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Multi-Material Printing of Multi Lengthscale Bio-composite Membranes

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Abstract

We present advances in a water-based fabrication and design approach for the 3D-printing of functionally graded bio-composites. These developments demonstrate further refinement and large-scale deployment of Water-Based Digital Fabrication in the construction of architectural-scale membrane structures. Fully biodegradable and composed of the most abundant materials of our planet —cellulose, chitosan, and water—these bio-composites represent a versatile system for the construction of dynamic membrane structures with graded mechanical and optical properties across scales.

The described digital fabrication system, based on the Fabrication-Information Modeling (FIM) approach, enables the simultaneous printing and mixing of water-soluble materials through real-time feedback-driven control logics. In this way, we explore programming material behaviors through the design of diffusion-based material gradients in biocomposite hydrogels and examine the impact of local and global material cues on the full-scale assembled structure. Novel generative design methods integrate material-specific fabrication parameters and enable data-driven, hierarchical organization of the manufacturing workflow. The parametric control of the chemical composition in the 3D printing process allows for the customization of structural, environmental performative parameters of the structures across scales. In combination, these methods comprise a scalable fabrication system for the creation of adaptable, biodegradable membrane structures with minimal need for environmental control.

Keywords: Additive manufacturing, membrane structures, water-based fabrication, biocomposite, material ecology, fabrication-information modeling, functionally graded materials, architectural hydrogels

1. Introduction

Due to their abundance and ability to safely biodegrade, the manufacturing of biopolymers at scale holds promise as a sustainable and versatile material system. Current developments in biopolymer 3D-printing have centered on the creation of small-scale scaffolds for biomedical purposes [1,2], the extrusion of biopolymers suspended in thermoplastics, and the manufacturing of photocurable resins containing small amounts of biopolymers but may not be fully biodegradable [3]. Scalable systems for the additive manufacturing of pure biopolymer structures remains largely unexplored. Here, we outline further developments in a Water-Based Digital Fabrication system capable of constructing large-scale functionally-graded bio-composites (materials composed of several biopolymers) at room-temperature [4]. Furthermore, we describe a prototype that demonstrates these capabilities at scale.

Previously we demonstrated the fabrication of purely chitosan-based rigid structures at a length scale of up to 4m, as well as multi-layer bio-composites with the ability to dynamically transition between flexible "skin" and rigid "shell" behaviors through the application of humidity [4, 5]. A compelling characteristic of these bio-composites is the ability to dramatically vary material and optical properties through relatively small changes in composition, which is ideal for dynamic variations within a single continuously fabricated object. The implemented fabrication system combines room-temperature extrusion of organic biopolymer gels with the precision and operating scale of industrial robotics. An end effector is outfitted with a pneumatic extruder containing a 300mL cartridge with a detachable nozzle. Nozzle diameter and pressure are varied according to the viscosity and particle diameter of the extruded colloid. Furthermore, extrusion pressure and feed-rate (the speed at which the robot travels) are varied in real-time in order to alter the rate of material deposition. These fabrication parameters are typically assigned prior to printing based on previous experimentation, but they can also be altered "on-the-fly" based on the designer's observations or data gathered during fabrication.

Printed biopolymer gels solidify through evaporation over time. During this process, they continue to flow at a rate proportional to their viscosity and that of the surrounding gels. This provides a window during which materials can flow into one another, leading to the creation of gradients that surpass mechanical resolution limits, and a smooth transitional zone between materials with no need for dithering [5, 6].

We examine the capabilities of this technology and demonstrate a prototype in which both smooth and discrete transitions between materials within large-scale bio-composite membranes are implemented. This represents the embedding of non-diffuse internal structures for the purpose of reinforcing otherwise flexible materials. Smooth transitions allow materials of dissimilar mechanical and optical properties to be mixed during fabrication while bypassing the need to allow hydrogels to fully dry between layers.

2. Methods

2.1 Bio-composite Composition

Each biopolymer hydrogel is composed of 20%-40% apple pectin (VWR, Radnor PA) with varied amounts of 85%-deacetylated chitosan (VWR, Radnor PA), microcrystalline cellulose (VWR, Radnor PA), methylcellulose (VWR, Radnor PA), and natural vegetable glycerin (Essential Depot, Sebring FL) added in order to tune material properties, dissociation rate, and environmental response [8]. Regardless of composition, extruded biopolymers retain their ability to biodegrade more rapidly than many synthetic polymers and bioplastics, with dissociation times in saline submersion ranging from 1 hour to 90 days, according to geometry and material composition. Colloids are mixed in 600mL batches at 2℃ and printed immediately in order to avoid stratification of substrates with varying density.

In order to create optical gradients across the fabricated membranes, natural pigments and dyes are incorporated into bio-composites of each base composition. These include: 1% turmeric powder (Healthworks, Scottsdale AZ), 4% beetroot powder (Bulk Supplements, Henderson NV), 1% maya blue mineral pigment (Natural Pigments, Willits CA), and 1% squid ink melanin (Yamamoto Industry, Tokyo, Japan). Regions with no added color maintain the natural amber hue of apple pectin.

2.2. Fabrication System

We implement a custom pneumatic extrusion system programmed with instruction-feed capabilities that can be affixed to a multi-axis industrial robot or three-axis gantry [7]. Print-nozzle diameters ranging from 12-20GA were utilized according to material viscosity and particle size. Pneumatic and motor control systems allow for pressure and feedrate to be controlled within ranges of 0-400kPa and 0-

8000mm/min, respectively. These parameters are tuned in order to suit specific materials and geometries.

2.3. Computational Workflow

The set of points continuously visited by the end effector without a complete stop in movement is referred to as the toolpath. A print may consist of several toolpaths due to scale or geometry. Weights corresponding to the amount of a material to be deposited at a point are assigned to every vertex in a toolpath. Weight values are stored as an N-dimensional vector w, where N is the number of materials in use across the entirety of all toolpaths within a print. The magnitude of $||w(x,y,z)||$ for any coordinate in a toolpath is 1 for any print where total material deposition is equal at all points, as is the case for even-thickness membranes. During fabrication, material weight values are used as an interpolation parameter between minimum and maximum extrusion pressures, with maximum values corresponding to extruded lines of equal thickness for each material.

Ideally, toolpath geometry varies independently of material weights. In order to achieve this, weight vectors are stored within a mesh that is decoupled from toolpath geometry. Once a toolpath has been established, vertices are populated with weights via a weighted nearest neighbor interpolation, with a search radius of 1 cm. Weight vectors that have been assigned to toolpath vertices are then divided into their individual components and print parameters are generated for each material. Each vector component is used as an interpolation weight between a maximum and minimum ranges for fabrication parameters such as pressure. For continuously extruded heterogeneous membranes, the following pressure ranges were used in printing toolpath-invariant prints with a federate of 800mm/min and a nozzle width of 18GA: 28% pectin, 5% glycerin printed at 0-60kPa, 32% pectin, 2% chitosan, 4% glycerin printed at 0-100kPa, 32% pectin, 4% chitosan, 3% glycerin printed at 41-185kPa.

3. Results

3.1 Continuously Graded Biopolymer Membranes

In contrast to other multi-material printing methods, the water-soluble nature of the extruded colloids allows for a level of diffusion during the drying process. This enables material distributions to equalize into gradients that surpass the system’s mechanical resolution, thereby creating smooth gradients even between materials of different viscosities. Such gradients can be used to transition between materials of differing properties or to interpolate between materials and create regions with hybrid properties not found in any individual biocomposite. As previously reported [2], even small changes in biopolymer composition can result in large changes in material properties. This dramatically increases the range of material properties that can be created with a relatively limited material palette.

3.2 Embedded Patterning of Discrete Material Changes

Discrete transitions in materials can be created within a single biopolymer membrane by printing separate toolpaths into membranes before they dry completely. Membranes dry at room temperature and 30% humidity for 10 to 24 hours until they reach a semisolid state where further material extrusions sink into the membrane, but do not diffuse and lose integrity. The resulting embedded pattern does not impact the surface features of the membrane, which maintains its geometry within the continuous print.

3.3 Hierarchical Structure Design

The structure follows a hierarchical design logic that organizes rigid skeletal elements, flexible membrane elements, and a semi-rigid reinforcement pattern. Skeletal elements are additively manufactured by fused deposition with a reusable photopolymer with high strength and elastic modulus. The composition of flexible elements is continuously varied according to structural and environmental parameters. Semi-rigid embedded reinforcement patterns are composed of 32% pectin, 4% chitosan, 4% glycerin, and 1% squid ink melanin.

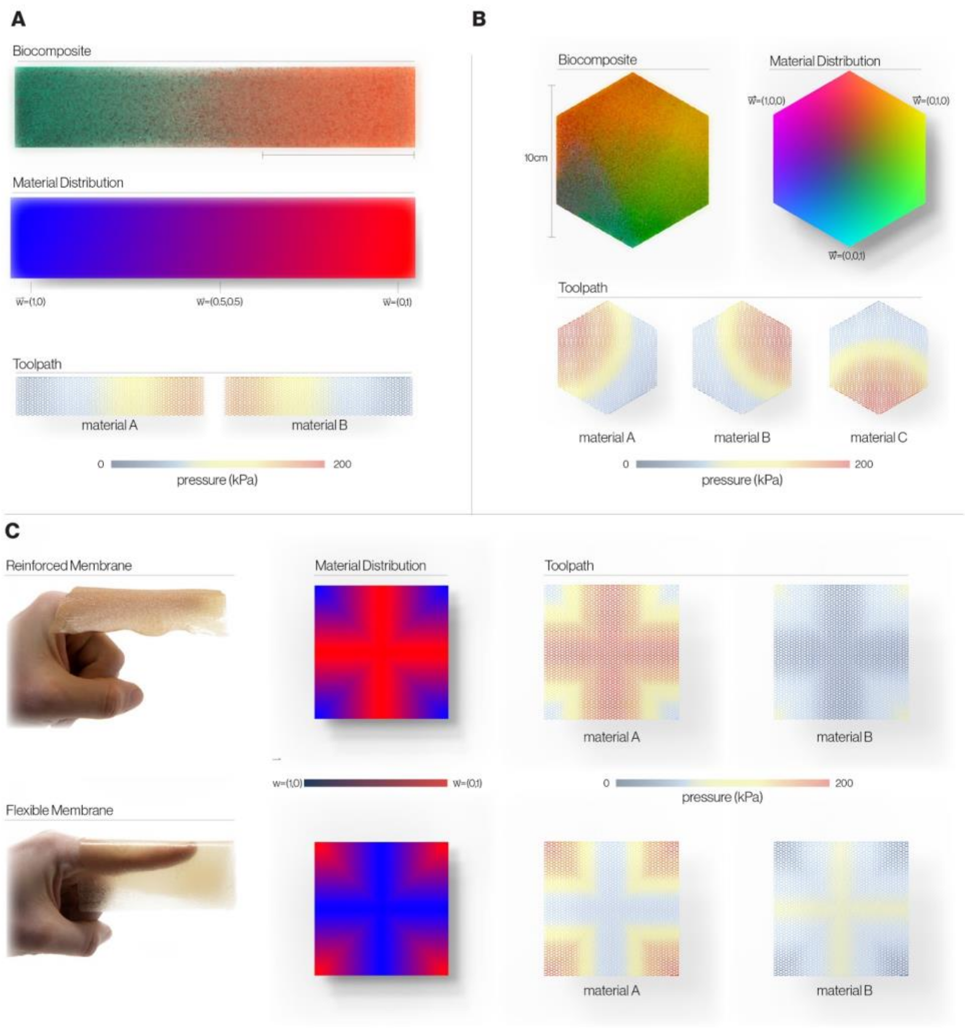

Figure 1: Continuously graded biopolymer membranes for 2 (a) and 3 (b) materials are shown for toolpath invariant prints. Material weights are stored in a vector w(1,... N) where N is equal to the number of materials used. Each vector coordinate is then mapped to pressure values across a toolpath. High- and low-end pressures are specific to each extruded material. Colloids with dissimilar material properties can be blended within a toolpath invariant print to produce different mechanical effects (c).

3.4 Global Geometry and Skeleton

The base geometry features anticlastic curvature shaping an elongated enclosure, open on one side to differentiate interior and exterior space. The base mesh is subdivided into a series of 20 developable quad strips that further define the membrane boundaries. Larger segments are consolidated towards the center of the structure while the smaller sections are arranged closer to the edge of the structure.

The boundaries of the mesh patches are extracted to form the borders of the membrane panels and a further resampling of the quad strip topology ensures the developability of the membrane surfaces. The first hierarchical layer defines the panel size as well as sizing of the main structural branches. Large panels in the center require a stiffer and thicker cross section to ensure stability of the structure, while thinning out towards the top and sides. The parametric model blends in between the various member sizes while ensuring the developability of the panels in between.

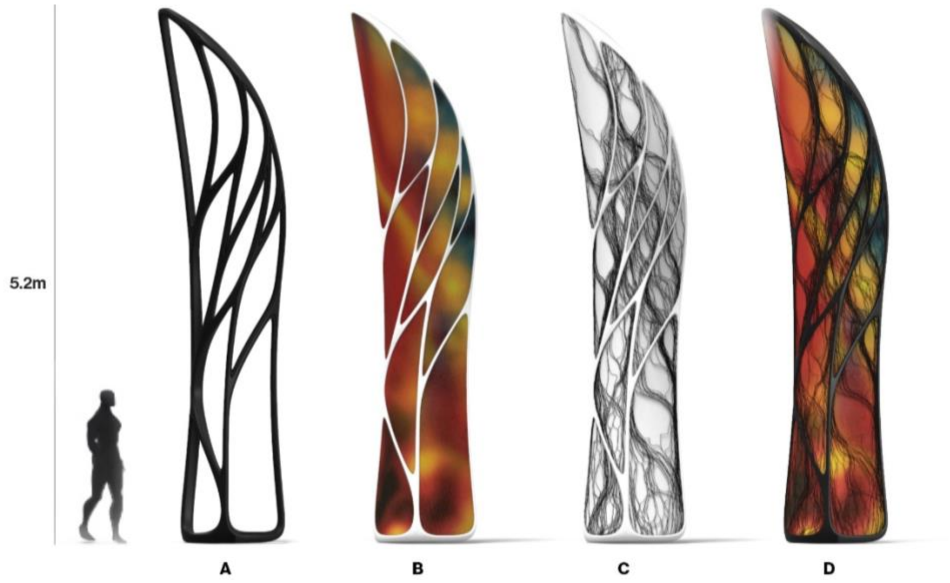

Figure 2: A prototypical structure is defined hierarchically beginning with rigid skeletal elements (a) followed by a continuously graded heterogeneous biopolymer membrane (b) and a semi-rigid embedded venation pattern that links the two previous levels (c) to create a continuous form (d).

3.5 Continuously Graded Flexible Membranes

Material gradation in flexible membrane elements can be divided into two categories: gradation of mechanical properties and gradation of optical properties. Importantly, these parameters are independent of one another. Both mechanical and optical properties are assigned according to global parameters including surface irradiance, structural load, and membrane curvature. Surface irradiance is calculated via sun-path simulation. Membrane curvature refers to the normal curvature of modeled biopolymer membranes and is highest where each panel bends to press-fit into rigid skeletal elements. Structural load and surface topology data is used to create a measure of surface flexure that represents the degree to which non-reinforced membranes would flex or bow.

A distribution of 28% pectin, 5% glycerin and 32% pectin, 2% chitosan, 4% glycerin is linearly assigned according to membrane curvature with the more flexible composition being assigned to the edges of panels where press-fitting occurs. To create optical gradients, surface irradiance, membrane curvature, and surface flexure magnitudes are used as parametric inputs to linear functions, the outputs of which were mapped to gradients. By specifically assigning gradient color limits, patterns are generated to template interactions key species in an ecology.

The first uses an aposematic pattern that incorporates motifs used in a number of species to repel predators [9], such as dark regions with high-contrast red and yellow patterning with sharp transitions. The second global pattern incorporates smooth gradients of blue and yellow hues visible to certain pollinators but not to other insects [10]. A third pattern combines shades of yellow that serve to attract aphids [11] with gradients of red similar to those used in ladybug sexual selection [12], thereby staging a hypothetical encounter between the two species. These patterns are selectively combined to create a single global color scheme with regions from each individual map.

3.6 Semi-Rigid Embedded Elements

Semi-rigid material containing 32% pectin, 4% chitosan, and 3% glycerin is embedded into flexible membranes in order to provide a level of reinforcement in areas distant from rigid skeletal elements. In order to create a pattern serving the simultaneous goals of reinforcing areas prone to flexure, minimizing disruption to optical patterns, and responding to structural irradiance, a modification of the *Boids*

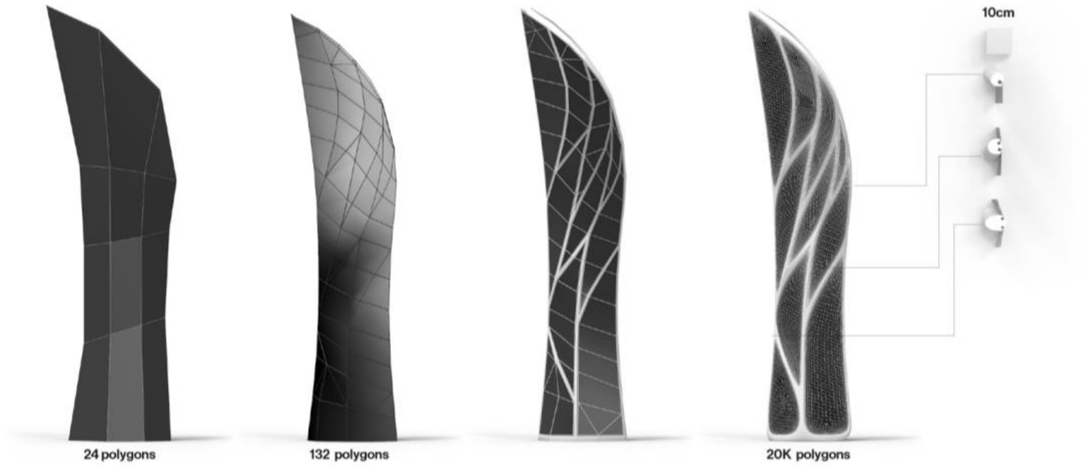

Figure 3: From left to right, low polygon representations of the base geometry, parametric mapping of skeletal member thickness based on load, generation of skeletal geometry based on developable mesh segment boundaries, and thickness assignment to skeletal members. Low and high polygon representations are operated on simultaneously.

Figure 4: Environmental data associated with global scale is modeled for structural irradiance, panel membrane curvature, and surface flexure for the structure in its assembled state (a). Global maps are translated into 2D representations (b) that guide the distribution of material and color for flexible membrane elements (c) and semi-rigid reinforcement patterns (d).

artificial life program is used with cohesion, separation, and path-following behaviors tethered to data from environmental maps [13]. Reinforcement patterns are generated by two distinct swarms of 125 and 85 agents respectively, the trails of which are then cross-linked via kernel-density estimation-based edge bundling [14]. The first swarm originates at the structure's base and follows a weighted navigation system designed to maximize the traversal of gradient boundaries where colors remain constant while maintaining a distance of 3.25cm from neighboring agents. Priority is given to steering that directs agents towards regions with high levels of flexure that require reinforcement. The second swarm originates from the structure's lower-ventral region and attempts to align to the gradient of the flexure map while separating in regions of low irradiance and gathering more densely in regions of high irradiance. The cross-linking of trails from both swarms is intended to provide lateral strength to patterns without the need for strictly lateral agent motions.

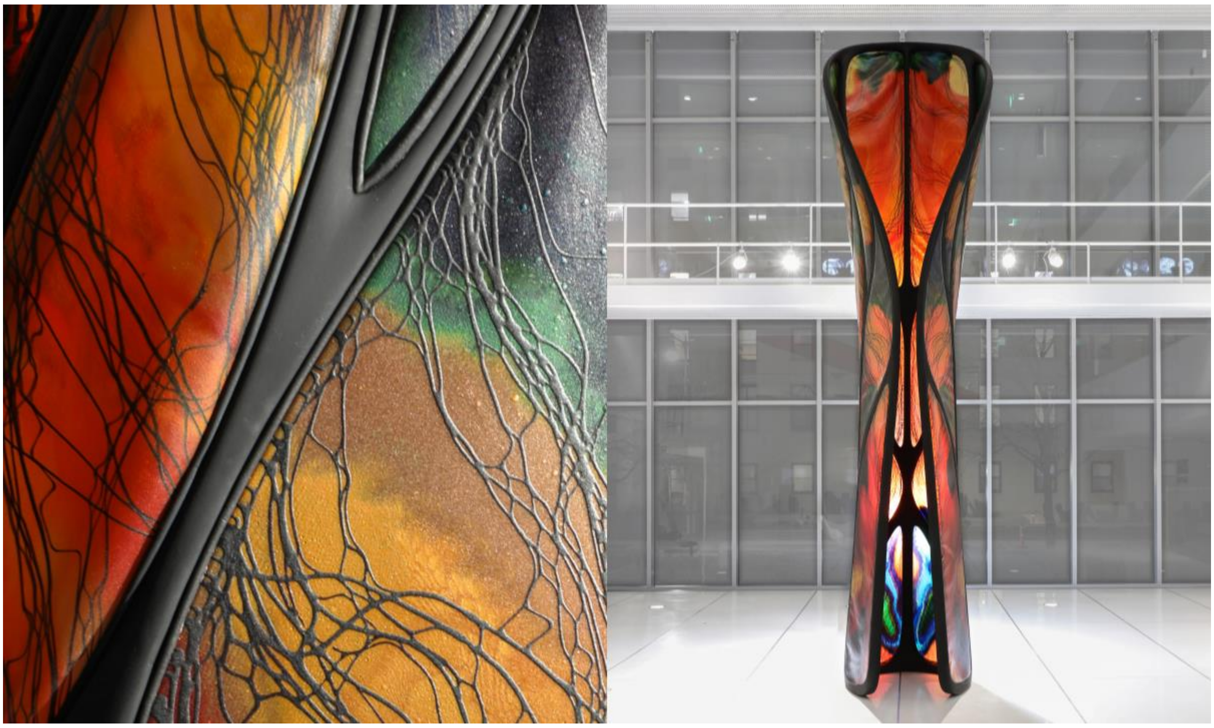

Figure 5: Close up (left) and front elevation (right) views of assembled structure.

4. Discussion and Outlook

This research demonstrates novel methods for the design and digital fabrication of hydrogel membranes with structural hierarchy across scales. Combined with Fabrication-information Modeling, the workflow presented enables the creation of smooth transitions between materials surpassing known mechanical limits per resolution. Importantly, the scalability of these methods allows for the digital fabrication of structures where a range of material properties are controlled and expressed both visually and functionally. We introduce the possibility to gradually vary material stiffness, environmental reactivity, age, and degradation through our custom digital fabrication platform. We plan on further fine-tuning material behaviors in terms of controlled decay and deformation under changing temperature and humidity, both structurally and visually. Further integration between rigid and flexible elements could be achieved through the incorporation of strong bio-composites with minimal deformation. This would allow for the creation of freestanding structures that transition between functional elements without the need for assembly. The architectural-scale fabrication of these techniques not only ensures the scalability of the described methods, but highlights developments in materials science, computation, and fabrication that allow sustainable, bio-degradable materials to be augmented with high-performance capabilities. These developments point towards a future where structures fabricated from natural, abundant components transcend the limitations of conventional materials.

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