again requires complicated toolpath constraints to maximise the effect of including fibrous reinforcement, and is incapable of producing any reinforcement in the z-axis across neighbouring layers.

An improved system would allow complete control over the internal microstructure of a printed component with either microfibrinous reinforcement or functional microparticles. In order to achieve this an alternative to FFF is required due to the difficulty of manipulating particles within a viscous material being extruded at high temperatures. The design shown

in this work combines the electromechanical aspects of FFF printing with the curing process of stereolithographic (SLA) printing. This is advantageous as SLA printing utilises a stationary reservoir of photocurable resin, which is selectively cured by a light source (commonly a projector or laser).

In order to include microstructure within SLA printed components, a process is required which can manipulate microparticles within the resin tank prior to curing. There are a number of existing methods capable of manipulating suspended particles, which make use of magnetic, electric and acoustic fields, as well as flow induced alignment techniques.^{6,17,20} Electric and magnetic fields are generally only capable of producing nematic phases and would not allow complex structures to be realised. Magnetic field alignment also typically requires that the microparticles be pre-treated with superparamagnetic nanoparticles to increase their response to an incident magnetic field, which interfere with the intended effect of the microparticles. Due to the dynamic nature of flow induced fibre alignment, it is not compatible with SLA printing, which requires a static resin tank. Acoustophoresis has been shown to be effective as a method for manipulating and arranging particles suspended in a host fluid.²¹ Opposing transducers produce counter-propagating plane waves in the fluid medium, which generates a standing wave field with steep acoustic pressure gradients. This leads to acoustic radiation forces being exerted on the suspended particles, with the amplitude of the force dependent on the acoustic contrast factor between the fluid medium and the particles. In general particles with a greater density than the fluid are manipulated to acoustic pressure nodes, and those with lower density towards pressure antinodes.^{5,10} With a simple 1-dimensional pressure field, the neighbouring pressure nodal planes (and therefore trap locations) are separated by $\frac{\lambda}{2}$ where λ is the wavelength of the wave in the fluid medium.^{11,22}

Acoustophoresis has proven useful in a number of fields, including biological applications such as cell sorting and building scaffolds for tissue engineering.^{8,12} Ultrasonic manipulation is (relative to optical tweezers) easily upscaled and less damaging to biological cells.² Using ultrasonic manipulation to arrange particles within a resin system has been demonstrated in existing publications,^{3,9,16,19} and mechanical characterisation of such parts demonstrated that structural anisotropy was achieved (8% stiffness and 43% strength anisotropy along the two principal directions).²⁰ More recently, 3D printing of components with unidirectional ultrasonically aligned microfibrils has been demonstrated, as well as dynamically realigned orthogonal fibres within a single printed layer.¹⁴ This method effectively separates the printing and microstructural formation processes, enabling greater freedom over the microstructures being formed. This work extends this concept to more complex structures using glass microfibrils.

Experimental Method

Printer

The printing stage used was built from an open source design (Prusa Mendel Iteration 3), with the thermoplastic extrusion head removed and replaced with a 50mW violet (405nm) laser module. The control board is an Arduino Mega2560 with an Ultimaker Shield v1.5.7 with Marlin firmware. The laser diode was fitted into a module containing a screw adjustable 6mm diameter lens, allowing for variable focus of the laser. A 1mm aperture is used at the end of the laser module to ensure a beam of uniform radial intensity. The module was oriented such that the laser beam was aimed at the print bed. The ultrasonic rig was placed on the print bed under the laser module as depicted in Figure 1.

Ultrasonic Rig

The design of the ultrasonic rig consists of a laser cut polymethyl methacrylate (PMMA) frame glued to a standard microscope slide (76mm by 26mm by 1.2mm). The frame has two small outer cavities, which house the ultrasonic transducers, and a larger central cavity where the resin/microparticle mixture was placed. The microfibrils used are (Lanxess MF7904, length 50 μm , diameter 14 μm). The transducers are piezoceramic plates made of lead zirconium titanate (Noliac NCE 51), and are held in place by small springs (Lee Spring CIM 040EG01S). The use of separate cavities for the transducers and resin allows for the transducers to be reused after printing and negates the risk of cured resin adhering to the transducer faces. The cavities housing the transducers are filled with water to create a heatsink, allowing the transducers to be driven at higher voltages without damaging the ultrasonic rig. The central printing cavity was filled with a mixture of Spot-A LV low viscosity photocurable resin and microfibrils. Two different rig designs were used, one containing a single opposing pair of transducers with dimensions 35mm by 2mm by 0.975mm as shown in Figure 1(a) and another with 3 opposing

Figure 1: Schematic representation of ultrasonic rigs used to pattern microparticles within photocurable resin.

pairs of transducers with dimensions 15mm by 2mm by 0.975mm as shown in Figure 1(b). Both sets of transducers have a first through-thickness resonance of 2.35MHz.

With a single pair of opposing transducers, a simple standing wavefield is produced by driving the two transducers with the same phase, frequency and voltage, which aligns the suspended particles along nodal planes parallel to the transducer faces.

Using 3 pairs of opposing transducers, more complicated ultrasonic fields can be generated, allowing for the formation of complex microstructures using particles of different dimensions and aspect ratios. In particular, hexagonal lattice arrangements of dense particles can be realised with a six transducer array, with the potential to trap low density particles at pressure antinodes in the centre of each hexagonal cell. It is also possible to switch specific pairs on and off, which allows for a greater variety of unidirectional orientation angles of particles.

Printing Process

The print movements are defined by standard G-code commands, which are generated automatically by a Matlab script from an input DXF sketch file. As with most printing processes, an outline of the shape is cured prior to filling in the rest of the shape to improve the surface finish of the printed part. Initial test prints determined that a speed of 20mm/s for the laser carriage was ideal for adequate curing of a 1mm thick sample of resin within the central cavity of the ultrasonic rig. To incorporate the laser module into a standard 3D printer, the module was powered by an auxiliary fan output on the printer control board, which was used to switch the laser. A single I-shape was used for all print samples to demonstrate that parts were printed and not moulded from the cavity shape, with the dimensions of the printed part shown in Figure 3. Each sample takes approximately 20 seconds to print. A schematic representation of both the printer and ultrasonic rig are shown in Figure 2.

Results and Discussion

Dual Array

As aforementioned, with two opposing transducers microparticles are aligned along nodal planes. With a driving frequency of 2.35MHz, nodal planes are separated by $\sim 300\mu\text{m}$ as shown in Figures 4 and 5. Figure 5 also demonstrates that the fibres extended to the edge of the printed component and maintain alignment to this edge. With a speed of sound in the resin

a)

b)

Figure 2: Schematic representation of the printer and the ultrasonic manipulation device. (a) Laser module aimed at the resin cavity in the ultrasonic rig (b) Cross-section of two-transducer ultrasonic device with labelled regions. Here P=PMMA, W=Water, PZT= lead zirconate titanate transducers, R= Spot-A LV low viscosity photocurable resin.

$c = 1400\text{ms}^{-1}$, the theoretical spacing of nodal planes at 2.35MHz is $\Delta x = \frac{\lambda}{2} = 297\mu\text{m}$. A driving voltage of $75V_{pp}$ is used across both transducers.

It can be seen that the fibres are not perfectly orthogonal to the edge of the printed component, which is a result of slight imperfections in the ultrasonic rig such as transducer misalignment, as well as misalignment of the ultrasonic rig with the axes of the gantry. Figure 6 is an SEM image of the fracture surface of an I-shape component split perpendicular to the fibre orientation, showing the bundle of fibres end-on, extending $\sim 50\mu\text{m}$ into the thickness of the print layer. The depth and width of the fibre bundle is dependent on the fibre volume fraction in the resin.

Hexagonal Array

Using a 6 transducer array, a hexagonal pattern of pressure nodes are formed, causing the dense glass microfibres to form a hexagonal lattice structure, as shown in Figure 7. All transducers are again driven with a frequency of 2.35MHz , with a voltage of $50V_{pp}$. A lower voltage is used than in the two-transducer device due to the smaller resin cavity size. Figures 7, 8 and 9 are all from a single sample, wherein initially all six transducers were driven to produce a hexagonal microstructure, as shown in Figure 7.

As aforementioned, it is possible to switch chosen pairs of transducers off with the hexagonal array device to produce various microstructures. An example of such a structure is shown in Figure 9, where a single opposing pair of transducers was deactivated mid-layer, giving a unidirectional alignment of fibres with pressure antinodes (seen as microfibre spacing) along each trapping plane. Due to larger effects from streaming, the nodal planes themselves are less clearly defined than in the simple unidirectional example. This is a problem that could be solved by using more precisely manufactured acoustic rigs and including matching impedance layers on the back faces of the transducers. Figure 8 represents shows the transition between hexagonal lattice arrangement of the microfibres and spaced unidirectional alignment which occurs

Figure 3: Representation of the print shape used for all samples in this study.

Figure 4: Microscopy image of unidirectionally aligned fibres within a printed sample, with edge of part shown at top.

Figure 5: Microscopy image of unidirectionally aligned fibres within a printed sample, with edge of part shown at top.

Figure 6: SEM image of fracture surface of sample with unidirectionally aligned fibres running perpendicular to surface.

when dynamically varying the microstructure. As the reorientation process is not instant, regions of the printed component contain a combination of the initial and final microfibre arrangements. This is easily avoided by pausing the print process whilst altering the acoustic field.

Concluding Remarks

This work has demonstrated the first example of ultrasonically arranged complex microstructures within a 3D printed component. This is achieved through the use of a novel modified SLA 3D printing technique in conjunction with an ultrasonic manipulation device. Future testing is required to determine the effects of the microparticle inclusions on the structural properties of the printed material. Previous work has shown the effect of including unidirectionally aligned glass microfibres on the mechanical properties of a moulded resin sample.²⁰ Fibres of varying length scales were independently manipulated within a single part, and previous work has indicated that dynamic realignment of fibres is possible mid-layer. This real time control over microparticle inclusions enables 3D printing of functional composite materials without the necessity for complicated toolpaths which compromise print quality. A new generation of 3D printed functional materials

Figure 7: Optical microscopy image of glass microfibres arranged in a hexagonal lattice structure.

Figure 8: Optical microscopy image of transition region between unidirectionally aligned microfibres and hexagonally arranged lattice structure.

Figure 9: Optical microscopy image of unidirectionally aligned microfibres with spacings along trap lines.

may be produced by substituting the particles used in this study for others such as resin-filled capsules for self healing purposes, electrically conducting materials for structural computers or piezoelectric fibres for energy harvesting.

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