# Additively Manufactured Polymeric Metamaterials

#### Subjects: Mechanics

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Metamaterials are architected cellular materials, also known as lattice materials, that are inspired by nature or human engineering intuition and provide multifunctional attributes that cannot be achieved by conventional polymeric materials and composites. There has been an increasing interest in the design, fabrication, and testing of polymeric metamaterials due to the recent advances in digital design methods, additive manufacturing techniques, and machine learning algorithms.

architected materials

lattices

polymeric composites

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## 1. Introduction

The persistent quest of scientists for candidate materials and designs is attributed to the advent of cutting-edge technologies. With advances in material synthesis and additive manufacturing techniques, the fabrication of these materials and designs has become conceivable. In recent years, metamaterials emerged as one of the leading-edge technologies enabling the production of multi-functional structures on macro- and nano-scales. Generally, metamaterials are defined as artificially engineered materials mimicking nature-based architectures synthesizing extreme material properties that are rarely observed in bulk material form. Metamaterials, also known as lattice, architected or cellular materials/structures, are multi-functional materials executing numerous functions by virtue of tailored electromagnetic <sup>[1]</sup>, optical <sup>[2]</sup>, acoustic <sup>[3]</sup>, thermal <sup>[4]</sup>, and mechanical properties <sup>[5]</sup> for diversified purposes.

The recent advancements in additive manufacturing and material synthesis allowed the integration of polymeric metamaterials in real-world applications. For example, Veerabagu et al. <sup>[6]</sup> reviewed the progress of polymeric auxetic metamaterials utilized in tissue engineering and medical devices, such as stents and sensors. Fan et al. <sup>[7]</sup> characterized different types of polymeric metamaterials according to their electromagnetic and acoustic features, for example, electromagnetic metamaterials made of photo-curable resin are functional in terms of electromagnetic cloaking, while acoustic and thermal metamaterial made of ABS and epoxy resin are functional in terms of acoustic absorption and ultralow thermal conductivity, respectively. Furthermore, Al Mesmari et al. <sup>[8]</sup> characterized different types of mechanical metamaterials involved in impact absorption and load-bearing applications. However, there is still an apparent gap in categorizing the current polymeric metamaterials in terms of their mechanical properties, whether it is due to the various topologies, printing techniques used to manufacture the parts, the polymeric materials used, etc.

## 2. Design of Polymeric Metamaterials

Metamaterials are usually arranged in a periodic network of structural elements or repeating patterns. This network of lattices exists on a wide range of scales, from the nanoscale to the macroscale, and is now a candidates for design in additive manufacturing. Importantly, the metamaterials' effective working properties, such as their mechanical capabilities, can be modified by engineering the macro units to form specific microstructures with desired functional responses. **Figure 1** illustrates examples for each major and subclass of lattice materials whose architectural multifunctional properties have been extensively explored in the literature <sup>[9][10][11][12]</sup>. It is worth noting that some lattices use the same recurring unit cell through the entire latticing bounds, and are referred to as periodic lattices (**Figure 1**b). The alternative form is for the cells to be randomly connected across the domain and are referred to as stochastic lattices (**Figure 1**a).





## 3. Fabrication of Polymeric Metamaterials

Additive manufacturing (AM) is the commonly used fabrication technique to produce polymeric metamaterials. The base material could be supplied in many forms, such as photosensitive resins, filaments, viscous polymer inks, and thermoplastic powders. Different techniques of AM are utilized to produce polymeric metamaterials, where the most common techniques include binder jetting (BJ), sheet lamination (SL), vat photopolymerization (VP), material extrusion (ME), powder bed fusion (PBF) and material jetting (MJ) <sup>[13]</sup>. Each one of the aforementioned AM techniques is unique in terms of working principle and base material form <sup>[13][14]</sup>. **Figure 2** describes the mechanism of the aforementioned AM techniques.



**Figure 2.** Illustration of (a) Vat photopolymerization, (b) Material jetting, (c) Material extrusion, (d) Binder jetting and (e) Sheet lamination polymer additive manufacturing processes.

VP is a widely used AM process for manufacturing polymeric materials due to its well-known high resolution and part accuracy. As shown in **Figure 2**a, VP makes use of radiation (i.e., visible light and ultraviolet (UV) radiation) to construct the digital specimen through selectively polymerizing photosensitive liquid resins in a vat (i.e., liquid resin) <sup>[15][16]</sup>. The conception of stereolithography (SLA) VP technology has fueled the large-scale manufacturing of parts with higher resolutions <sup>[15][16]</sup>.

Two-photon photopolymerization (2PP) is a direct laser printing technique which is another category of photopolymerization-based additive manufacturing technique known for its ability to print parts at a much higher resolution than SLA <sup>[13][17]</sup>. 2PP employs the two-photon absorption (TPA) concept, where two photons drive the electronic transitions rather than a single photon with visible or UV-based VP during the photopolymerization process <sup>[18]</sup>. TPA is derived from using a high photon-density laser beam, for example, a pulsed femtosecond laser. The part is built layer-wise and across the workspace by changing the focus of the laser in-line with the part's geometrical configuration within the resin.

MJ is another polymer additive manufacturing technique whose process, as demonstrated in **Figure 2**b, is comparable to conventional 2D inkjet fabrication, where a liquid material is either deposited onto the workspace from the inkjet print-heads through a continuous process, or deposited-on-demand during manufacturing <sup>[13]</sup>. Afterward, the layer-wise deposited liquid material is solidified through photopolymerization. Due to the ability to use multiple printheads, the MJ technique is widely used for printing composites or multi-material components from a wide choice of materials such as resins, reactive materials, thermoplastics, wax, etc. <sup>[14]</sup>.

Contrary to the above-discussed additive manufacturing techniques, ME, also referred to as fused filament fabrication (FFF) or fused deposition modeling (FDM), involves a progressive extrusion of viscous inks, polymer pellets or filaments through an orifice at elevated temperatures <sup>[19]</sup> onto the build platform as shown in **Figure 2**c, after which they solidify through cooling. FDM is the most commercially-viable additive manufacturing process, where thermoplastic filaments are used as feedstock. However, with the incorporation of extrusion-based systems in the recent development of the FDM process, pellets can be used as feedstocks which facilitate the processing of wider types of thermoplastics <sup>[20]</sup>.

BJ is a powder-based process that employs an inkjet printhead to selectively fuse the powder materials (e.g., metals, and ceramics, while polymers are mostly used as binders <sup>[21]</sup>) in a powder bed by dropping liquid binders while mimicking the shape of the 3D object as shown in **Figure 2**d. Due to the non-reliance of the BJ process on the heat source, much larger parts can be built through this process at a relatively low cost. Nevertheless, the absence of melting or sintering processes gives rise to relatively weak or flimsy and porous parts made by BJ, hence, the need for post-processing (typically through liquid infiltration and thermal sintering) of the fabricated part <sup>[13]</sup>. Unlike the BJ process, in the PBF technique, a powdered material is selectively fused using a heat source (e.g., laser or electron beam) on a powder bed. For example, the selective laser melting (SLM) method utilizes a laser source to completely fuse metal particles, thus, producing metallic components with higher mechanical resistance <sup>[8]</sup>. As the PBF technique involves recurring heating and cooling cycles during the solidification of deposited layers, the printed components experience the accumulation of unfavorable residual stresses.

With the SL process, 3D objects are printed by stacking and laminating feedstocks, which include papers, ceramic tapes, thermoplastic foils, and metallic and woven fiber composite sheets in the form of thin rolled sheets or foils. SL is a cost-effective process and can be used for fabricating relatively larger structures. The typical SL process is described in **Figure 2**e. Laminated object manufacturing (LOM) is one type of SL process that employs both subtractive and additive approaches to form 3D objects through the bonding of sheets. Inspired by this process, other cutting construction principles have been designed such as laser cutting, water jet cutting and computer numerical control (CNC) milling, and bonding types such as ultrasonic welding thermal bonding and adhesive bonding <sup>[13]</sup>. LOM is not widely used for commercial manufacturing due to issues related to the inception of internal cavities derived from the cutting process which contributes to unacceptable material waste <sup>[13]</sup>.

## 3.1. Optimizing the Fabrication Procedure of Polymeric Metamaterials

Geometrical imperfections and defects generated due to improper manufacturing deteriorate the mechanical performance of additively manufactured metamaterials <sup>[22]</sup>. The topic of enhancing the manufacturing fidelity of lattice materials has become of interest to several industries. For example, Holmes et al. <sup>[23]</sup> demonstrated the capability of Gyroid structures made of flexible thermoplastic polyurethane (TPU) to replace commercial soft padding foams to mitigate clinical conditions like pressure ulcers. Along the same lines, the manufacturing fidelity of lattice materials includes examining the deviations between the as-built (i.e., the printed) and the as-designed (i.e., CAD design) parts in terms of mass and geometry. Commonly, micro X-ray computed tomography (micro CT) is utilized to construct a 3D model of an as-built part to compare its manufacturing fidelity to an as-designed part. In fact, optimizing the process parameters is one of the key factors that improves manufacturing fidelity and mitigates the geometrical defects associated with developing lattice materials. Sala et al. <sup>[24]</sup> addressed the manufacturing complexities related to printing flexible honeycomb, Schwartz Primitive, and Gyroid lattices made of TPU using FDM. Certain process parameters were tuned to probe the optimum printing combination, such as printing temperature, retraction speed, retraction distance, printing speed, and fan speed.

Myers et al. <sup>[25]</sup> analyzed the effect of FDM process parameters (layer height, flow rate, and printing speed) on the printability and compressive strength of Schoen Gyroid and Schwartz Primitive structures made of PLA. A full factorial design was conducted to identify that there are three critical process parameters that influence the geometrical accuracy of a given print: layer height, flow rate and print speed, with four process parameters having a minor effect on the geometrical accuracy: nozzle temperature, build plate temperature, travel speed and retraction distance. It was found that the flow rate of material during printing (i.e., extrudability) was the most significant statistical parameter that influenced the geometrical accuracy of the test prints. Lower flow rate caused under extrusion (smaller geometrical dimensions) while higher flow rate caused over extrusion (larger geometrical dimensions). Furthermore, layer height was the next significant statistical parameter that affected the accuracy of the z-dimension (i.e., the height) and the strut diameter. Moreover, print speed had the smallest effect on the geometrical accuracy since it mainly controls the printing speed and layer adhesion.

Although the FDM process is most commonly utilized to fabricate polymeric composites, the PBF process produces parts with lower geometrical defects and higher machining accuracy when compared to the former <sup>[26][27]</sup>. As described by Gao et al. <sup>[28]</sup> in a comprehensive review, the interlayer bonding in the FDM process is known to be weak. Yet, many authors reported that geometrical deviations and surface roughness exist between the as-build and the as-designed parts even when using PBF processes <sup>[29][30][31][32]</sup>.

Fabricating a lattice material consisting of closed cells, such as plate lattices, is another challenge when considering the PBF process. These structures require the creation of circular holes on their surfaces for extracting the trapped and unfused powder material. This results in creating stress concentrations and decreasing the elastic moduli of these structures <sup>[33][34]</sup>. As an alternative, the material extrusion technique has been implemented by many authors to produce polymeric closed-cell lattice materials since it does not require the extraction of unprocessed material <sup>[35][36][37][38][39]</sup>.

#### 3.2. Polymeric Composite Materials

Synthesizing lattice metamaterials that are capable of performing multiple structural functionalities increased the interest in multifunctional materials that produce structures with conflicting mechanical properties, such as high ductility with improved mechanical strength. The present section introduces major techniques that are implemented to produce multi-functional lattice metamaterials, particularly, the utilization of composite materials and multi-material additive manufacturing techniques. In brief, a composite material is defined as the assembly of multiple materials composed of a matrix that is strengthened by a reinforcer <sup>[40]</sup>.

#### 3.2.1. Fiber Reinforced Composites

Fiber-reinforced composites are utilized to synthesize materials with a high strength-to-weight ratio. Different types of fibers are utilized to reinforce a host matrix, such as carbon fibers, glass fibers, and aramid fibers. For a thorough review of the different types of fibers, the readers are encouraged to refer to Prashanth et al. <sup>[41]</sup>. Furthermore, fiber alignment is one of the factors controlling the strength of fiber-reinforced composites.

Besides optimizing the fiber orientation in a host matrix, enhancing their fabrication procedure improves the mechanical behavior of the considered structure. Wang et al. <sup>[42]</sup> introduced a fabrication procedure to manufacture triangular corrugated structures (TCSs) made of continuous carbon fiber (CCF) reinforced thermosetting epoxy (EP) composite. The single-stroke printing path ensured a strong connection between the corrugated core and the face sheets. Subsequently, the CCF/EP TCS samples outperformed the ones made of unreinforced nylon, short fiber-reinforced nylon, and CCF-reinforced nylon in terms of compressive strength, stiffness, and energy absorption.

**Figure 3** illustrates the mechanical properties of reinforced and unreinforced polymeric lattice materials in terms of uniaxial modulus, ultimate strength, and specific energy absorption (SEA). The data of Schwartz Primitive were taken from a study conducted by Diamantopoulou et al. <sup>[43]</sup>, in which the sandwich construction of the Schwartz Primitive cell wall involved IP-S photoresist as the core material and alumina as the skin material. **Figure 3**a–c demonstrate that reinforcing the IP-S photoresist with variant weight percentages of the alumina enhanced the uniaxial compressive modulus, the SEA, and the ultimate strength of the considered structures, respectively.



**Figure 3.** Bar charts demonstrating the mechanical properties of reinforced and unreinforced polymeric lattice materials: (a) Uniaxial modulus. (b) Specific energy absorption (SEA), and (c) Ultimate strength <sup>[42][43][44]</sup>.

### 3.2.2. Polymeric-Derived Ceramic Composite

Polymeric-derived ceramic (PDC) composite emerged as a potent material that exhibits high temperature resistant and excellent mechanical properties <sup>[45]</sup>. **Figure 4** illustrates the general layout of producing an additively manufactured PDC lattice structure. Slurry preparation involves the mixing of polymeric and ceramic precursors with necessary chemicals to produce a precursor slurry or a precursor melt for laser/light-based <sup>[46]</sup> or extrusionbased <sup>[47]</sup> additive manufacturing techniques, respectively. When prepared for laser/light-based additive manufacturing, surfactant and dispersant are used to enhance the dispersity of ceramic particles into a polymeric resin. A photoinitiator is implemented to trigger the photopolymerization of the slurry mixture when exposed to ultraviolet light during 3D printing. After multiple heat treatment procedures, the printed precursor is pyrolyzed at high temperatures into a ceramic matrix to fill the voids in the printed precursor in a procedure known as polymer impregnation and pyrolysis (PIP). This procedure is repeated until a dense polymeric-derived ceramic composite is obtained <sup>[48][49]</sup>.



**Figure 4.** (a) General layout of producing an additively manufactured PDC lattice structure. (b) Demonstrating the effect of reinforcement content on the linear shrinkage of PDCs. (c) Demonstrating the effect of reinforcement content on the bending and compressive strengths of PDCs. (d) The effect of sintering/pyrolysis temperature on the compressive strength of PDCs.

Preparing a PDC composite for material extrusion technique, such as the DIW procedure, requires a material with efficient rheology (filament flowability). In DIW, the process material is provided as ink/paste composing of a polymeric material loaded with ceramic particulates. Very similar to the FDM procedure, an extrusion nozzle is utilized to deposit the ink under a controlled flow rate <sup>[50]</sup>. Liu et al. <sup>[46]</sup> elaborated on the rheological behavior and the linear shrinkage of carbon fiber-reinforced SiC composites produced via the DIW process. Linear shrinkage quantifies the decrease in fiber length when undergoing certain types of heat treatment. It was reported that increasing the content of carbon fiber decreased the viscosity of the printing inks which resulted in an excellent shear-thinning behavior and improved the extrudability of the ink for the DIW process.

Digital light processing (DLP) technique is a vat photopolymerization additive manufacturing process that is commonly utilized to fabricate lattice materials made of photosensitive ceramic slurry. DLP operates under similar

principles as the SLA procedure; however, a UV-light source and optical systems are utilized instead of a laser beam to cure a prepared slurry in the DLP procedure <sup>[51]</sup>. Su et al. <sup>[52]</sup> proposed a fabrication procedure in which low-cost and environmentally insensitive precursors are utilized to produce  $Li_4SiO_4$  powder with enhanced microstructural stability and higher phase purity when compared to the existing  $Li_4SiO_4$  PDC.

#### 3.2.3. Cementitious Composite

Concrete has been widely implemented in the construction industry due to its high compressive strength capability. However, owing to its brittleness and low tensile strength, reinforcing the cementitious composite with metals and polymers emerged as a viable solution to enhance their ductility <sup>[53]</sup>. Most commonly, steel rebars are used as reinforcement elements to improve the ductility of cementitious materials at the cost of corrosion problems <sup>[54]</sup>. As an alternative, 3D-printed polymeric lattices are utilized as reinforcement structures to enhance the ductility and improve the crack resistance of concrete without initiating corrosive reactions. Salazar et al. <sup>[55]</sup> enhanced the ductility of ultra-high-performance concrete (UHPC) by reinforcing it with octet-truss lattices made of PLA and ABS materials. The polymeric lattices were placed into beam-shaped molds and then infiltrated with UHPC to form the lattice-reinforced concrete beams. It turned out that strengthening the UHPC with a 33.7% PLA reinforcing ratio enhanced the peak load and the toughness by 54.6% and 8650%, respectively. The addition of PLA into the UHPC cementitious composite completely shifted the failure mode from brittle to ductile.

The aforementioned studies indicate the increasing interest in utilizing 3D-printed polymeric lattice materials as spatial reinforcing elements to enhance the ductility and strength of cementitious composites. It was highlighted that the enhancement of certain types of mechanical properties is attained at the cost of worsening some other performance markers. To this end, it is believed that the utilization of multi-objective optimization tools would support the enhancement of conflicting mechanical properties without deteriorating a specific property at the cost of another. Furthermore, since the performance of polymeric lattice materials is sensitive to temperatures, the current state of the art seems to lack explanations for the effect of thermal gradient on the mechanical performance of cementitious composites reinforced with polymeric lattice materials.

### 3.3. Multi-Material Additive Manufacturing

Despite the distinguished mechanical performance of metamaterials, achieving a combination of conflicting properties is challenging with single material composition <sup>[56]</sup>. For example, ceramic-based metamaterials are superior in terms of compressive strength but inferior in terms of energy dissipation due to their brittle nature <sup>[57][58]</sup>. To this end, multi-material additive manufacturing is exploited to produce structures characterized by conflicting properties, such as attaining efficient stiffness and energy absorption capabilities. For example, Yavas et al. <sup>[59]</sup> fabricated honeycomb structures composed of variant ratios of hard and soft materials, in which the hard shell was made of PLA to maintain stiffness, and the soft core was made of TPU to enhance energy absorption. The considered structures were fabricated using a multi-material FDM printer equipped with a dual extrusion system such that each nozzle deposits one type of material.

## 4. Mechanical Characterization of Polymeric Metamaterials

### 4.1. Uniaxial Compression Tests

One of the simplest mechanical tests that exist is the uniaxial compression test, where a specimen is used to conduct the test and investigate the compressive behaviors of polymeric metamaterials. Various mechanical properties can be obtained from uniaxial compression tests, such as the elastic modulus, yield strength and ultimate strength.

Due to their superior properties, many studies have investigated the mechanical properties of TPMS lattices under uniaxial compression tests <sup>[60][61][62][63][64][65][66]</sup>. Uniform TPMS lattices have been investigated by Abueidda et al. <sup>[62]</sup>, specifically sheet-based TPMS cubic samples of Schwartz Primitive, Schoen's I-WP and Neovius. The cubic samples were printed using the SLS technology with PA12 material, where different specimen sizes were investigated with a unit cell edge length of 1.5 cm each. The elastic modulus and the ultimate strength of these three sheet-based TPMS lattices were investigated for a relative density range of 5% to 26%. It was found that the I-WP and Neovius structures had higher uniaxial compressive moduli and uniaxial strengths than the Primitive lattice.

In a subsequent study, Abueidda et al. <sup>[64]</sup> investigated the Gyroid sheet-based TPMS lattice in a similar manner and found that the uniaxial compressive properties of the Gyroid rank between the Neovius (highest properties) and the I-WP lattices. Similarly, another study <sup>[63]</sup> investigated uniform TPMS lattices. However, both sheet-based and strut-based TPMS lattices were investigated in this study, based on the Diamond and Gyroid topologies.

## 4.2. Bending Tests

The flexural properties of materials are important as well to characterize the performance of polymeric metamaterials, which can be obtained using three-point or four-point bending tests. Several studies have attempted these tests on polymeric metamaterials and reported their flexural properties. A couple of studies investigated the flexural properties of polymeric honeycomb structures <sup>[67][68]</sup>. Li and Wang <sup>[68]</sup> conducted three-point bending tests on composite sandwich structures made of acrylic-based photopolymer honeycomb as the core structure. Two different honeycomb structures were considered: the conventional honeycomb and the re-entrant honeycomb. In terms of the face sheets, three different face sheet materials were used to manufacture different specimens, which were acrylic-based photopolymer, woven carbon fiber reinforced polymer and unidirectional carbon fiber reinforced polymer. By investigating the flexural stiffness, flexural strength and the energy absorption of these specimens, it was found that the conventional honeycomb structures had higher values than the re-entrant honeycomb at a corresponding relative density. However, due to the relatively homogenous stress distribution, the re-entrant honeycomb sandwich constructions exhibited an interesting global failure mode.

### 4.3. Impact Tests

In order to quantify the amount of energy absorbed by a material during a fast collision (in a scale of milli-seconds), one of the common mechanical tests performed is the impact test. This gives a measure of a material's toughness which is an important parameter in various applications. De Castro et al. <sup>[67]</sup> performed a Charpy impact test on similar PLA honeycomb sandwich structures that were used for three-point bending, with the three different core designs: hexagonal honeycombs oriented (i) out-of-plane and (ii) in-plane, and S-shape corrugated. With a similar mass for all three samples, the results showed that energy absorption of the out-of-plane oriented and S-shape corrugated honeycomb structures was similar and higher than the in-plane oriented honeycomb sandwich structure

Furthermore, low-velocity impact tests have been performed on plate lattices. Andrew et al. <sup>[37]</sup> conducted impact tests on polymeric plate lattices printed with thermoplastic using SLS printing technology. Six types of core topologies were used which included SC, BCC and FCC and the three hybrid structures of SC-BCC, SC-FCC and SC-BCC-FCC. All plate specimens were made of  $2 \times 2 \times 2$  number of unit cells with 16 mm unit cells. The study focused on two specimen categories which were plate-lattice specimens with constant relative density and constant plate thickness. For the former case, the relative density was set to be 35% while the thickness of the plates in different architectures varied depending upon the volume of the plates in each lattice. For the latter case, the plate thickness of all specimens was set at 1 mm while the relative density varied between the specimens. From the low velocity impact tests carried out on the different constant thickness specimens, the specimens were ranked from the highest energy absorbed to the lowest as follows SC-BCC-FCC > SC-BCC > SC-FCC > SC-FCC > SC-FCC > SC-BCC > SC-FCC > S

## 4.4. Other Tests of Polymeric Metamaterials

Looking away from the uniaxial compression, bending and impact tests, there is a scarcity of mechanical tests of polymeric metamaterials. Although there have been attempts to numerically model the mechanical behavior of TPMS structures under complex loading, such as biaxial, shear, torsion and combination of other loading types <sup>[69]</sup>, experimental investigations would provide a clear image of the performance of these metamaterials in a variety of applications.

## 5. Conclusions

Although several lattice architectures have been proposed in the literature with the sole purpose of meeting the desired engineering function, there are still aspects related to metamaterial designs that are yet to be explored to the best of the authors' knowledge. Often, strut-based, plate-based and comb-based lattices are derived explicitly using CAD tools which is a time-consuming process. It will be interesting to be able to construct these classes of lattice materials implicitly as the mathematically derived TPMS lattices to accelerate their designing and facilitate functional grading of their topological properties. In addition, given the major and rapid advancements in additive manufacturing, there is an increased demand for using such metamaterials in applications that do not involve uniaxial compression loading only. There is a scarcity of investigations on the mechanical properties in loading

conditions such as uniaxial tension, biaxial, shear, torsion and a combination of such loading conditions, which is required to have a clear image of the performance of these polymeric metamaterials in a variety of applications. This is needed to speed up the implementation of additive manufacturing into various applications. It is important not to forget the role of machine/deep learning in filling the current gaps in the additive manufacturing process.

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