

**Water-based Digital Design and Fabrication:
Material, Product, and Architectural Explorations
in Printing Chitosan and its Composites**

by

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Submitted to the Program in Media Arts and Sciences,
School of Architecture and Planning
in partial fulfillment of the requirements for the degree of

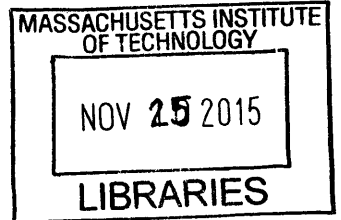
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Abstract

Conventional digital design tools display little integration between shape formation and materialization resulting in disassociation between shape and matter. Contrarily, in the natural world shape and matter are structured through growth and adaptation, resulting in highly tunable and hierarchically structured constructs, which exhibit excellent mechanical properties. Working towards integration, rigorous bottom-up natural material studies have resulted in a novel multi-scale digital design and fabrication platform that is precisely tailored, viable, and a rich companion to develop sustainable explorations across application scales integrating shape and matter control. Specifically, the research introduces design and environmental motivations driving novel sustainable digital manufacturing of water-based biomaterial structures at the architectural, and product-scale. Water is harnessed to tune biomaterial properties, to guide shape formation by natural evaporation, and to fully recycle and reuse material structures. Initial outcomes demonstrate self-supporting structural constructs displaying multi-functionality informed by graded material properties and hierarchical distribution depositions. I discuss contemporary literature in water-based manufacturing, and detail methods of the novel additive fabrication platform that combines a robotic positioning system and customized multi-barrel deposition system. Important contributions of the platform development as a design companion serve to advance sustainable digital manufacturing and propel it towards biologically inspired and material-informed techniques. Integrated material-based design studies, novel technology development, and sustainable motivation, produce an invention that outputs functional biodegradable products, reduces the need for external energy sources for fabrication, operates at room temperature, uses mild chemicals, and could embed productive microorganism cultures due to the biocompatibility of the materials used, pointing towards new possibilities for digital fabrication of living materials. Finally, the work advocates for the designer to play the role of a cohesive thinker, as well as a rigorous science and aesthetics explorer, able to seed novel processes that emerge from material studies towards digital design and advanced fabrication.

Keywords: new design companions; material-driven design; additive manufacturing; water-based digital fabrication; bio-materials catalogue; environmental engineering; architectural design; product design; biological design;

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Chapter 1

Introduction

This thesis is structured as follows: the present chapter, Chapter 1, introduces architectural and environmental motivations driving this work as well as intended research contributions and outlines the importance of material practices as new design companions within the case study of water-based digital design and fabrication of bio-material structures across application scales. Chapter 2 discusses contemporary literature in the role of water-based materials and processes in biology and manufacturing. The core of this document, Chapter 3, proposes a novel water-based fabrication platform and supports the approach by detailing methods and results. The last chapter, Chapter 4, concludes with an analysis of the thesis contributions and perspectives for future work.

1.1 Research Motivations

This thesis is motivated by architectural design and environmental engineering and falls at the intersection of bio-materials science, robotic digital fabrication, and material-informed computational modeling. I will investigate how water and water-based materials can be used in digital fabrication to internally organize, manufacture, and recycle structured objects and architectural parts.

1.1.1 Architectural: Manufacturing Designs

While contemporary digital fabrication tools are able to produce geometrically sophisticated objects and structures, such constructs are typically not sustainable insofar as material and energy use are considered (Oxman et al. [2012]). In contrast, assembly observed in nature employs mild chemicals, produces little to no waste, and uses small amounts of energy to produce multi-functional and adaptable systems (Vincent [2012]).

Water exists throughout nature and in all biological materials (Wiggins [1990]). It provides for an invisible scaffold designed to assemble basic molecules into structures with complex functions and property gradients. Examples include the giant squid beak where water content contributes to a significant stiffness gradient (Dumitriu [2004], Vincent [2012], Miserez et al. [2008]). The squid's beak is one of the hardest and stiffest wholly organic materials known. Water-storing proteins,

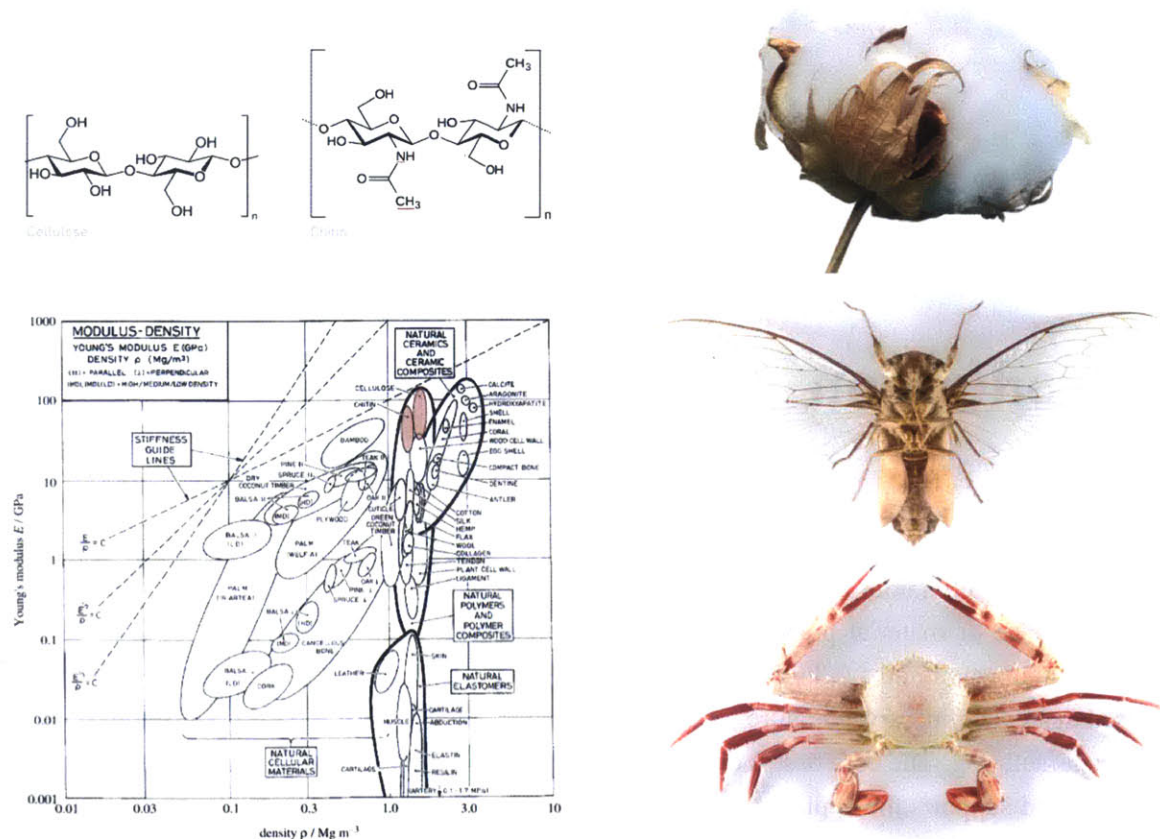


Figure 1-1: Structural polysaccharides such as chitin and cellulose are very abundant in nature. Cellulose provides structure for plants and its most pure form is the cotton flower. Chitin supports the exoskeleton of insects and crustaceans. The elastic moduli and tensile strength of chitin and cellulose are similar to those of engineering ceramics and metal alloys.

and polymers are carefully combined to generate its internal structure contributing to exceptionally graded material properties. When hydrated the beak exhibits an extreme functional gradient across its structure: it is sharp and firm at the tip with a tensile strength of 35MPa, and 10 times more compliant at the rim with a tensile strength of 3.5MPa (Miserez et al. [2008]). Another example of water-mediated structural performance is the chitin-made insect cuticles and crustacean shells. These structures accomplish dual functionality enabled by variable hydration. Chitin can be found, at once, in hard composites that shield insects, and in the flexible tissue found in their joints. Despite chitin's extraordinary properties, and in spite of its abundance, it is mostly used in pharmaceutical and dietary products (Lieleg and Ribbeck [2011], Dumitriu [2004], Vincent [2012], Rinaudo [2006], Gutowska et al. [2001]), and has not yet been deployed in large scale manufacturing of consumer goods (Fernandez and Ingber [2014], Mogas-Soldevila et al. [2014]).

Indeed, water is known to initiate or activate chemical reactions in biological materials and, in the engineering disciplines, it is especially relevant in the context of programmable materials (Krieg and Menges [2013], Reichert et al. [2015], Tibbits [2014]). Within traditional building construction, water can be found in common construction materials such as clay, concrete or plaster, where

it also acts as an initiator/activator. Metals and glass too, involve water as a lubricator and cooler for efficient manufacturing. More recently, water-as-activator has been used in programmable composites where hydrophilic polymers, which expand 150 percent when exposed to water, are used at the joints of segmented strands for underwater 2D or 3D shaping processes (Tibbits [2014]). Additionally, recent research demonstrates a new way to utilize wood as a responsive building envelope. Wood is a hygroscopic material that can absorb water molecules causing it to shrink and swell in the tangential direction of the fibers (Krieg and Menges [2013]). As a result, thin wooden sheets can be induced to find their shape in response to humidity changes when tuning the fiber direction in the exposed components (Krieg and Menges [2013], Reichert et al. [2015]). Common to all of these examples is the use of water as an agent for material processing and object shape change. This is contrary to its use in the biological world as a building block for internal organization, environmental tunability and biodegradability.

1.1.2 Environmental: Engineering Sustainability

Man-made plastics are challenging to reuse or degrade (US Environmental Protection [2013]). Only about 9% of plastic waste generated in the US is recyclable, while 27% of glass, 34% of metal, and 65% of cardboard are routinely recycled (US Environmental Protection [2013]). In the construction industry, synthetic plastics derived from petrochemicals are vastly used and make up most of pollutant non-recyclable waste (US Environmental Protection [2013]). While it could provide an alternative to fuel-based plastic materials, the use of regenerated bio-materials for product, architectural, and engineering applications is still not widespread (Fernandez and Ingber [2014]). This low use rate exists in spite of the fact that natural materials display exceptional mechanical properties (Fernandez and Ingber [2014, 2013], Gibson and Ashby [1997]). Cellulose provides structure for plants, and chitin supports the exoskeleton of insects and crustaceans (Dumitriu [2004], Vincent [2012]). The elastic moduli and tensile strength of structural polysaccharides such as chitin and cellulose are similar to those of engineering ceramics and metal alloys (Gibson and Ashby [1997]) (Figure 1-1).

Natural materials, and specifically polymers and polysaccharides, provide for a vast renewable resource produced at much higher rates than man-made synthetic polymers (Figure 1-2). Consequently, polysaccharide derivatives could replace existing synthetic fuel-based polymers, provide new property combinations for relevant applications, and play a key role in future manufacturing implementations (Dumitriu [2004], Gibson and Ashby [1997]).

1.2 Intended Contributions

1.2.1 Theoretical: New Design Companions

Biological structures adapt to external stimuli by growth-induced material property variation resulting in hierarchically structured forms. Conventional digital design tools and processes, by contrast,



Figure 1-2: Natural polymers and polysaccharides provide for a vast renewable resource produced by organisms such as insects or crustaceans at much higher rates than man-made synthetic polymers.

lack in robust integration of computational design tools, digital fabrication environments and the material itself. Design driven by material behavior - such as digitally fabricated internal structures informed by load-responsive arrangements as can be found in the biological world - can help and consequent matter tuning by digital design and reconnect conceptualization and materialization of contemporary complex designs. I believe that such practices will enable a shift of both mindset and tools from geometry-centric design processes to material-centric approaches in digital design and fabrication.

1.2.2 Applied: Water-Based Fabrication Research

In this thesis I demonstrate an applied case study of Material Studies as New Design Companions surveying the methods and results of bottom-up material-driven design. Specifically in my work on

Water-Based Fabrication Research focused on the exploration of water as a fundamental material building block able to unravel novel aesthetics as well as extraordinary structural performance via additive fabrication of natural composites. Specifically, I use water as; (1) an initiation agent, to chemically formulate the composite solution; (2) an activation agent, achieving optimal viscosity and "fabricability" properties; (3) a shape formation agent via natural evaporation; (4) a recycling agent for the reuse of the deposited materials; and (5) a life sustaining agent, enabling the containment of living micro-organisms for biological augmentation. My research lies at the intersection of bio-materials science, digital design and additive manufacturing. I present a platform and its associated methods for the design and production of small to large-scale multifunctional constructs that have the capacity to interact with their environment and that are fully recyclable upon contact with water.

Chapter 2

Background

2.1 The Role of Matter

In the natural world shape results from bottom-up material organization, sophisticated property gradation and functional hierarchies developed over time within single material systems (Vincent et al. [2006], Vincent [2012]). As a result, natural structures such as bamboo, silk, or bone, display extraordinary properties. Gaining deeper insight into, and a better understanding of, the formation processes associated with these natural phenomena would greatly benefit the design of digital fabrication tools and technologies (Gibson and Ashby [1997], Mogas-Soldevila et al. [2014]).

2.1.1 Matter & Biological Design

Structures in nature are shaped according to the principles of minimum inventory for maximum diversity (Pearce [1990]). Organisms use freely available energy, simple molecular chemistry, mild chemicals, and abundant resources to build complex medium-, and large-scale structures, such as bird's nests or termite mounds (Pearce [1990], Vincent [2012]). Biological structures are locally attuned and responsive, resilient to disturbances, and fully recyclable (Vincent [2012]). Most bio-material structures are generated at ambient conditions with low energy and using water (Vincent [2012], Mogas-Soldevila et al. [2014]).

2.1.2 Matter & Manufacturing Design

Current design practice is characterized by the domination of shape over matter. Consequently, virtual shape-defining parameters are typically prioritized over physical material and fabrication constraints, which are often considered only in hindsight, following a geometric-centric design phase (Menges [2007], Oxman [2011], Marenko [2014]). Recent advances in direct digital manufacturing are enabling a shift from a geometry-centric to a material-centric design practice (Oxman [2011], Sabin [2013], Duro-Royo et al. [2014b]). Today, designers have access to highly sophisticated computer-aided fabrication hardware that is contributing to the development of new material-driven

design processes (Nicholas and Tamke [2012], Cabrinha [2008], Krieg and Menges [2013], Duro-Royo et al. [2014b]). As the two domains - the physical and the digital - interact during the early stages of form generation, new and unexpected structural and morphological potentials may emerge that approach natural morphogenesis increasing efficiency and augmenting performance (Menges [2007], Ramsgaard Thomsen et al. [2012], Marenko [2014]).

2.2 The Role of Water

The role of water in the human body, and in a large number of natural processes, is essential to natural growth and shaping. Consequently, soft matter is assuming a growing role in fundamental science and in technological applications. Soft materials can rarely be understood on one length and time scale alone (Peter and Kremer [2010]), and their transition into solids by water evaporation entails a fascinating change in internal organization. Time-based multi-scale manufacturing and shaping prediction of water-based bio-materials needs further research.

2.2.1 Water & Biological Design

Water exists in all biological materials. It provides an invisible structure driven by the interaction of intrinsic chemistry and the external environment. Water assembles basic molecules into structures with complex functions and property gradients such as the one found in the beak of the giant squid, where the water content contributes to a stiffness gradient (Dumitriu [2004], Vincent [2012], Miserez et al. [2008]). The squid's beak is one of the hardest and stiffest wholly organic materials known, and in its internal structure, water, proteins, and polymers are carefully combined to generate exceptional material properties. Once hydrated, this material system exhibits extreme degrees of property differentiation across its structure: it is sharp and firm at the tip, and soft and adaptable at the rim (Miserez et al. [2008]).

Another example of water-mediated performance is the chitin-based material system that composes insect cuticle and crustacean shells. It accomplishes a dual function enabled by differential hydration. Chitin makes both the hard composite material that shields insects, and the flexible tissue in their joints. Interestingly, despite chitin's extraordinary properties, and it being the most abundant organic compound on earth after cellulose, it is not yet used in manufacturing of consumer products (Fernandez and Ingber [2014], Mogas-Soldevila et al. [2014]). I intend to advance research in this direction.

The interaction of water with the polar chemistry of biological surfaces, whether inside or outside the cell, is the primary driver of mechanical and morphological properties (Vincent [2012]). The aqueous extracellular matrix of cells surrounds tissues and organs and provides mechanical support and regulation of survival, migration and differentiation. Two types of extracellular macromolecules compose the matrix (Katharina Ribbeck, personal communication, Fall 2014, MIT). Biological hydrogels are networks of protein-polysaccharide chains that typically contain 90-99% water. They surround biological functional entities such as cells, tissues, organs, or entire organisms. These gels

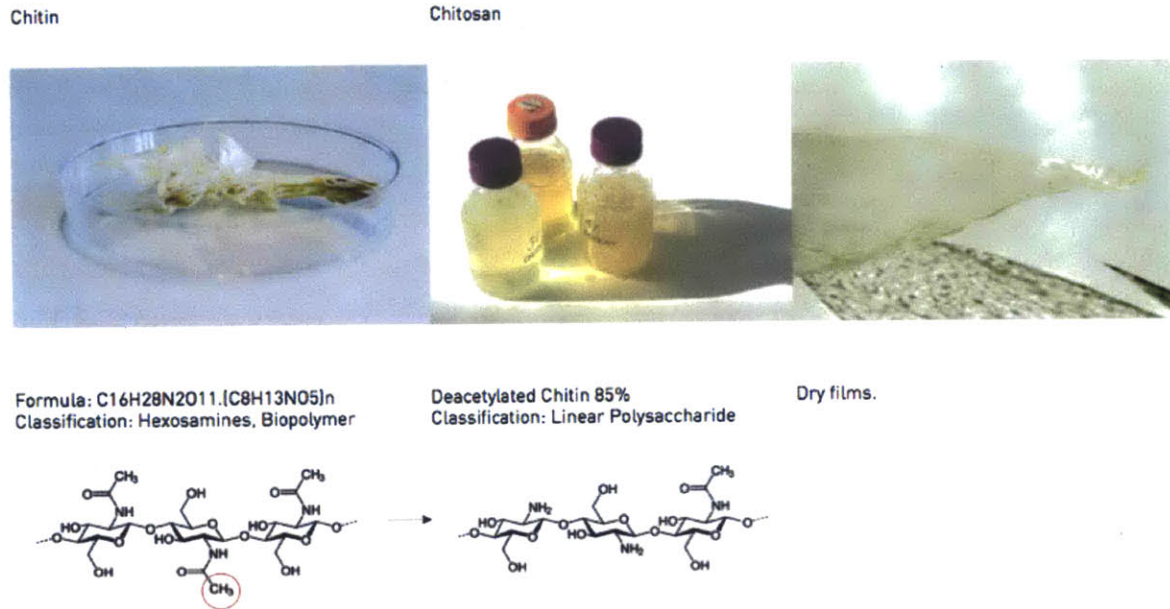


Figure 2-1: Chitin is a renewable resource and is isolated from crab and shrimp waste. Chitin can be made water soluble by deacetylizing it into chitosan that can be transformed into gels that form strong films by natural water evaporation.

can cover a wide spectrum of material properties, ranging from relatively soft and viscous-like to highly elastic (Lieleg and Ribbeck [2011]). Biological hydrogels establish and regulate the mechanical properties of cells and tissues and serve as lubricants in joints or on epithelial surfaces (Lieleg and Ribbeck [2011]). However, the physiological role of biological gels is not limited to their mechanical performance. They can also form selective barriers that control the exchange of molecules between different compartments (Lieleg and Ribbeck [2011]).

For instance in the human body, water performs a manifold of crucial functions. It maintains the health and integrity of every cell, keeps the bloodstream flowing through blood vessels, helps eliminate the byproducts of metabolism such as excess electrolytes and urea, it regulates body temperature through sweat, moistens mucous membranes such as those of the lungs and mouth, lubricates and cushion joints, reduces the risk of cystitis by keeping the bladder clear of bacteria, aids digestion and prevents constipation, moisturizes the skin to maintain its texture and appearance, carries nutrients and oxygen to cells, serves as a shock absorber inside the eyes, spinal cord and in the amniotic sac surrounding the fetus in pregnancy (Wiggins [1990]).

Water typically makes up from 80 to 95% of the mass of growing plant tissues. Cell walls allow the build up of water-induced turgor pressure within each cell that contributes to rigidity and mechanical stability of non-woody plant tissue, and is essential for many physiological processes including cell enlargement, gas exchange in the leaves, or transport of nutrients. Long-distance water movement is crucial to the survival of land plants (Raven et al. [2005]). Turgor pressure also enables another interesting phenomena; water-based actuation. The movement occurs when regions of cells lose pressure. When the plant is disturbed, sections on the stems are stimulated to release chemicals which force water out of the cell vacuoles and diffuse it out of the cells, producing a loss of cell

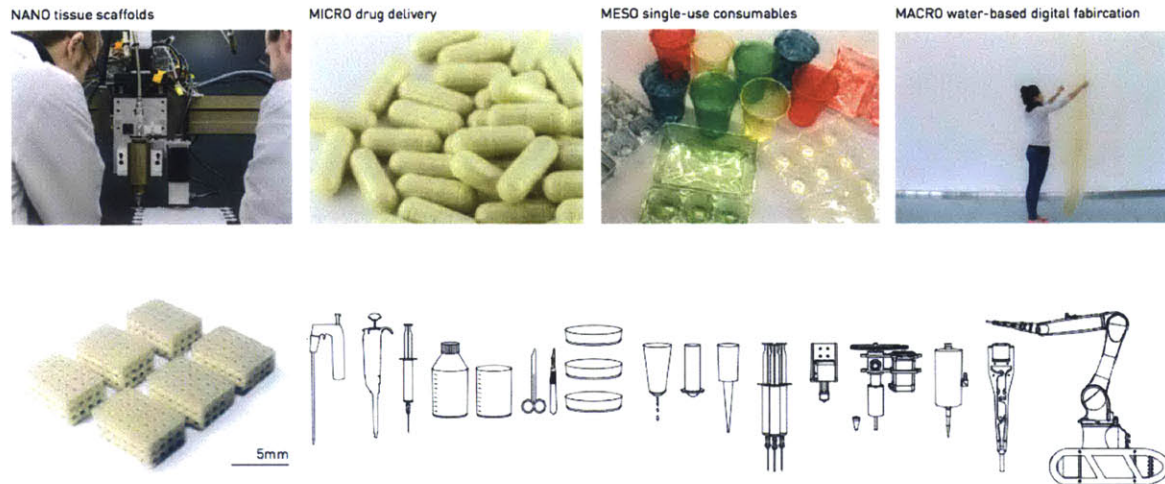


Figure 2-2: The properties of chitosan and other polysaccharides allow for the development of advanced materials used in multiple scale products.

pressure and collapse (Raven et al. [2005]).

2.2.2 Water & Manufacturing Design

For the past decade, manufacturing of hydrogels has become a rapidly evolving technique to produce nano-featured biocompatible tissue scaffolds for tissue engineering purposes (Li et al. [2005], Gutowska et al. [2001], Melchels et al. [2012]). There are mainly three different bio-fabrication techniques known to achieve fully interconnected soft-material 3D structures: laser-based, nozzle-based (also known as robotic dispensing) and printer-based systems (also known as inkjet printing) (Billiet et al. [2012], Malda et al. [2013]). The fabrication of hydrogel scaffolds requires mild processing conditions. Therefore, additive-manufacturing techniques such as fused deposition modeling (FDM) or selective laser sintering (SLS) cannot be used as they generally involve harsh processing conditions (Mogas-Soldevila et al. [2014], Fernandez and Ingber [2014]).

Recent research in tissue engineering proves that biodegradable scaffolds from chitosan (Figure 2-1) - deacetylated chitin from ground shrimp shells, a by-product of the fishing industry - and sodium alginate - a gum extracted from the cell walls of abundant brown algae - show significantly improved mechanical and biological properties (Li et al. [2005]). Moreover, it has been shown that chitosan enables the manufacturing of medium three-dimensional (3-D) objects, thereby offering a new pathway for large-scale production of fully compostable engineered components with complex forms.

Recent developments deploy conventional manufacturing methods such as injection casting into epoxy resin molds to produce chitosan-based functional 3D industrial products, such as fully compostable cups and egg storing containers (Fernandez and Ingber [2014, 2013], Rinaudo [2006]). The non-toxicity, water solubility, capacity for swelling, and chemical versatility of chitosan allows for the development of advanced functionalized materials that can be used in multiple scales such as

in drug delivery, tissue scaffolding, consumer products or large scale architectural parts (Figure 2-2) (Mogas-Soldevila et al. [2014], Fernandez and Ingber [2014], Billiet et al. [2012], Malda et al. [2013]). **I am interested in researching low-energy and mild-condition deposition systems in order to design large-scale functionally graded structures with environmental capabilities for applications in architectural and product design.**

Chapter 3

Proposal

3.1 New Design Companions

Water-based Digital Design and Fabrication research serves here as a case study positioned at the intersection of bio-inspired formation, material science, sustainable design and digital fabrication. It belongs to a stream of thought and practice that engages emerging technical material explorations in the design world. I envision New Design Companions as a novel way of legislating laws for a material-based design exploration seeding the production of new technologies and with product results that cannot be a-priori imagined (Figure 3-1).

Driven by aqueous material formation, in Water-based Digital Design and Fabrication research, a novel robotically controlled system is developed to deposit natural water-based organic structures at ambient conditions, using mild chemicals and low amounts of energy (Duro-Royo et al. [2015], Mogas-Soldevila and Oxman [2015]). It additively manufactures biodegradable and lightweight composite constructs with functional, mechanical, and optical gradients across length scales (Duro-Royo et al. [2015], Mogas-Soldevila et al. [2014, 2015a]).

3.1.1 Water-based Design Research

Novel bio-materials design and natural aqueous formation offer in Water-based Design Research a new structural design perspective combining a crustacean-derived biopolymer with robotic fabrication to shape constructs that interact with the environment utilizing graded material properties for hydration-guided formation (Figure 3-2).

Structural, manufacturing, and environmental design templating strategies are established informing the design of constructs and their material makeup. Biomaterial-driven design process are presented here resulting in a novel structural system and a custom robotic manufacturing platform designed to deposit water-based composites unveiling novel functional, mechanical, and optical gradients across length scales. Components are form-found through evaporation patterns informed by the geometrical arrangement of structural members and the hierarchical distribution of material properties. Each component is designed to take shape upon contact with air and dissolve upon contact with water.

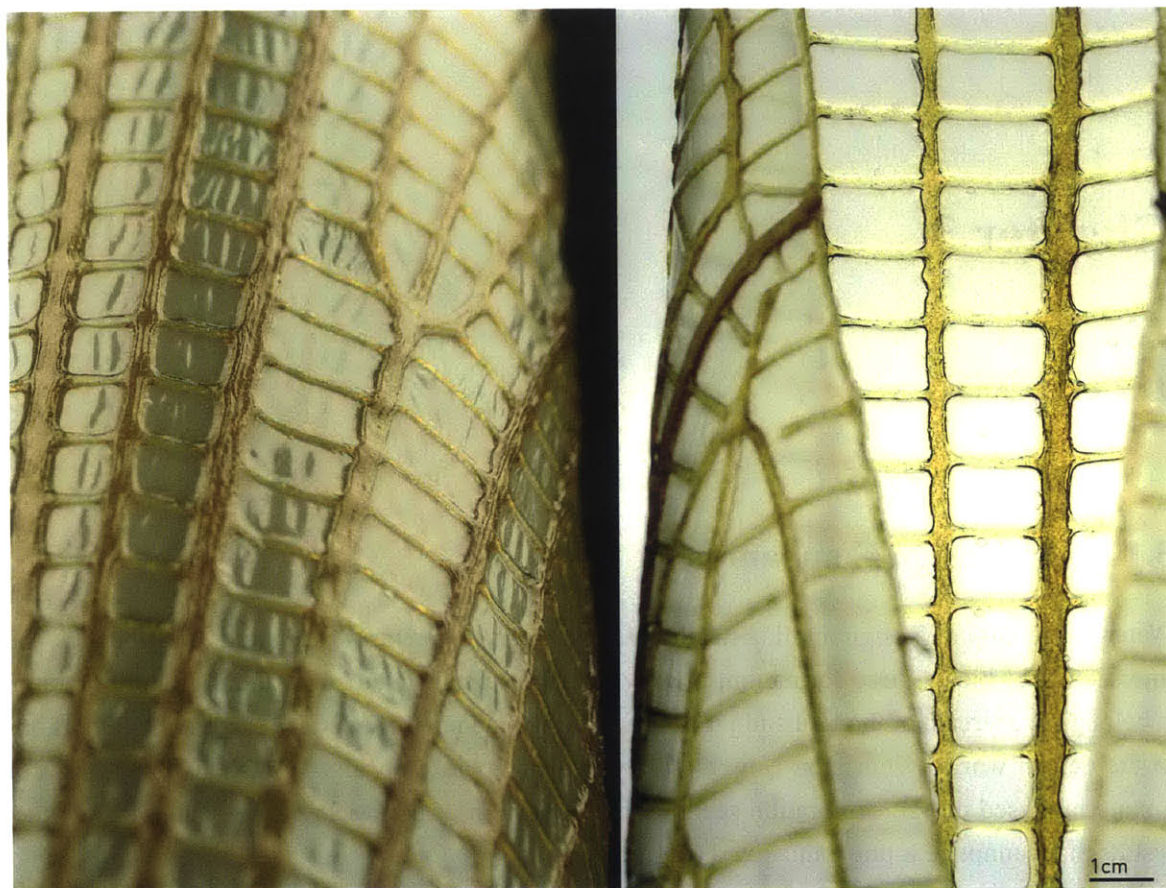


Figure 3-1: Close-up back and front images of a 3m-long hierarchically structured bio-plastic column form fabricated using a material-driven computational workflow integrating digital design with a novel robotic additive manufacturing platform.

Below are shown the principles and method applied as a unique case demonstrating the material ecology design approach through additive manufacturing of lightweight, biocompatible and materially heterogeneous structures. Initial results demonstrate a wide range of structural behaviors that represent a novel approach to material-informed biodegradable structure formation by design and hold great promise for the future of sustainable manufacturing (Mogas-Soldevila et al. [2014]).

3.2 Methods for Water-based Digital Fabrication

3.2.1 Mechanical Biomaterials Digital Fabrication

Mechanical Biomaterials Digital Fabrication is an enabling technology for additive manufacturing (AM) of biodegradable hydrogel composites through an extrusion system that can produce large-scale 3D shapes and objects omitting the need of molds. This allows achieving a wide range of geometrical forms combining varying hydrogel and aggregate concentrations in the design of functional gradients. The platform employs volume-driven extrusion combined with high-capacity repositories converging into a single nozzle. Composites can be pre-mixed and extruded, or mixed statically at

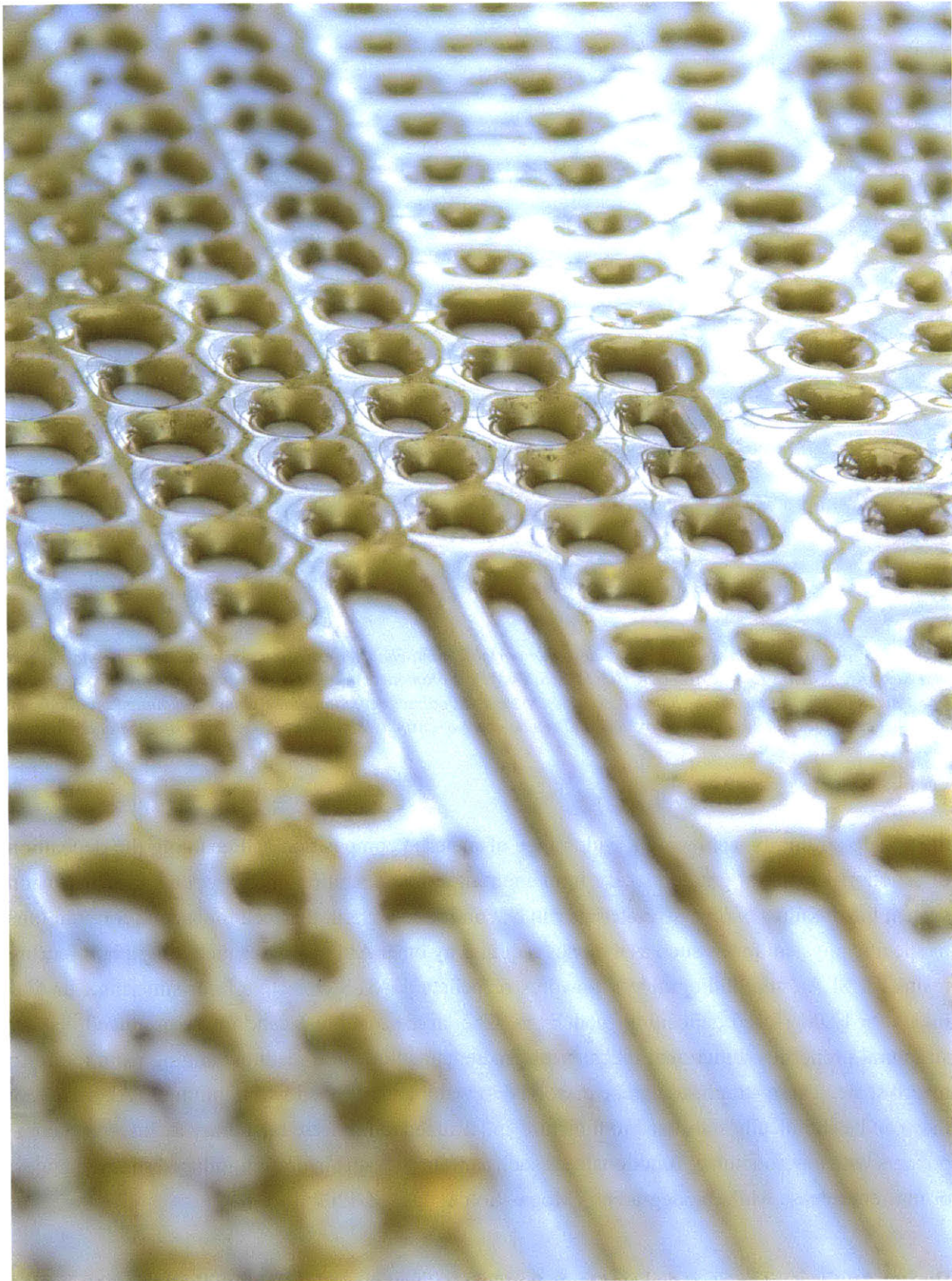


Figure 3-2: Water-based Design Research offers a new structural design perspective combining a crustacean-derived biopolymer with robotic fabrication to shape constructs that interact with the environment utilizing graded material properties for hydration-guided formation. Here a hierarchical robotic deposition of chitosan biomaterial is curing under controlled evaporation.

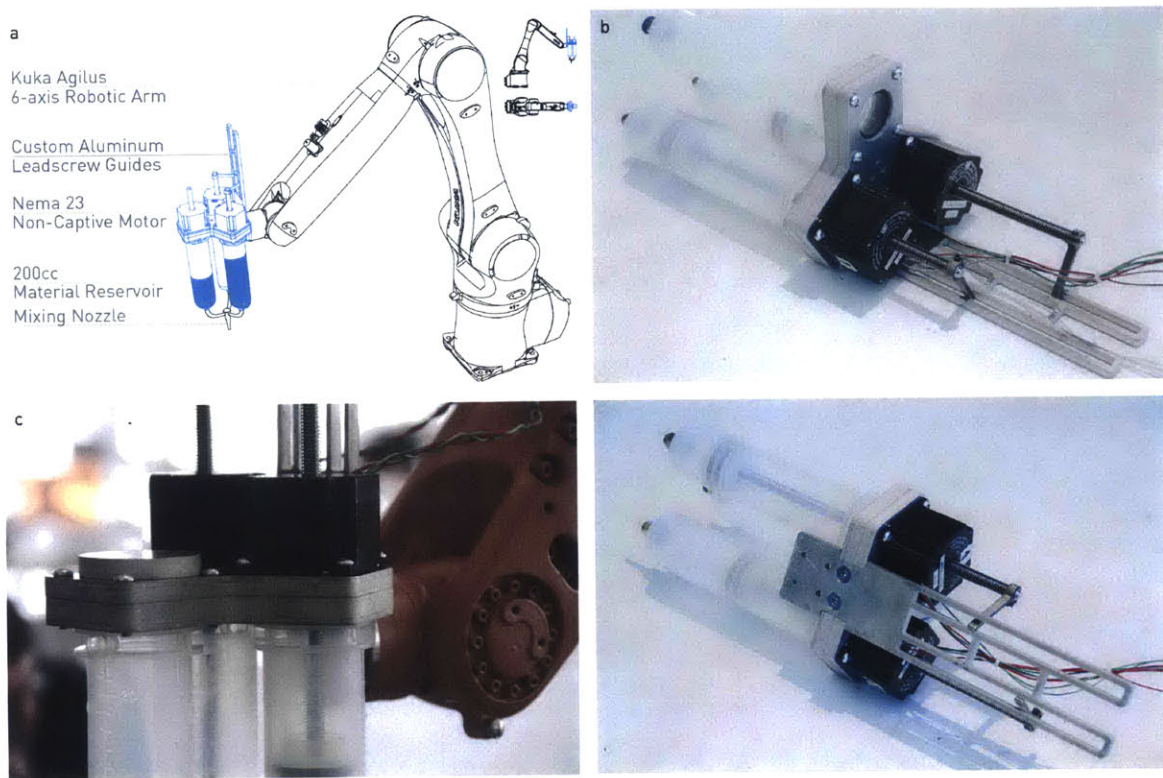


Figure 3-3: a) Experimental setup: custom-designed portable multi-chamber extrusion system mounted as end-effector of a Kuka KR AGILUS robotic arm (KR 10 R1100 SIXX WP). b) Robotic end-effector with mounted extrusion system. c) Front and side view of the three-chamber extrusion system with direct actuation of the plungers mounted on aluminum water-jet cut plates allowing for deployment and replacement of custom nozzle tips. Figure from Mogas-Soldevila et al. [2014].

the nozzle on-the-fly, achieving a wide range of material composition and enabling the deposition of functional material gradients. By implementing a low-operation energy open-ended extrusion system the mixtures of diverse materials can be controlled without the need for special formulations or specific physical forms such as the ones required by other AM processes including; photo-curing resins for SLA (stereolithography), powders or pellets for SLS (selective laser sintering), or filaments for FDM (fused deposition modeling). The large-scale bio-plastic forms produced display structural properties, functional mechanical gradients and material features ranging from micro to macro scale. These structures can be dissolved in water and recycled within minutes. Alternatively they can be chemically stabilized to moisture and utilized to fabricate temporary architectural components with environmental functionalities such as water storage, hydration-induced shape changes, or nutrient release while biodegrading (Mogas-Soldevila et al. [2014]).

Design Analysis: The designed system is portable with an overall weight of 4kg including loaded syringes and deposition material, and can be attached to an existing numerically controlled platform. Typical flow rates range from 8mm³/s to 4000mm³/s while linear motion varies between 10mm/s and 50mm/s; nozzle sizes range from 0.5mm to 8mm inner diameter. The dispensing head moves in

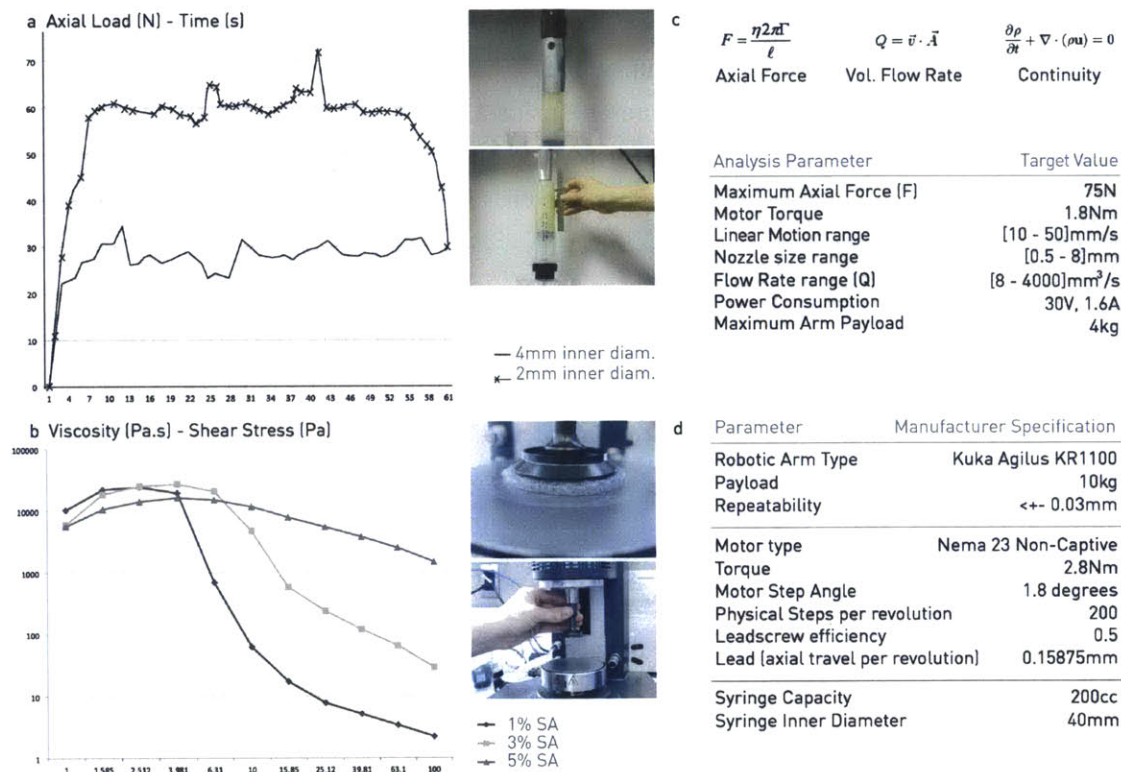


Figure 3-4: a) Colloids tested in a 200cc syringe with a 40mm inner diameter, and a 4mm and 2mm nozzle inner diameter. The sample requires an averaged axial load of 60N to display a consistent flow-rate. b) Colloids tested under AR-G2 TA Instruments viscometer display shear thinning properties that improve extrusion of the material due to the presence of sodium alginate (SA) in the matrix. SA content in the matrix is tested at 1%, 3% and 5% concentrations. c) Initial calculations and volumetric flow-rate physical testing provide insights to dimension the extrusion system for viscous colloids. d) Summary of assembly capabilities and specifications. Figure from Mogas-Soldevila et al. [2014].

three dimensions, within the robotic arm envelope, while the fabrication platform is stationary. The dimensional limits of the process, when using a Kuka Agilus KR1100, are 1000mm in length and 500mm in width in order to deposit rectangular constructs. The portable extrusion system can be mounted on robotic arms with a larger envelope or other CNC machines. Figure 3-3 demonstrates the mechanical experimental setup (Mogas-Soldevila et al. [2014]).

Dimensioning Tests: Key experiments executed in the design of the extrusion system where performed using an Instron mechanical tester and a shear measurement AR-G2 viscometer. From the initial testing, a 60N axial load is required to extrude the colloids from a 200cc syringe with a 0.7mm nozzle, and a motor torque is calculated for direct drive of 1.8Nm. In terms of travel speed a maximum robotic arm end-effector displacement of 50mm/s is required in order to ensure proper extrusion of the materials (Figure 3-4a, Figure 3-4c). Colloids tested under shear rates of 1Pa to 100Pa over 10min display properties that favor extrusion of the material, due in part to the presence of the sodium alginate additive in the chitosan matrix [16]. These favorable properties are due to the enhanced shear thinning behavior that sodium alginate provides, where the viscosity of the fluid decreases when the shear stress applied is increased. This change in the properties of the material

is caused by shear-induced reorganization of the polymer chains to a more stretched conformation, which leads to decreased entanglement. For fabrication purposes, this implies a decreased shear stress at the high shear rates that are present inside a nozzle, followed by a sharp increase in viscosity upon deposition, facilitating the extrusion and printing fidelity of the viscous matrix [4], [16]. Concentration of sodium alginate of 3% w/v is chosen for sample production in Figure 3-4b (Mogas-Soldevila et al. [2014]).

Experimental Setup:

1) Existing Motion Platform: The extrusion system is attached to an existing motion platforms end-effector, a Kuka Agilus 6-axis robotic arm, in order to take advantage of its high precision displacement and repeatability capabilities.

2) Extrusion System Design: The extruder design is composed of two chambers utilizing plunger actuation for viscous colloids and an additional chamber to handle dry or granular dispensing of natural fillers such as cellulose chopped fibers, fine sand, or natural polymer powders (Figure 3-3c). Bipolar stepper motors with non-captive lead-screws (Figure 3-4d) linearly advance the materials to the tip by pushing the syringe plunger. The motors are mounted to CAD-CAM water-jet cut aluminum frames that provide hardware support and vertical guides to prevent the lead-screws from turning, thereby transmitting the rotor actuation force to linear motion.

3) Nozzle System Design: The system provides for the nozzles to be custom designed to step the 8mm reservoir opening down to different diameters and material configurations for extrusion. For the samples presented here 3D-printed single nozzles are designed with an inner diameter of 0.5mm, 0.7mm, 2mm, 3mm and 4mm that can be mounted at the end tips of other complex nozzles. A three parallel chamber nozzle is designed with 2mm final inner diameter, a tri-axial extrusion nozzle with 3mm final inner diameter, and a three material static mixing nozzle with 3mm final inner diameter. Nozzle schematics are displayed in Figure 3-5.

Control and Operation: The desired objects are modeled in a computer-aided design platform and custom code is written to slice surface-based geometry into layers, build a line-based tool-path from curves, and stream the tool-path steps to a central interface in the computer. The central interface computes input and output data from the design platform and to and from the mechanical parts of the system (robotic arm and extrusion stepper motors). Various machine paths are tested and evaluated for extrusion consistency and mechanical extrusion behavior starting with straight lines and moving into cylindrical paths and layered prismatic depositions. The tool-paths are processed and transmitted to the robotic arm via a server socket, and to the motors via serial signal. Both output signals are synchronized for smooth control. The robotic arm sends back actual positions that can be compared with the targets sent for consistent coordinated feed / speed ratios. The interface takes into account forward and reverse motor drive to prevent material dripping. A stepper motor transmission code is developed that receives data from the central interface and sending signals to two driver boards that are set up to run the two-stepper motors simultaneously thereby enabling variable

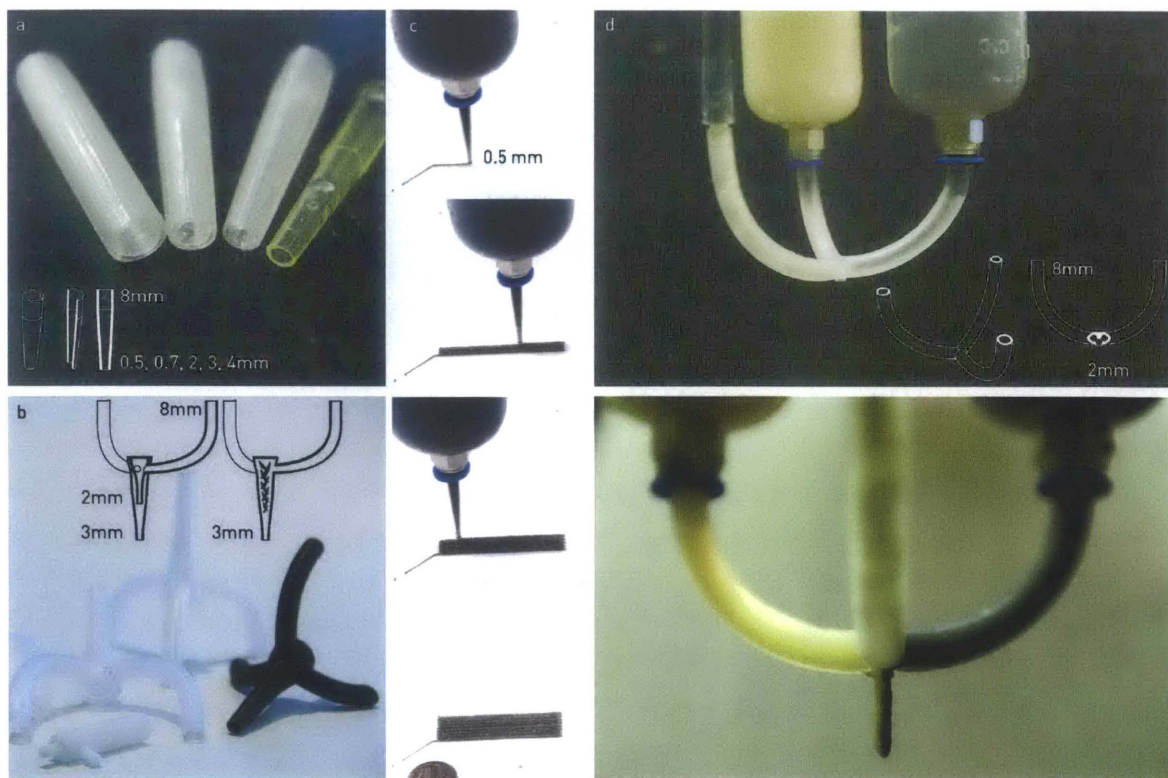


Figure 3-5: a) Single nozzle designs and tips. b) Triaxial and static mixing nozzle designs. c) Continuous extrusion with a single nozzle with 0.5mm feature size. d) Parallel material extrusion nozzle depositing colloids mixed with white and black food coloring. (Nozzle line drawings indicate inner diameter measurements, are superimposed in their corresponding nozzle design photographs, and represented in perspective and section view) Figure from Mogas-Soldevila et al. [2014].

extrusion rates and mixing ratios. For the motor control hardware a micro-controller board is used and bipolar drivers are powered through an external bench power supply at 30 V and 1.6 Amps in correspondence with the motor specifications. Robotic movement and extrusion are not explicitly linked, which allows for independent control of each motor-driven syringe, and the displacement of the robotic arm end-effector. With the ability to vary each of these parameters individually in succession, there is more flexibility in the development of the tool as an experimental fabrication platform (Mogas-Soldevila et al. [2014]).

Mechanical Assembly: The extrusion system is attached to an existing motion platforms end-effector. The existing platform is a Kuka KR AGILUS robotic arm, model KR 10 R1100 SIXX WP. The platform weighs 54kg with a 10kg payload and a maximum reach of 1101mm. It has 6 axes, a ± 0.03 mm repeatability and employs the KR C4 compact control system. The stepper motors are equipped with non-captive Acme lead-screws with 9.5mm diameter and 635mm length. The single nozzle assemblies use Oxford Universal plastic pipette tips of a 0.7mm, 1mm and 2mm inner diameter at the tip. The complex nozzle fabrication use the Formlabs Form1 SLS (selective laser sintering) 3D-printer with clear photo curable resin, 0.5mm layer height, no internal support and high exterior support density for maximum channel resolution to minimize extrusion calculation

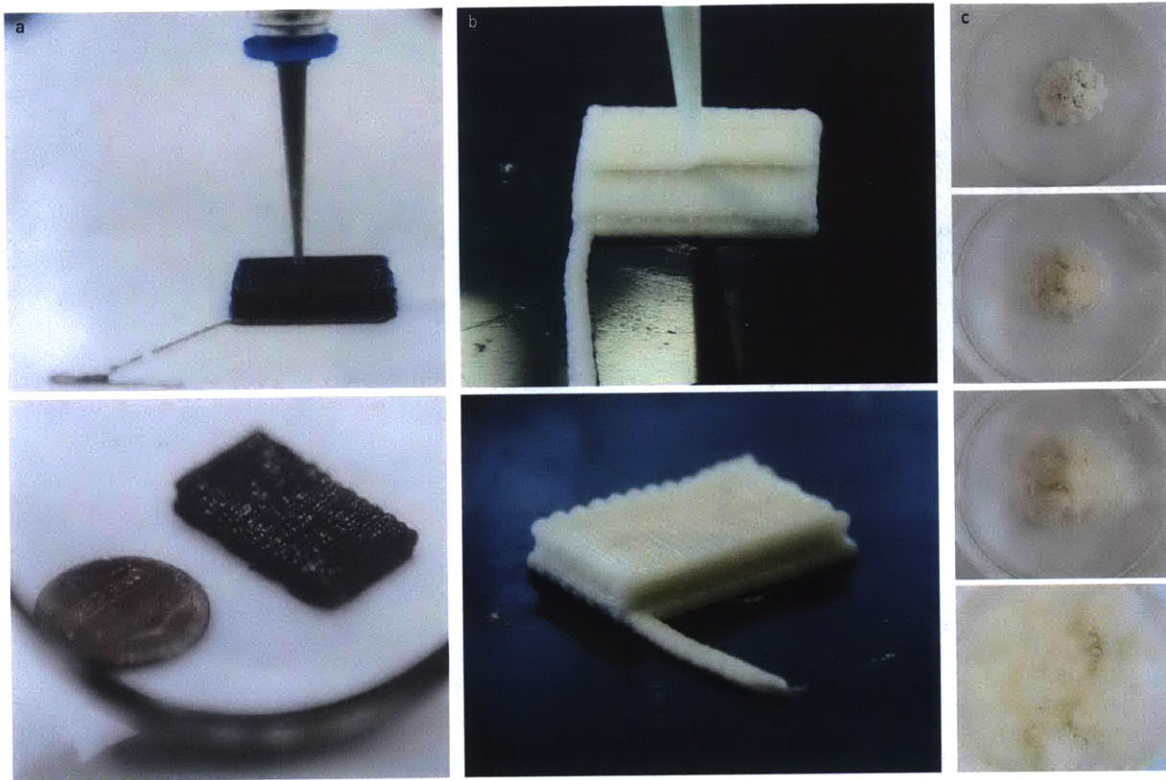


Figure 3-6: (columns a, b, c). a) A 2x1x0.5cm prism printed using a 0.7mm nozzle at 20mm/s printing speed to produce unidirectional layer orientations. The result is shown under a 1.5 augment magnifying lens in bottom column. b) A 2x1x0.5cm prism printed using a 0.7mm nozzle at 20mm/s speed to produce bidirectional layer orientations. The result is shown in bottom column. c) A printed 15g dried chitosan-based cone shaped composite is dissolved at room temperature in 200ml of water over 20min, demonstrating colloids biodegradability. Figure from Mogas-Soldevila et al. [2014].

error due to material friction at the nozzle. The mounting of the mechanical parts uses 5mm and 10mm aluminum machinable sheets cut with a numeric control abrasive water-jet cutting machine (OMAX Corporation, USA) (Mogas-Soldevila et al. [2014]).

Hardware: A Mastech Linear DC power supply, model 30V 5A HY3005F-3, with triple outputs and dual adjustable outputs (0-30V and 0-5A) is used to power two bipolar motor stepper drivers Mondo Step 4.2, with an output current of 1A to 4.2A, a supply voltage of 20VDC to 50VDC and a step frequency of 0 to 300Khz. The bipolar non-captive linear actuators Haydon 57F4A, have a step angle of 1.8 degrees, a linear travel per step of 0.0079375mm and a force versus linear velocity of 1200N at speeds below 1mm/sec. The board where the system is controlled from an Arduino Mega 2560, with a computer supplied input power of 5V and an output power of 3.3V or 5V, incorporating a high performance, low power Atmel AVR 8-Bit micro-controller (Mogas-Soldevila et al. [2014]).

Software: Geometric tool paths providing control and operation of the extrusion system are designed with Rhino3D modeling software (RHINOCEROS, Robert McNeel and Associates, USA) and its scripting plugin Grasshopper. Using the geometric kernel library of the plugin, custom

C-sharp code is written to transmit fabrication instructions to the central interface. The central interface is written in C++ using the Eclipse IDE environment (2013 The Eclipse Foundation) which computes input and output data from the design platform to and from the mechanical parts. Transmission is achieved with an Ethernet UDP socket and via a serial USB signal. The stepper motor transmission code is developed in C code using the cross-platform open-source Arduino IDE (by Arduino Software) (Mogas-Soldevila et al. [2014]).

Initial Results in Layered Depositions: Initial material bonding and flow-rate consistency tests were performed resulting in simple prisms (Figure 3-5c). Constructed 2x1x0.5cm prisms with a single nozzle setup, display minimum feature sizes of 0.7mm and 1mm measured with a digital caliper of 0.0254mm accuracy. Unidirectional and bidirectional toolpaths were tested with significant improvement in layer bonding in the bidirectional samples (Figure 3-6a, Figure 3-6b). Geometrically patterned tool-paths with 0.7mm thickness, displaying structural inertia when dried and detached from the substrate, were designed in anticipation of future development for object infill (Mogas-Soldevila et al. [2014]).

Initial Results in Parallel Depositions: Tests with a three-chamber nozzle (using only 2 material chambers) display 2mm feature size when two colloids with different coloration are extruded from the two actuated syringes. Figure 3-5c displays successful parallel extrusion of the materials. This interesting feature could be used in future work to deposit dual material geometries with different internal and external properties. This could avoid boundary de-lamination issues between the two materials as the colloids are bonded together while extruded through the multi-material chamber at the nozzle. All samples are composed of chitosan-alginate matrix and chitin powder aggregate with white and black food coloring for visual contrast (Mogas-Soldevila et al. [2014]).

Initial Results in Graded Depositions: Material mixing at the nozzle provides for mechanical property variation within the samples, as well as for controlled material gradients with the intention of mimicking spatial organization and material composition strategies found in Nature. Figure 3-7a displays the deposition process and result of a 30cm long design modeled after the maple seed. The structure obtains its inertia from chitosan-alginate reinforcement lines that follow the structural bending diagram of the construct. The clear color and tensile strength of the infill films are provided by a chitosan 3% interstitial filling. Figure 3-7b illustrates the deposition process and result of a 50cm long dragonfly wing design. The structure obtains its inertia from the cell pattern of chitosan-alginate reinforcement paths, and its tensile strength from chitosan 6% interstitial films. Deposition of chitosan concentrations is achieved from 1% to 10% w/v in 1% acetic acid solution through static mixing at the nozzle. When mixing the gels with a sodium alginate thickening agent, as well as viscosities with deposition-compatible properties still displaying the optical density qualities of the gels. In Figure 3-7c the deposited range contains a gradient of concentration and of thickening agent; from 1% chitosan and 10% sodium alginate, to 10% chitosan and 1% sodium

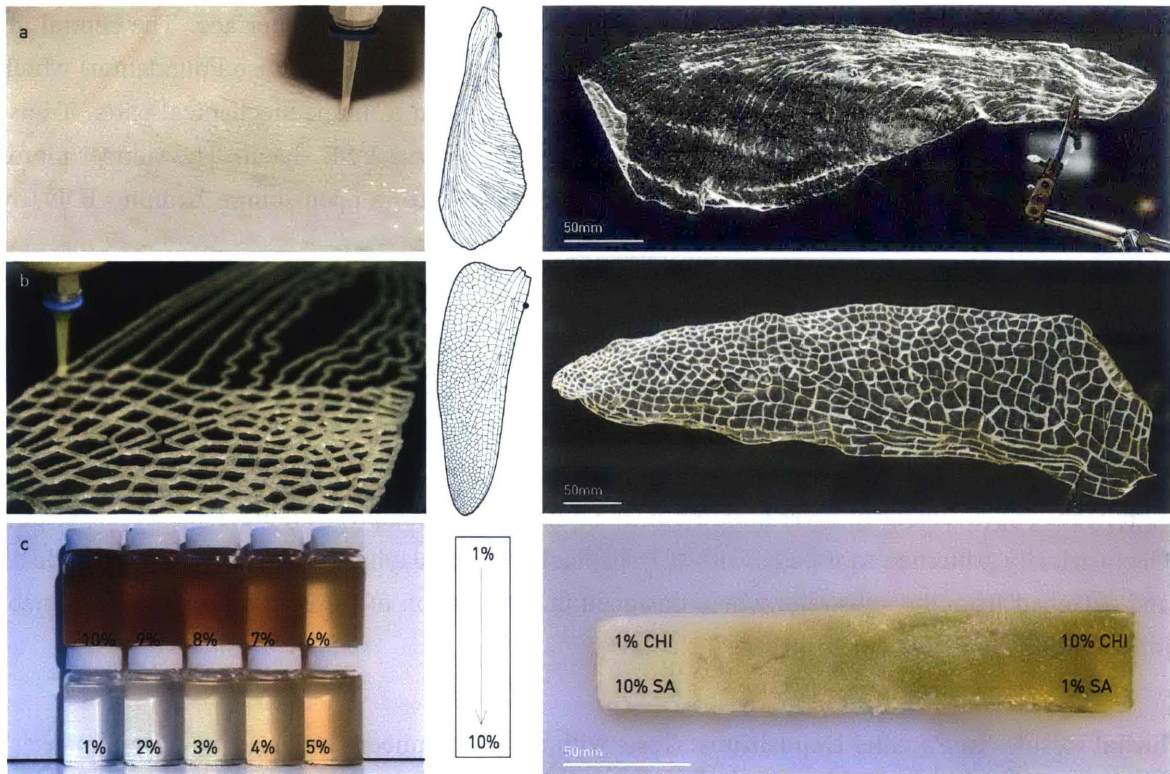


Figure 3-7: (rows a, b, c). a) Water-based deposition of a 30cm long material patch modeled after the maple seed. The patch exhibits reinforcement trajectories aligned with the anticipated structural bending. b) Water-based deposition of a 50cm long material patch modeled after the dragonfly wing. The patch exhibits structural inertia. c) Chitosan concentrations range from 1% to 10% w/v in 1% acetic acid solution. Gradient chitosan concentrations are statically mixed at the nozzle with sodium alginate as thickening agent. Both sodium alginate content and chitosan concentration differences also provide for optical density variation. The gradient ranges from: 11% aggregate; 1% chitosan (CHI) and 10% sodium alginate (SA), to 10% chitosan (CHI) and 1% sodium alginate (SA). Figure from Mogas-Soldevila et al. [2014].

alginate (Mogas-Soldevila et al. [2014]).

Initial Results in Biodegradability: I have evaluated that a 15g conical prototype of 30mm diameter base and 30mm height put to dry overnight, will dissolve in 200ml of water at room temperature after 20min (Figure 3-6c). The samples can have longer stability to moisture by submerging them in a 4% NaOH w/v aqueous solution for 5min once dried, and then dried again overnight. Nevertheless, chitosan-alginate composites will biodegrade over time when exposed to the environment. This capability can be used to design temporary and highly ecological architectural-scale parts that can interact with the environment by contributing to soil nutrient levels over time during decay. Plant growth on a 40mm diameter and 8mm height disc of mixed chitosan and plant soil is tested with grass seeds growing over a week with no loss of disc consistency or integrity (Mogas-Soldevila et al. [2014]).

I have presented Mechanical Biomaterials Digital Fabrication as the combination of natural ag-



Figure 3-8: Perspective view of the manufacturing platform installed at the MIT Media Lab composed of (1) a positioning robotic arm (Kuka KR AGILUS robotic arm, model KR 10 R1100); (2) a customized multi-barrel extrusion head for water-based materials; (3) a security envelop for operation of the robotic assembly; (4) a 1m-wide and 5m-long deposition bed; (5) a computer-controlled evaporation system composed of 100 fans; and (6) a 9m-long workspace and storage cabinet. Figure from Mogas-Soldevila et al. [2015a].

gregates with chitosan and sodium alginate hydrogel matrixes with other organic aggregates, to deposit biodegradable prototypes using mechanically actuated extrusion methods. The approach is demonstrated by building and testing the mechanics and controls of a multi-chamber portable extrusion system that can be attached to numeric control multi-axes platforms. The technology was developed in collaboration with Jared Laucks, Arthur Petron, Markus Kayser, Steven Keating and Jorge Duro-Royo. The utility of the system is demonstrated by fabricating depositions of natural composite-material objects. Preliminary results display consistent volumetric flow-rates, sub-millimeter to macro-scale features, and graded properties. Water-based Robotic Fabrication holds potential applications for fully recyclable products or architectural parts with graded mechanical, optical and environmental properties such as water storage, hydration-induced shape change, or full disintegration over time.

The approach and enabling technology presented contributes to the field of sustainable additive manufacturing of biodegradable material-systems. The technology enables the deposition of functionally graded constructs with structural hierarchies made of basic building blocks and processed via water and low impact chemicals. The technology enables the deposition and curing of constructs at room temperature, with no external energy sources other than mechanical extrusion and displacement of the nozzle.

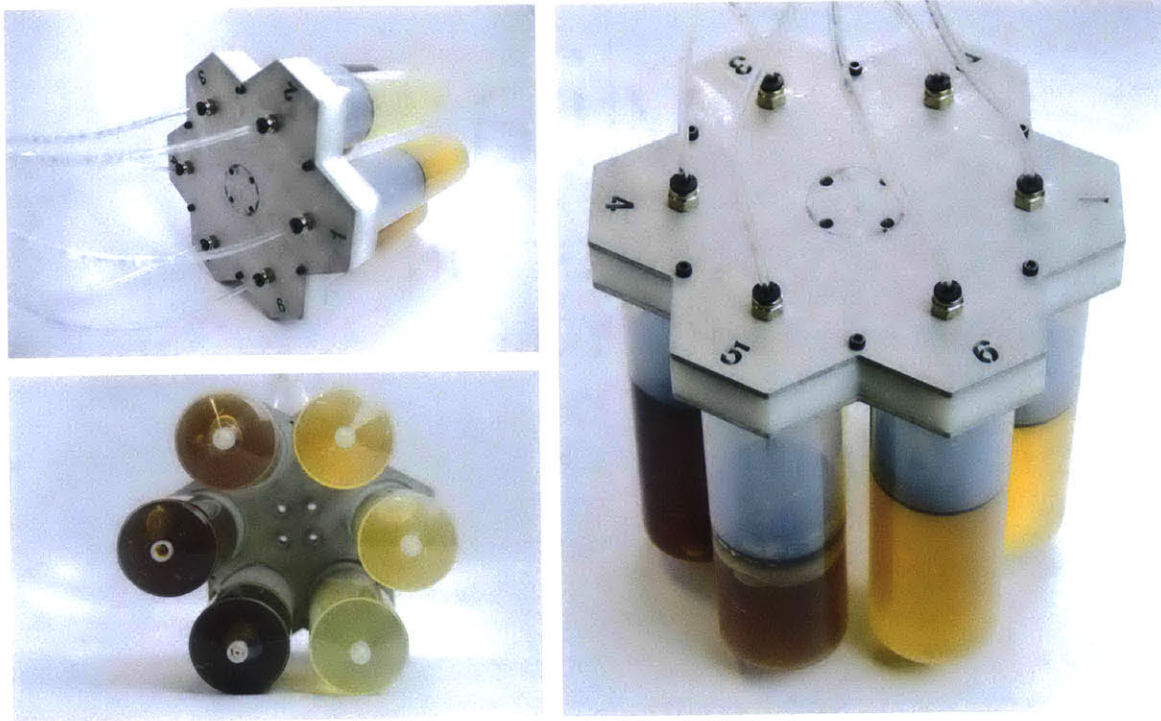


Figure 3-9: Customized multi-barrel extrusion head for diverse viscosity water-based materials.

3.2.2 Pneumatic Biomaterials Digital Fabrication

Further research is presented in this section that improves the mechanical fabrication platform prototype by designing a pneumatic extrusion system (using air pressure and solenoid valves), instead of the presented volume-driven system (using stepper-motors), that provides for the generation of more complex extrusion paths. In terms of further material explorations: cellulose chopped fibers provide enhanced mechanical properties of printed objects. In terms of advanced controls: mechanical property prediction models should be integrated to inform the variations of geometrical and chemical design of the objects. Figure 3-8 illustrates the designed pneumatic-driven manufacturing platform with fully synchronized fabrication parameters for position, speed and air pressure via material property constraints (Duro-Royo et al. [2015]). The platform is composed of an existing 6-axis robotic arm (Kuka KR 10 R1100) as a positioning system operated via Ethernet instructions (Figure 3-8(1)); a customized pneumatic deposition platform operated via serial instructions carrying 6 barrels of different structural capacity hydrogels (Figure 3-9, Figure 3-8(2)); a security envelope for operation of the robotic system (Figure 3-8(3)); a 1m-wide and unlimited length print bed (Figure 3-8(4)); a computer-controlled evaporation system (Figure 3-8(5)); and finally a 10m-long workspace and storage cabinet (Figure 3-8(6)). The water locked within the polymeric materials allows for total self-bonding as well as self-repair of layers and sections of the print. Consequently, structures span a sliding print bed with virtually-infinite longitude allowing for structural construct generation beyond the robotic arm reach, which overcomes the gantry size limitation of current additive manufacturing technologies (Oxman [2011]).

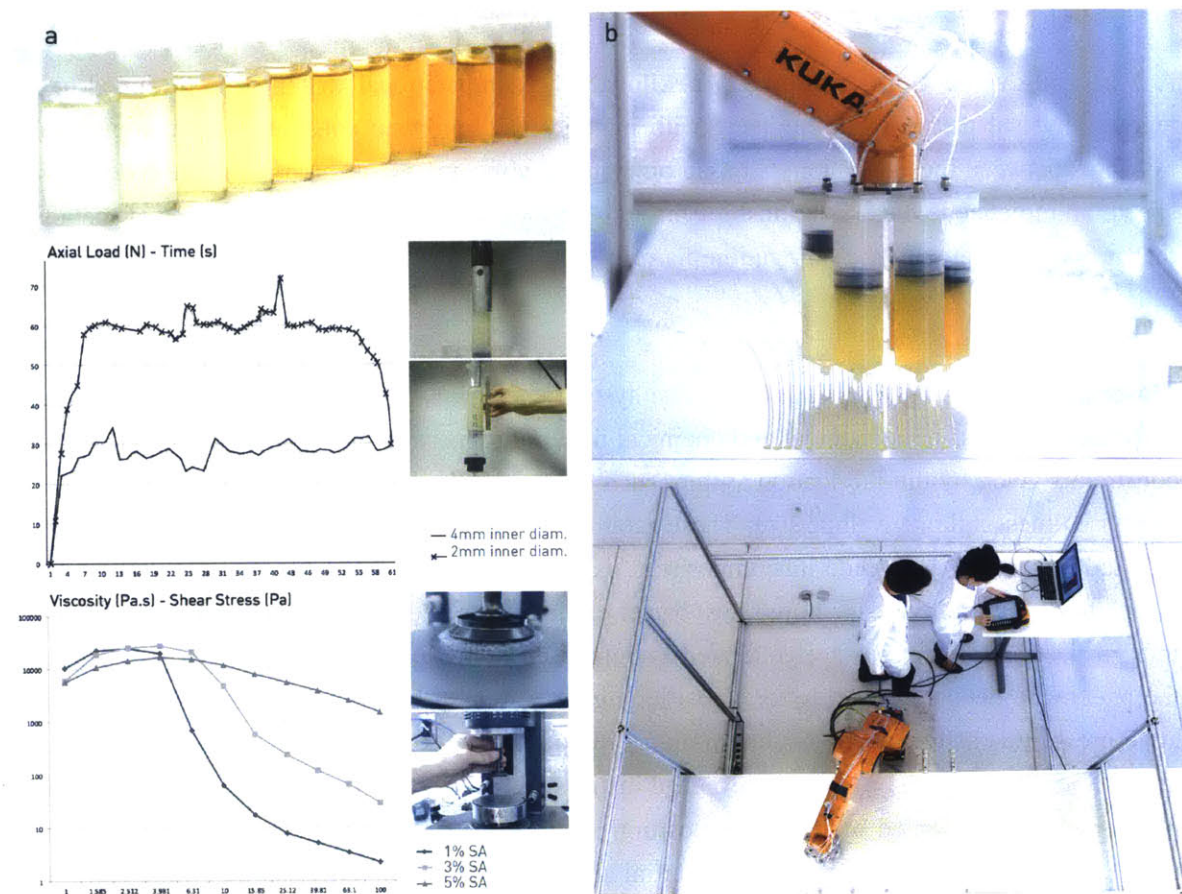


Figure 3-10: (a) Hydrogel material gradients vary from 1% to 12% concentration of biopolymer in water. Material characterizations such as axial load and shear rate are used to inform hardware design and software implementation. (b) The hardware platform is composed of a custom multi-barrel deposition system and a positioning robotic arm. Figure from Mogas-Soldevila et al. [2015a].

Hardware: The pneumatic assembly is designed to additively fabricate large-scale structures made of water-based materials. It is composed of six large-capacity clear plastic barrels containing hydrogels of different viscosities and is attached to the end-effector of an existing 6-axis robotic arm, connected to a positive and negative air pressure system. Both pneumatic assembly and robotic arm are digitally controlled to tune pressure and speed on-the-fly based on desired extrusion geometries and material properties (Figure 3-10b) (Mogas-Soldevila et al. [2015a]).

Software: An integrated design workflow is implemented in the C# language within the Rhinoceros3D platform; in C++ language as a stand-alone applet for mechanical synchronization of deposition and positioning platforms; and as custom firmware loaded into the deposition electronic assembly. Instructions generated by the software are implemented to achieve full synchronization of deposition and positioning, including the incorporation of material metadata with direct feedback to and from hardware (Figure 3-11a) (Mogas-Soldevila et al. [2015b]).

A Material-driven Computational Workflow: The computational workflow is designed to integrate physical parameters associated with material and hardware constraints, to compute relationships between these parameters based on material viscosity, extrusion pressure, and deposition speed; and to encode these relationships through customized instructions in the forms of metadata that is communicated to the fabrication platform (Duro-Royo et al. [2015], Mogas-Soldevila et al. [2015b]).

Material-driven Property Variation: The system encodes an array of independent parameters associated with basic mechanical and chemical material properties, as well as platform-dependent constraints given by the fabrication system such as viscosities; shear rates, barrel types, hardware response times, and/or envelope size. These parameters are then combined with design-specific variants such as nozzle type, material composition, as well as time-dependent pressure maps. Resulting calculations output necessary flow-rates, barrel refill patterns and positioning speeds. Finally, custom fabrication instructions are generated as output, encoding motion and extrusion commands to the pneumatic deposition and robotic positioning systems respectively. Computer-controlled negative pressure is achieved via a vacuum pump (v), positive pressure via a compressed air tank (c) and a digital PSI regulator (r). Synchronized instructions given by both deposition and positioning platforms are represented by the home position in 3D space (H), motion (M), extrusion initiation values based on material properties (E), partial and final position targets including time delays (T) and barrel refill pattern (R) (Figure 3-11a left) (Mogas-Soldevila et al. [2015b]).

Material-driven Hierarchical Structuring: Hierarchical structuring of printed components is obtained across three levels of resolution associated with the additive manufacturing process (Figure 3-11a, right, Figure 3-11b): At the first level of resolution, material is distributed locally through varying layering strategies and stiffness gradients. At the second level of resolution, material is extruded in longitudinal regions with variable extrusion height and width associated with pressure data given by the deposition platform and the nozzle (Figure 3-11b). Finally, at the third level of resolution, global topologic effects are obtained across the entire construct through geometrically patterning the overall shape (Figure 3-11a, right). It is important to note that local layering, regional extrusion gradients, and global geometric patterning are designed and fabricated in 2-dimensions (2D). Specifically, designs are not modeled in 3D; rather, they are composed of 2D extrusions that are associated with, and informed by, material properties, robotic arm speed and nozzle speed. Depositions of material hierarchies with varying stiffness are extruded flat on a two-dimensional substrate (Mogas-Soldevila et al. [2015b]).

Material-driven Shape Formation: Hydration guided shape formation is informed by both intrinsic constraints (relating to material properties, e.g. elastic modulus) and extrinsic constraints (relating to the environment, e.g. thermal maps). Designed and digitally fabricated constructs are

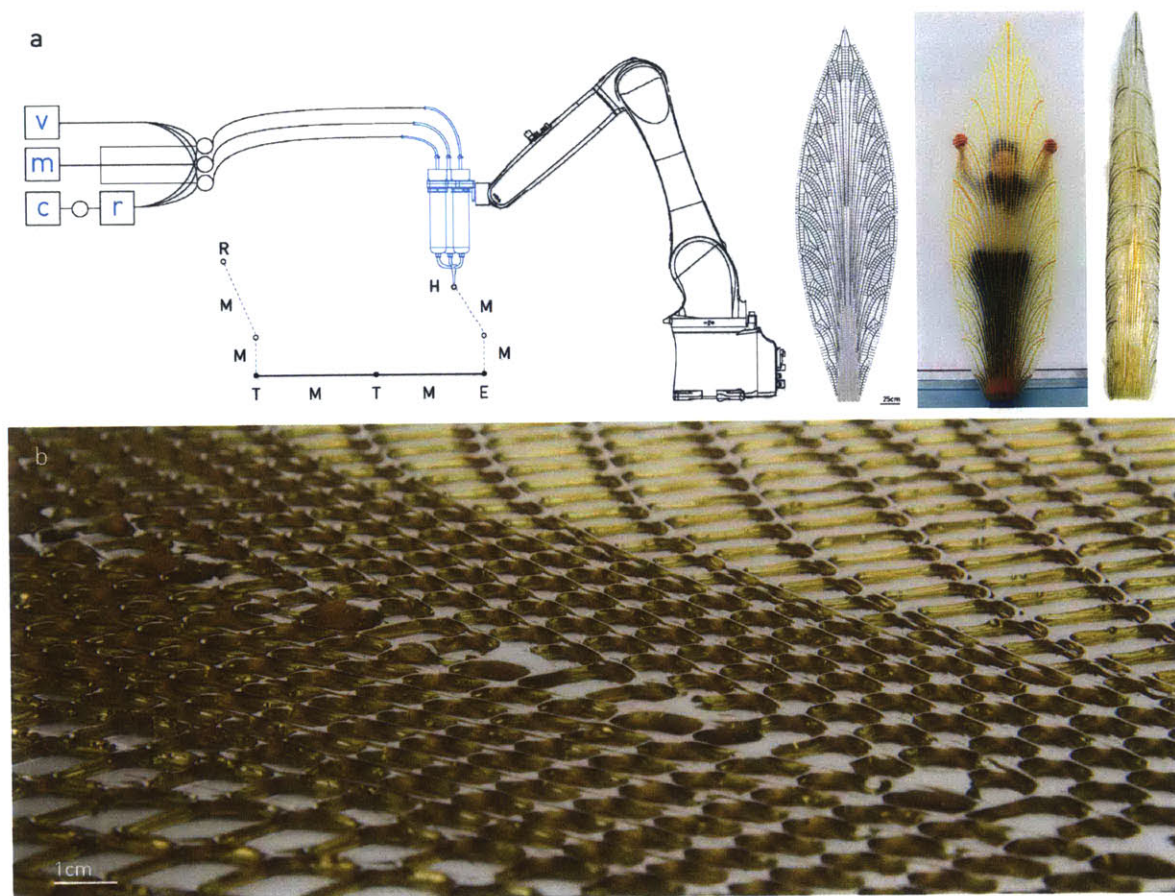


Figure 3-11: (a) Motion-extrusion instructions schema: a pneumatic assembly for material deposition is designed as a multi-chamber nozzle and attached to the end-effector of an existing positioning platform (Kuka KR AGILUS robotic arm; model KR 10 R1100 SIXX WP). (b) Close-up image of a hierarchically structured chitosan-based deposited construct. Figure from Mogas-Soldevila et al. [2015b].

left to dry flat, overnight, while positioned under a tunable fan array. Figure 3-11 a (right) displays a two-dimensional (2D) digital drawing, its fabrication protocol including structural hierarchies on three levels of resolution, and the final three-dimensional (3D) structure; a self-assembled column formed during drying. During the drying process, internal evaporation-induced stresses accumulate within the printed structures of the flat material (Figure 3-12a). Once removed, the constructs undergo folding as remaining moist content evaporates from their surface. Shape is therefore found or formed based on computationally templated material and environmental effects incorporated into the workflow. Figure 3-12b displays 2D-to-3D formation induced by digitally patterning material properties in a medium-scale structured grid removed from its substrate at an intermediate hydration stage. Additional research on predictive modeling of 3D shaping is currently under way to further advance the digital design tool by incorporating immediate simulation of 2D hierarchical designs (Mogas-Soldevila et al. [2015b]).

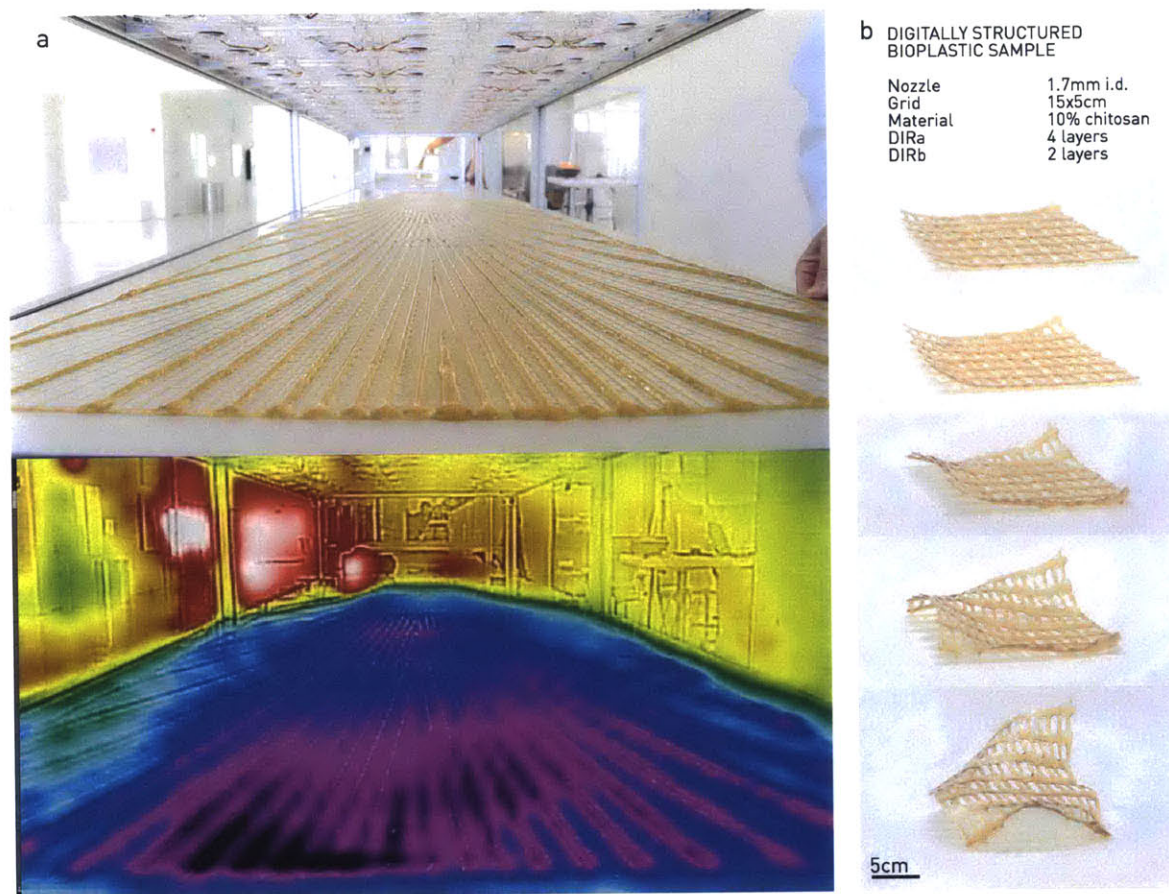


Figure 3-12: a) Internal stresses generated by natural evaporation contribute to the shaping of each structural component. (b) 2D-to-3D shaping stages of an additively manufactured structured chitosan grid removed from the substrate at an intermediate state of curing (left to dry and form-found within 12 hours). Figure from Mogas-Soldevila et al. [2015b].

3.3 Methods for Water-based Materials Engineering

Bottom-up bio-materials chemical design exploration focuses on chitosan, the second most abundant natural polymer on earth. Chitosan is a polysaccharide derived from chitin (Fernandez and Ingber [2014]). Chitins chemical structure is similar to cellulose and can be found in exoskeletons of insects and crustaceans as well as in some fungi (Vincent [2012]). The non-toxicity, solubility, capacity for swelling, and chemical versatility of chitosan allow for the development of functionalized materials that can be used in multiple scales such as drug delivery, tissue scaffolding, consumer products or architectural parts (Mogas-Soldevila et al. [2014], Fernandez and Ingber [2014], Billiet et al. [2012], Malda et al. [2013]).

For the purpose of this research, chitosan powder 85% de-acetylated and acetic acid are purchased from Sigma-Aldrich. Chitosan powder is processed into gel at 3%, 6%, 9%, and 12% w/v concentrations using 4% w/v acetic acid in aqueous solution. A concentration of 3% has a translucent appearance and displays similar consistency to watery honey. A concentration of 12% has an opaque appearance and displays the consistency of natural rubber.

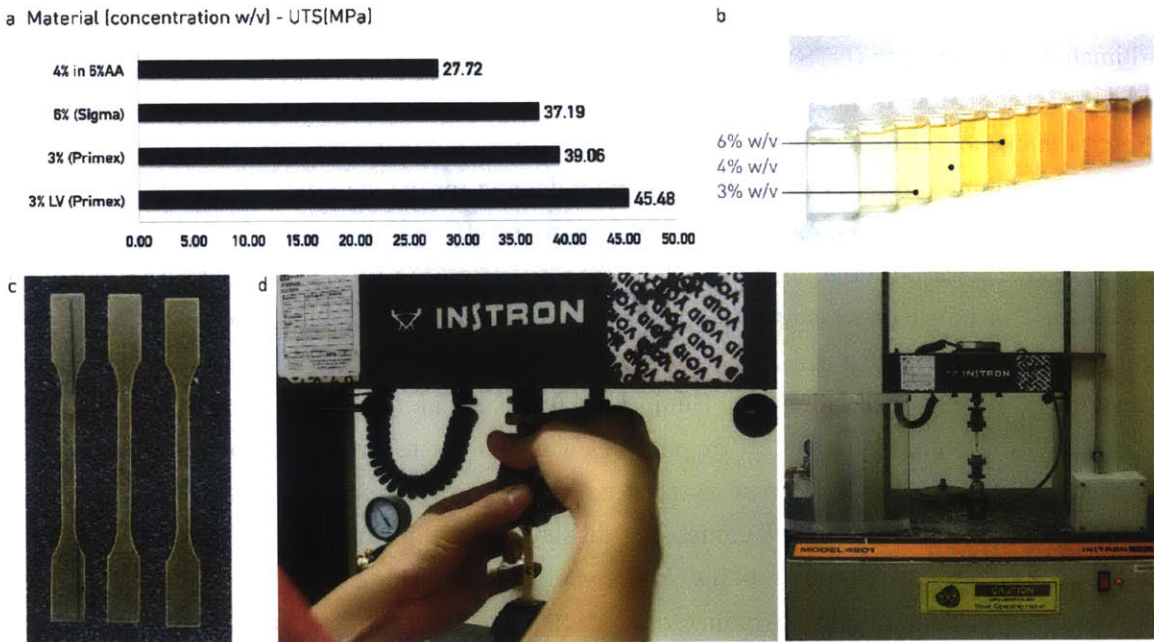


Figure 3-13: (a) Ultimate Tensile Strength (UTS) testing results of chitosan film dog-bones in different concentrations and purchased from different companies (b) Chitosan concentrations in gel state (c) 3D printed and laser cut dog-bones before being tested (d) Instron mechanical tester with a 9071.85kg reversible load cell. Figure from Mogas-Soldevila et al. [2015a].

The materials are characterized using a 200cc syringe with a 40mm inner diameter, and a 4 and 2mm nozzle inner diameter under an AR-G2 TA Instruments viscometer and an Instron mechanical tester with a 9071.85kg reversible load cell. Extensive empirical testing results demonstrate that a maximum axial load of 75N and maximum linear plunger motion of 50mm/s present a consistent flow rate with the different gel viscosities (Mogas-Soldevila et al. [2014], Mogas-Soldevila and Oxman [2015]). Such material-driven parameters are used to dimension and design the presented enabling technology for large-scale digital fabrication of chitosan gels.

Preliminary testing of deposited materials shows promising 40MPa UTS (Ultimate Tensile Strength) comparable to other 3D-printing filament materials such as Nylon, ABS (Acrylonitrile Butadiene Styrene) or PLA (Polylactic Acid). To perform UTS testing, deposited chitosan materials in different concentrations (3%, 4% and 6% w/v in 4% acetic acid in aqueous solution) and sourced from different manufacturers (Sigma Aldrich and Primex) are dried overnight into 0.5mm thick films and laser cut into dog-bones that are tested under an Instron machine (Figure 3-13).

Further research is being conducted in chitosan-based composites incorporating glycerin plasticizers for increased flexibility, as well as fibrous and granular fillers in order to generate components with compression and bending capabilities (Mogas-Soldevila and Oxman [2015]).

3.3.1 Composites Design Catalogue

The objects presented in this section, are fabricated and cured at room temperature, dramatically reducing the need for external energy sources other than air-actuated extrusion and displacement of

the nozzle head [16, 18, 19]. Initial results of deposited and dried chitosan bio-plastic films show an ultimate tensile strength (UTS) of 40mPa comparable to nylon or wood in the direction of the fiber (Figure 3-15d) (Mogas-Soldevila and Oxman [2015]).

Natural thickeners such as sodium alginate powder can be incorporated into chitosan gels to generate shear thinning effects in the material barrel, allowing for the materials to extrude easily under shear forces, and to become more viscous once extruded effectively defining the extruded geometry (Figure 3-15c). Various other fillers such as sand, soil, hemp fiber, seeds, calcium carbonate, sodium bicarbonate or polyacrylamide storing crystals, have been combined with chitosan and sodium alginate hydrogels (Figure 3-15b). Such structures display integrity after drying due to the binding capabilities of polysaccharide gels. Hemp long fibers, sodium bicarbonate, and polyacrylamide chitosan-based composites display irregularities, material heterogeneity, and pose extrusion issues due to the difficult control of chemical reactions or filler distribution at the nozzle. Sand, soil, and calcium carbonate chitosan-based composites display sub-millimeter surface resolution when dried due to the dense packing of their grains (Mogas-Soldevila and Oxman [2015]).

In future research other water-based materials can be used that are stored in air-tight extruder barrels at room temperature and that cure at room temperature by natural evaporation once out of the barrel. For instance, organic resins, polyvinyl alcohols, clay, plaster, calcium sulfate and calcium carbonate compounds in concrete. These materials can also be combined with granular or fibrous fillers. In future iterations, the platforms end-effector could be replaced with a heated nozzle and a filament material feed to extrude thermoplastics typically used in fused deposition modeling (FDM) such as poly-lactic acid (PLA), acrylonitrile butadiene styrene (ABS), nylon etc.

Initial Fabrication Results: These initial results encourage further research into natural volumetric fillers for 3D printing of fully recyclable and naturally biodegradable objects such as compost containers or vegetation planters harvesting chitosans fertilizer and plant growth promoting properties. Moreover, the antimicrobial effects of chitosan gels, combined with fibrous or granular fillers, could be applied to food packaging, exo-prosthetic devices, toys, or single-use consumer products such as cups or tableware (Mogas-Soldevila and Oxman [2015]).

In order to test and evaluate some of the materials designed, constructs are digitally fabricated layer by layer with composite materials containing; chitosan (CHI) in diverse concentrations (3%, 6% and 10% w/v) in acetic acid aqueous solution (1% w/v); cellulose microfibrils (CEL) in CHI solution (100% w/v in 3% CHI solution); and sodium alginate powder (SA) in CHI solution (4% w/v). Novel explorations on ceramic-chitosan composites have also been undertaken. In particular, insoluble kaolinite ceramic nano platelets in slurry (KAO) in 50%, 100%, and 150% by volume, are combined with chitosan (CHI), cellulose micro-fibers (CEL) and sodium alginate (SA) solutions in diverse concentrations and deposited in layers resulting in bio-ceramic composites inspired by naturally occurring nacre nano-scale arrangements. The multi-material 3D printing system is equipped with 6 material containers that can be connected to nozzles of varying sizes (from 0.7 to 9mm inner diam.) in order to achieve different resolutions along the printed objects. Importantly, due to the chemical makeup of the composites used, the platform can harness shrinking forces for

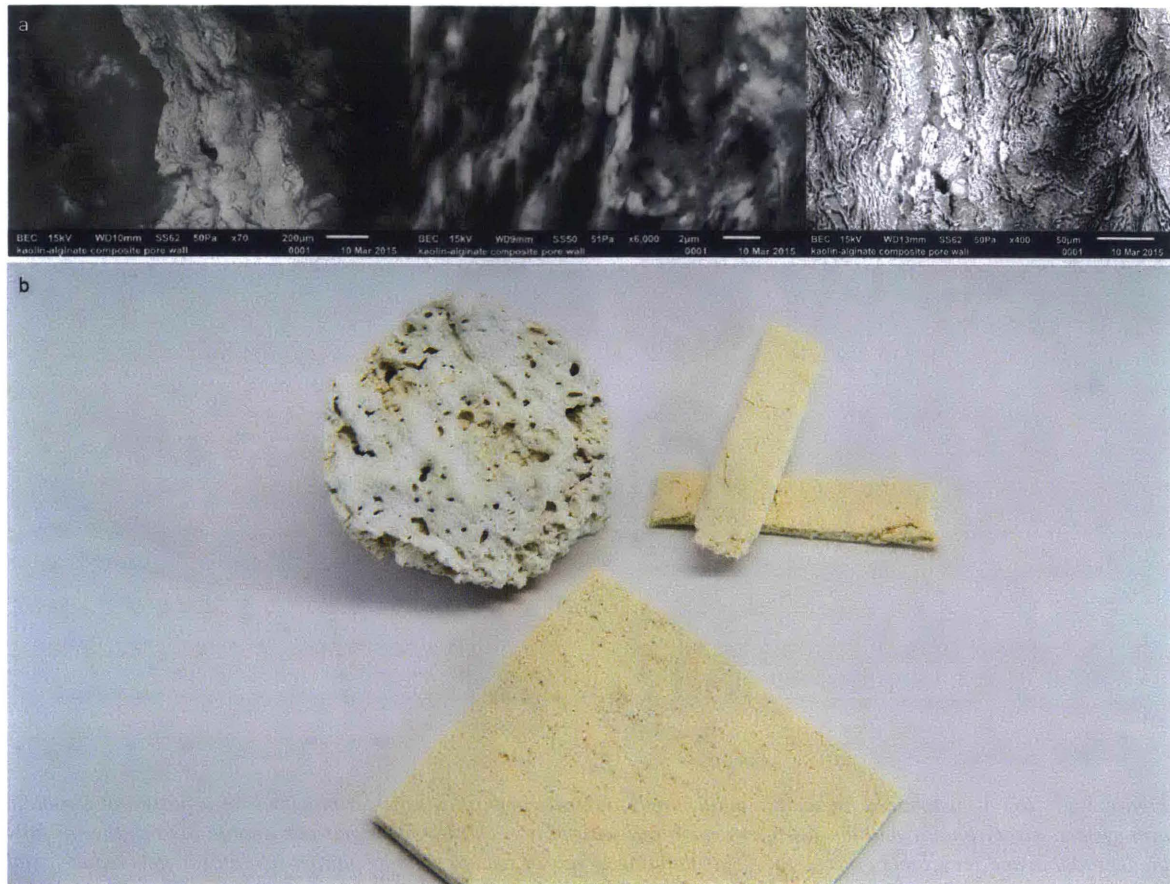


Figure 3-14: (a) Scanning electron microscopy of kaolinite-alginate composite. A tensile fractured pore wall at 200' micron, 50' micron and 2 micron scales, showing the size and local alignment of high aspect ratio kaolinite platelets. Adapted from (Lizardo [2015]). (b) KAO-SA and KAO-CHI-CEL tested samples provide insight for further research in bio-ceramic composites.

naturally induced shape formation. The wet depositions are layered flat and constructs find dry 3D shape by responding to internal directional evaporation stresses. Those stresses can be defined by geometrically designed heterogeneous patterns and computer-controlled material distribution along the extrusion process. Further detail about the novel computational control behind this deposition system can be found in Duro-Royo et al. [2015], Mogas-Soldevila et al. [2014], Duro-Royo et al. [2014a].

Initial Product Results: The resulting deposited objects display variable mechanical, optical, and biodegradability properties. For instance, in a 50cm long design inspired by the dragonfly wing, CHI and SA composites can be printed as structural bending-resistant venations, and clear CHI 3% solutions can be deposited and bonded to such venations to create a tension-resistant membrane in the construct [18] (Figure 3-16a top). The concentration of SA in CHI solution can be graded to provide for optical density variation from: 1% CHI and 10% SA, to 10% CHI and 1% SA (Figure 3-16a bottom). In another example, cellulose microfiber (CEL) in 100% w/v in 3% CHI solution is

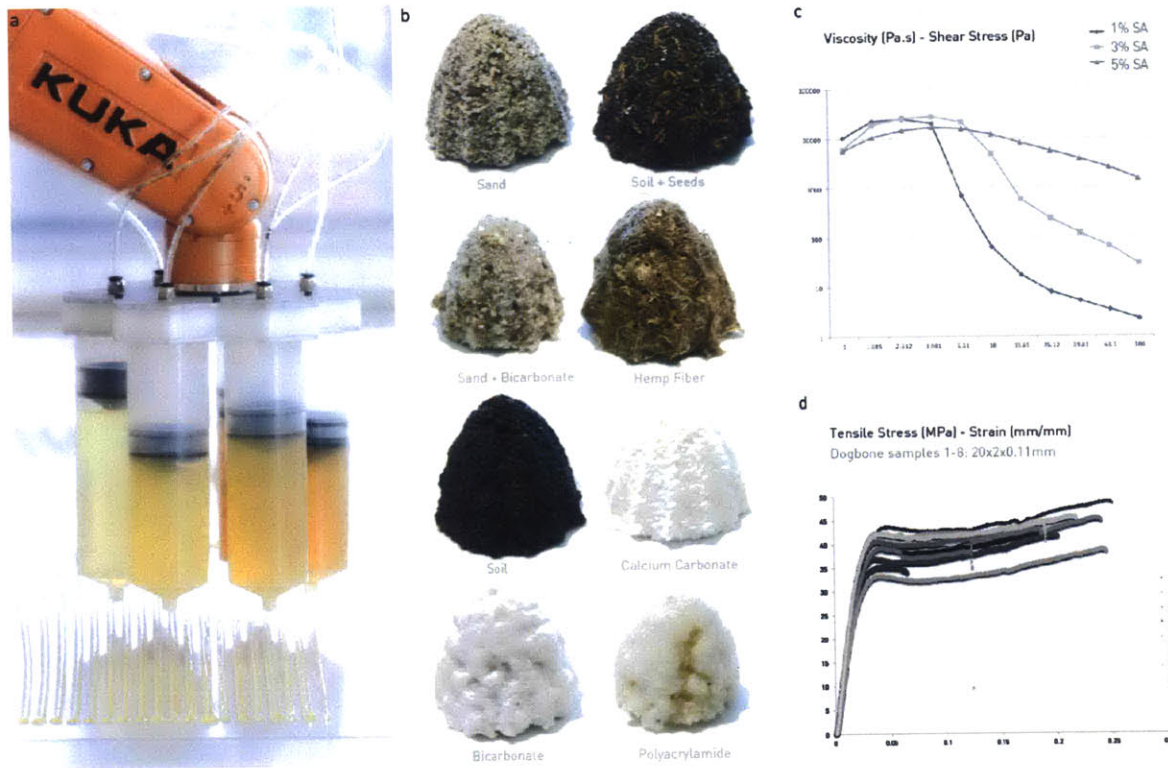


Figure 3-15: (a) A customized pneumatic multi nozzle natural composites extruder mounted on a positioning robotic arm system. (b) Granular and fibrous fillers combined with polysaccharide hydrogels can enable 3D printing of fully biodegradable and recyclable objects. (c) Shear thinning effects of sodium alginate powder in chitosan gels reduce axial forces needed to extrude colloids out of the barrel. (d) Chitosan bio-plastic films show an ultimate tensile strength (UTS) of 40mPa comparable to nylon or wood in the direction of the fiber. Figure from Mogas-Soldevila and Oxman [2015].

deposited in a 5x15mm grid and left to dry obtaining a fine 50cm length cylindrical structure (Figure 3-16b top). CEL extrusions can also be combined with 3% CHI depositions as in the membrane of a strong 1m long object with CEL spine-like venations (Figure 3-16b bottom). The combination of strong CEL members and tensile 3% CHI membrane results in a higher degree of structural performance of the final dried object (Mogas-Soldevila and Oxman [2015]). The deposited bio-ceramic samples based on kaolinite and chitosan are analyzed using scanning electron microscopy and display local alignment of the platelets (Figure 3-14) (Lizardo [2015]). Moreover, kaolinite is a soft white layered silicate clay mineral that constitutes the most abundant clay on Earth, its combination with chitosan and cellulose, the most abundant polymers on Earth, can be very relevant for macro-scale applications in architectural additively manufactured components under compressive as well as tensile loads. Further research need to be conducted in this direction.

Initial Tunability Results: Finally, CHI-only depositions can be mechanically and optically graded by using different concentration CHI gels in 1% w/v acetic acid aqueous solution (from 1% to 12% w/v) (Figure 3-16c bottom). Mechanical functional gradients can be accentuated by designing variable thickness extrusion path arrangements, or by using different nozzle sizes (0.7 to 9mm) for each extrusion path. Figure 3-16c shows part of a 3m tall construct inspired by leaf venations. The

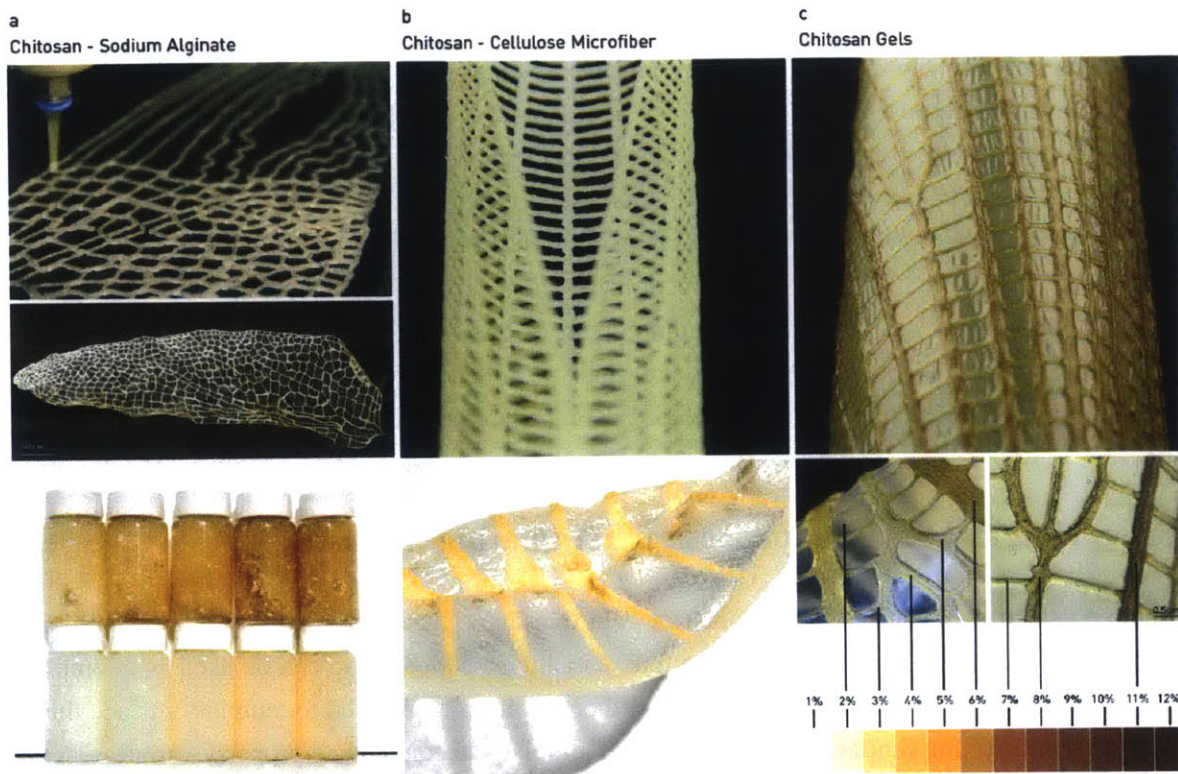


Figure 3-16: (a) CHI-SA composite defining a 50cm long construct inspired in dragonfly wing architecture. (b) CHI-CEL composites defining helical and cylindrical 50cm and 1m long grids and surface structures. (c) CHI multi-concentration composite describing a 3m long leaf venation pattern. Figure from Mogas-Soldevila and Oxman [2015].

structure combines CHI with varying concentrations organized as structural members and tensile membrane areas. Both areas are bonded by the self-repairing characteristics of the materials used [16]. These material systems and their properties can be deployed into temporary architectural panels, lightweight biodegradable car parts, wearable devices, or even bacterial scaffolds for biofuel production (as is further discussed in Section 3.5.3 of this Thesis).

3.3.2 Hydration Tunability Catalogue

A wide variety of raw biodegradable materials exist in Nature that display structural and environmental properties superior to those currently used for mass production of commercial products [20]. However, research into additive manufacturing processes for sustainable structures is still in its infancy, specifically in disciplines such as product engineering, product design and architectural design. Chitin, alginate and their respective chitosan and sodium alginate derivatives, are responsible for some of the most remarkable mechanical properties exhibited by natural materials, including seaweed gum, nacre, insect cuticle, and crustacean shells, and are as abundant on earth as cellulose, the main constituent of cotton or wood (Wiggins [1990], Lieleg and Ribbeck [2011], Vincent [2012]).

The Cycle of Water: In Nature, water provides for invisible support, its function is structural, mediating between the internal chemistry and the external environment. In Water-based Design Research water composes a full closed cycle as it is used in all steps of the design process - from material formation to product biodegradability. Specifically, in the research presented here water is used to; (I) chemically tune bio-materials to obtain different structural capacity properties, (II) enable "fabricability" in a liquid-to-paste extrusion-based digital manufacturing platform, (III) provide for the behavioral, functional and structural design of deposited constructs by hydration storage based flexibility, (IV) drive the shape-finding behavior of structures via hierarchical load distribution by differential evaporation patterns, and finally, (V) induce full recyclability and reuse of the bio-materials composing the objects produced with the fabrication platform.

3.4 Material Studies for Design Templating

I define design templates as frameworks for data translation that inform and encode material and immaterial performance criteria. For example, an anticipated load map can be used as a template for structural component thickness; whereas a thermal map can be used to template surface thickness and porosity for ventilation purposes. Such templates accommodate property variation in the course of the design process, while ensuring the implementation of the main algorithm. They tailor and integrate structural, manufacturing and environmental design templates relating to multi-scale feature resolution and global structure formation and performance.

In order to do so, Jorge Duro-Royo and I implemented a seamless computational process encoding virtual design and physical digital fabrication that relates to the MIT Mediated Matter Group's ongoing research on multi-dimensional, media and trans-disciplinary data informed design workflows. I demonstrate in this thesis the results of water-based fabrication: fully biodegradable architectural-scale structures that are additively manufactured, self-supportive, lightweight, and materially heterogeneous. Variable flow rates, differentially distributed material properties, and environmental control inform self-folding by shrinkage of the constructs as water evaporates from the water-based deposited gels (Duro-Royo et al. [2015, 2014a], Mogas-Soldevila et al. [2014]). Results display a range of structural behavior (Figure 3-17b) and represent a response to some of the planet's distresses related to the construction industry's unsustainable practices.

Bioinspiration: Initial bottom-up empirical studies involve the design and fabrication of functional patterns inspired by insect wing and leaf venation geometry, mimicking structure-and-skin graded architectures. In particular, dragonfly wings are passive lightweight structures supporting dynamic loading and employing high corrugation to increase stiffness and strength (Jongorius and Lentink [2010]) (Figure 3-17a). It is interesting to note that the Young's modulus of dragonfly wings can vary widely within the structure partly due to hydration patterns induced in the presence of chitin and proteins altering local properties (Donoughe et al. [2011], Jongorius and Lentink [2010]) and providing for both graded flexibility and graded tensile strength along the wing from the joint to the tip

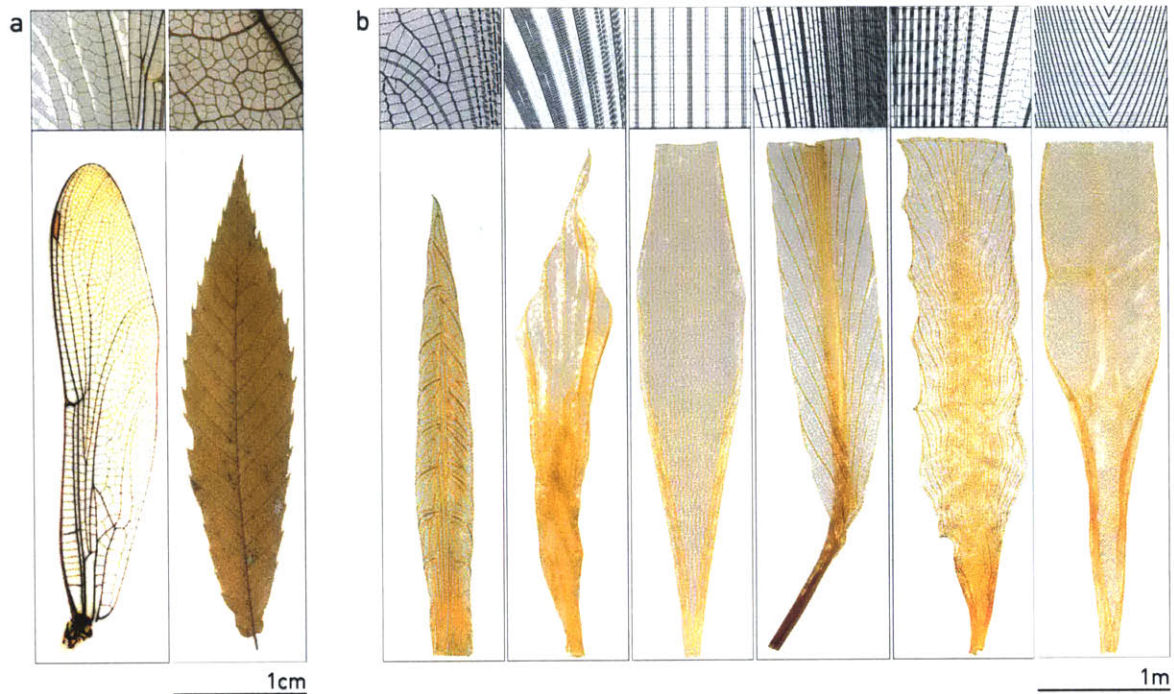


Figure 3-17: Experiments in hierarchical additive manufacturing strategies. (a) Top: close-up image of the structural pattern of a dragonfly wing and of the venation pattern of an Acer leaf (Image credit: CUPAC (Cornell University Plant Anatomy Collection, specimen 366, used with permission). Bottom: plan view and detail of a dragonfly wing and a dry leaf. (b) Top: detail of geometric trajectories for additive material distribution patterns (the gray shaded background represents film membrane infill). Bottom: front view of large-scale structural experiments when dried. Figure from Mogas-Soldevila et al. [2015a].

(Fernandez and Ingber [2014]). As insect wings, leaves are flat structures that maximize surface-to-volume ratios. Their venations are two-dimensional ramifying structures that tightly relate form and function performing both transportation of mass and energy, and distributing force fields across the surface area of the leaf structure (Roth-Nebelsick et al. [2001], Sack and Scoffoni [2013]). The highest mechanical stresses in leaves occur along their longitudinal axes; and transversal parallel veins contribute to stabilize bending forces (Figure 3-17a). Both longitudinal and transversal structural systems are lignin-based with a high elastic modulus, which, when combined with hydration and turgor pressure within cells, provides support and allows for high flexibility to reconfigure or fold under mechanical loading (Roth-Nebelsick et al. [2001], Sack and Scoffoni [2013]). Such support and functional variation strategies, as well as the use of primary and secondary structural systems, are applicable to architectural structural cantilevers, shells and spatial structures (Mogas-Soldevila et al. [2015a]).

Digitalization: Structural design patterns are synthesized combining strategies from both voronoi-based insect wing patterns, and leaf ramifying venations in order to obtain functional and mechanical gradients. Functional gradients, spanning column-like behavior to surface-like behavior, are obtained by negotiating geometrical pattern density and hydration rates (Figure 3-17b bottom). Me-

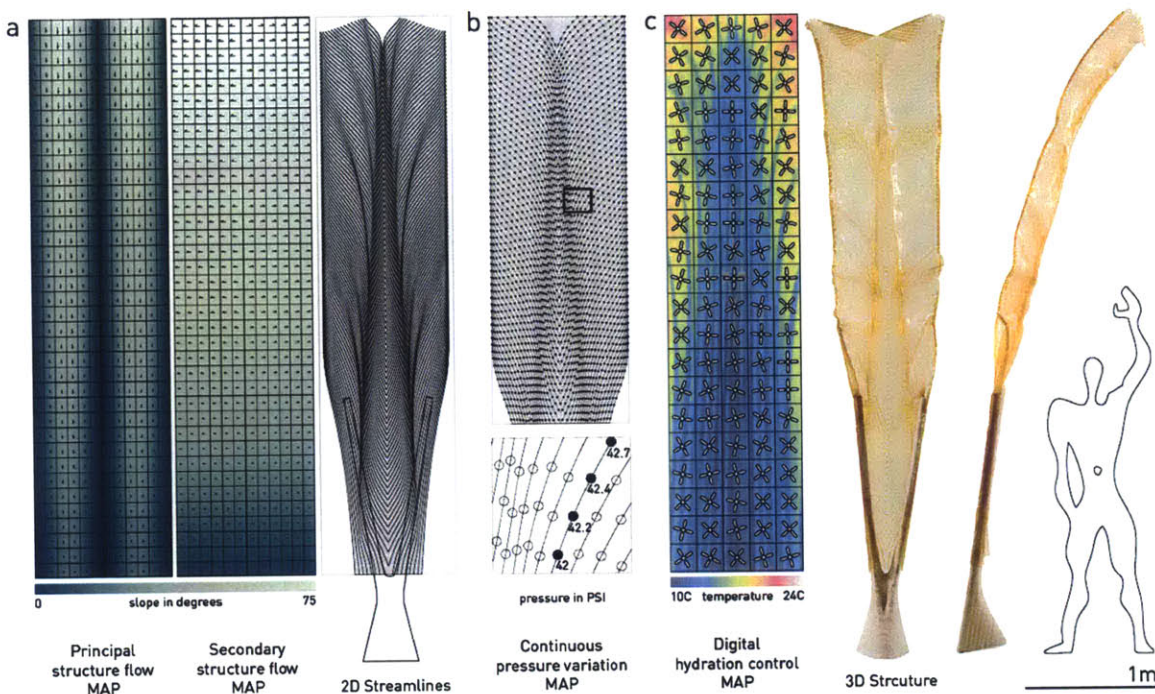


Figure 3-18: (a) Vector fields encode variation in principal and secondary structure, and are interpolated into streamlines of material deposition trajectories. (b) Pressure, speed and material concentration variation along structural streamlines is encoded into real-time instructions sent to the manufacturing platforms. (c) Computerized hydration control maps on top of deposited structures provide global evaporation-driven self-folding of the final functional 3D structure. Figure from Mogas-Soldevila et al. [2015a].

chanical gradients are obtained by modulating stiffness, pressure, and layering strategies. Finally, optical gradients are obtained that are directly proportional to structural gradients and are a result of grading material concentration from 3% w/v -translucent- to 12% w/v opaque - in deposited gels. The constructs are 1m to 70cm wide and range from 2.5 to 3m tall when dry (Figure 3-17b bottom). In the presented implementation, as in insect wings and many leaf types (Roth-Nebelsick et al. [2001], Sack and Scoffoni [2013]), the size of the local diameter of load bearing members with respect to the global size of the structure is key to guaranty self-support and bending stability for cantilevered performance of the constructs.

3.4.1 Structural Design Templating

In the constructs designed and presented next, structural template maps are designed based on hierarchical flow of loads encoded into principal and secondary structure streamlines. The principal structure is longitudinal to the global shape of the construct and provides for stiffness employing thicker diameter members and higher material concentrations as can be observed in leaf venation and insect wing structures. The secondary structure can be semi-parallel or semi-perpendicular to the principal structure and is composed of hierarchical networks of thinner member diameters and lower concentration materials. Parallel secondary structures allow for column-forming structures,

and perpendicular secondary structures allow for wall-forming structures. Figure 3-18a shows principal and secondary streamlined structural pattern generation by interpolating 2D vector fields from 3D scalar fields. Desired 3D surfaces are modeled and their gradient of slope mapped into 2D vector fields that represent local structural behavior. The vector fields are then computed into continuous principal and secondary directions of flow (Figure 3-18a) (Mogas-Soldevila et al. [2015a]).

3.4.2 Robotic Manufacturing Templating

Once the streamlines for principal and secondary directions are determined, hierarchical material distribution is implemented. There are three ways to determine differentiated material distribution to achieve selective structural capacity; pressure variation can be implemented along selected lines resulting in continuously varying material accumulation (Figure 3-18b); material concentration can be assigned to each trajectory resulting in stiffness gradients from lower to higher concentrations; and, finally, layering onto dry deposited material provides higher degree of reinforcement (Duro-Royo et al. [2015], Mogas-Soldevila et al. [2014]). Such strategies are encoded into position, speed, pressure, and material instructions that are sent in real-time to both positioning and deposition platforms (for further detail on manufacturing instruction generation see Duro-Royo et al. [2015, 2014a]).

3.4.3 Environmental Performance Templating

Hydrogel materials are deposited in geometrical and pressure patterns onto a flat substrate following instruction parameters. As water evaporates, shrinking forces accumulate across the networked gel and induce self-folding once the construct is removed from the substrate. In order to distribute hydration across large-scale depositions, a computerized evaporation control system can be easily implemented composed of 100 variable speed fans with differential control according to desired degrees of final hydration compared against thermal imaging of the deposition (Figure 3-18c) (see Mogas-Soldevila et al. [2015b], Duro-Royo et al. [2015] for additional detail). Hydration control allows for increased final folding in hydrated areas providing for structural interface joints as shown at the foot of the dry structure in Figure 3-18c. If uncoated, the structures will fully biodegrade providing nutrients as they decay in contact with the environment. Such materials can be fully recovered and reused if dissolved in water (Mogas-Soldevila et al. [2014]).

3.5 Resulting Water-based Products and Utilities

Presented here are variable property biomaterial-based components 3D printed at room temperature, using relatively little energy and mild chemicals. Applications of the design and manufacturing method range in scale and function from lightweight robotics such as flapping micro vehicles, cell growth promoting environments such as biocompatible wearable devices in contact with regenerating tissue, biofuel-producing bacterial culture supports (Patrick [2015]), fully compostable

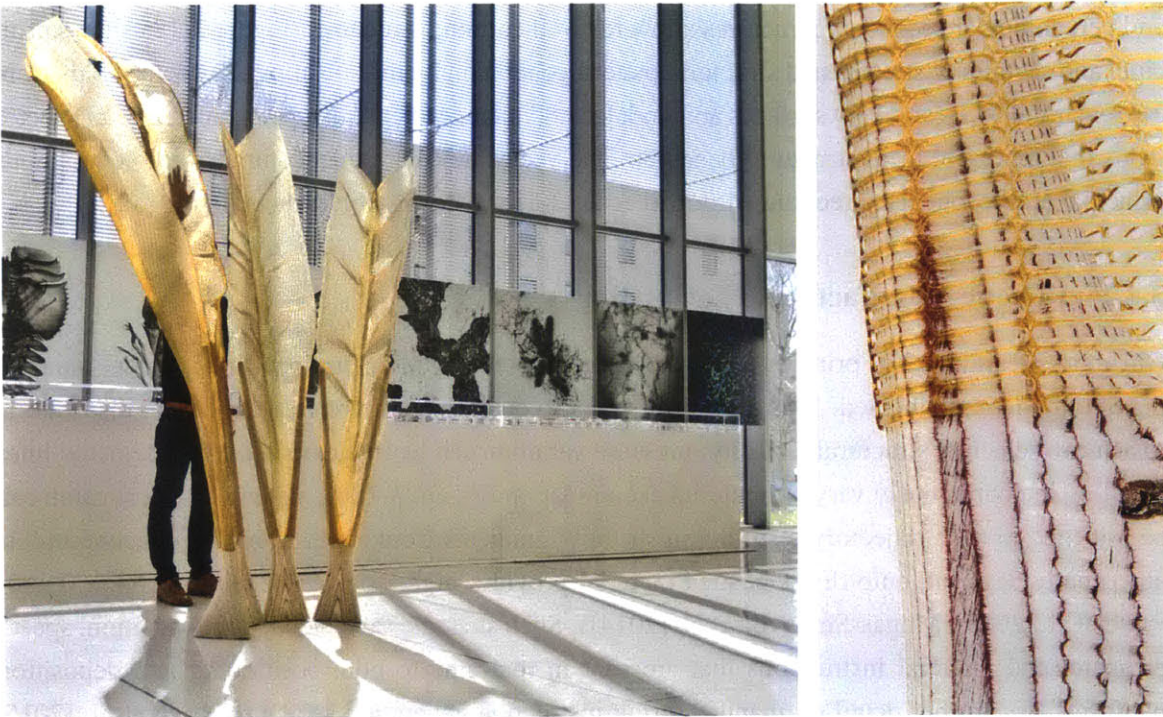


Figure 3-19: Perspective view of a typical structural assembly and research exhibit focusing on water-based additive fabrication and biological design at the MIT Media Lab, 2014.

consumables, ecosystem-enhancing constructs that replenish soils with nutrients as they decay, and temporary biodegradable architectural structures or building skins. Experimentation in the direction of embedding biofuel-producing bacterial cultures in chitosan printed samples is conducted with William Patrick at the MIT Mediated Matter Group, in collaboration with doctoral student Stephanie Hays at the MIT Silver Lab, and with Eleonore Tham at the MIT Lu Lab.

3.5.1 Macro: Architectural Design Applications

With biological material formation characteristics in mind, I present the production of architectural-scale structures that are not only derived from structural patterns in nature, but also manufactured from biological materials such that they highly interact with the environment as they take shape and ultimately biodegrade (Figure 3-19) (Mogas-Soldevila et al. [2015a, 2014]).

By deriving fabrication technologies, structural design, and environmental adaptability from material-driven research, novel cross-disciplinary multi-functional properties such as; optical gradation proportional to load-bearing capacity; merging of structure and skin functions within the same material system; energy and resources savings due to material environmental responsiveness; emergence of functionally-graded shape induced by time, air and water; or the possibility to flow biofuel-producing organisms within structural stress lines of the constructs. The lightweight robotically fabricated biomaterial structures represent an ecological and economical approach to materially heterogeneous formation by design, and an innovative exploration into future biocompatible living



Figure 3-20: Bioplastic-based fully recyclable packaging products composed of plasticized CHI 10% concentration and plasticized CHI 10% concentration with black food coloring additive.

spaces that can have a high impact in alleviating our planets resources and health issues.

3.5.2 Meso: Product Design Applications

Product-scale fully recyclable applications of this research are very promising and range in scale and function. Lightweight robotics applications can include flapping micro vehicles. Wearable devices include fashion items such as skirts and jackets (Figure 3-24), jewelry, purses, or baskets (Figure 3-22). In fashion items, flexibility and body adaptation are achieved through discontinuous robotic deposition of CHI 6%, 8% and 10%, on top of CHI 3% continuously deposited layer. Fully compostable consumables include packaging boxes (Figure 3-20), cups, cutlery or grocery bags (Figure 3-21) that could help alleviate plastic pollution affecting ocean ecosystems. Medium-scale recyclable facade components include flexible panels composed of chitosan or chitosan-cellulose composites (Figure 3-25). These bio-materials can be dyed with non-toxic food coloring products and achieve richer opacity qualities on top of the ones achieved by material concentration ranges (Figure 3-20).

3.5.3 Micro: Biological Design Applications

Chitosan is a naturally produced (Figure 3-26a) biocompatible material that is used in the medical industry as cell scaffold for tissue growth Li and Ortiz [2014], Gutowska et al. [2001], Melchels

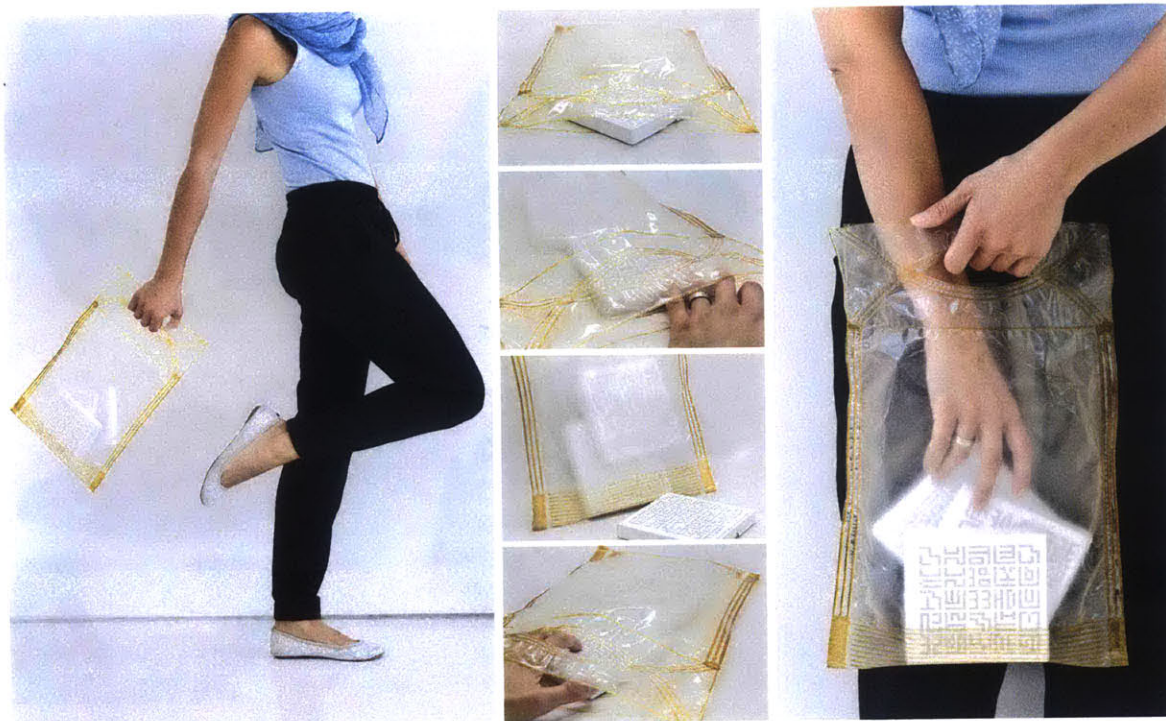


Figure 3-21: Bioplastic-based fully recyclable disposable grocery bag composed of plasticized CHI 10% concentration and plasticized CHI 3% concentration.

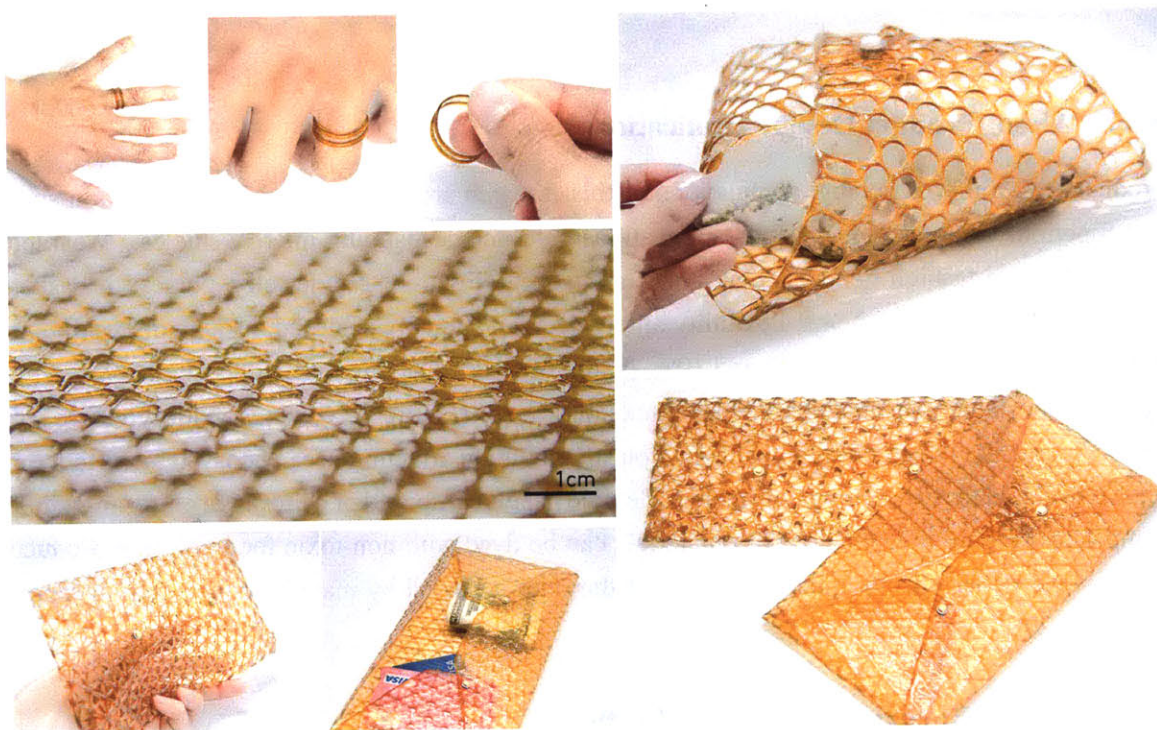


Figure 3-22: Bioplastic-based fully recyclable jewelry, baskets, and fashion clutches composed of hierarchically structured plasticized CHI 10% concentration.

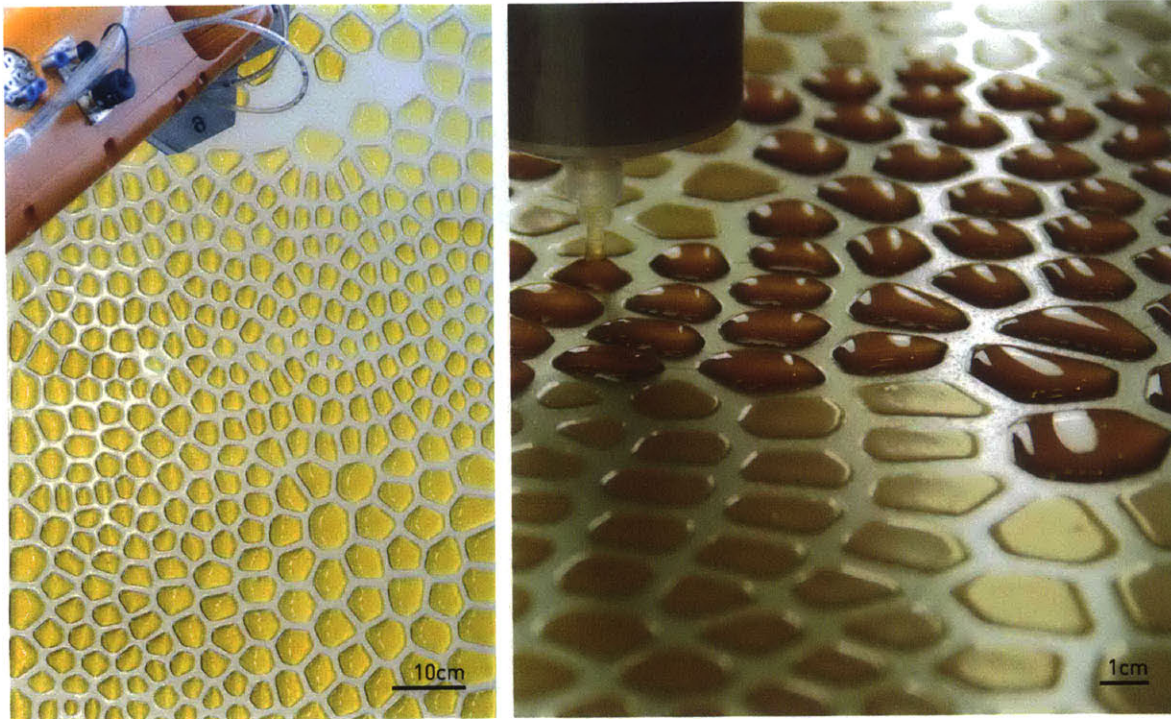


Figure 3-23: Discontinuous robotic deposition of CHI 6%, 8% and 10%, on top of CHI 3% continuously deposited layer. In future implementations, bacterial biofuel production pockets can be designed along with water-storing gels and embedded nutrient and collection devices.

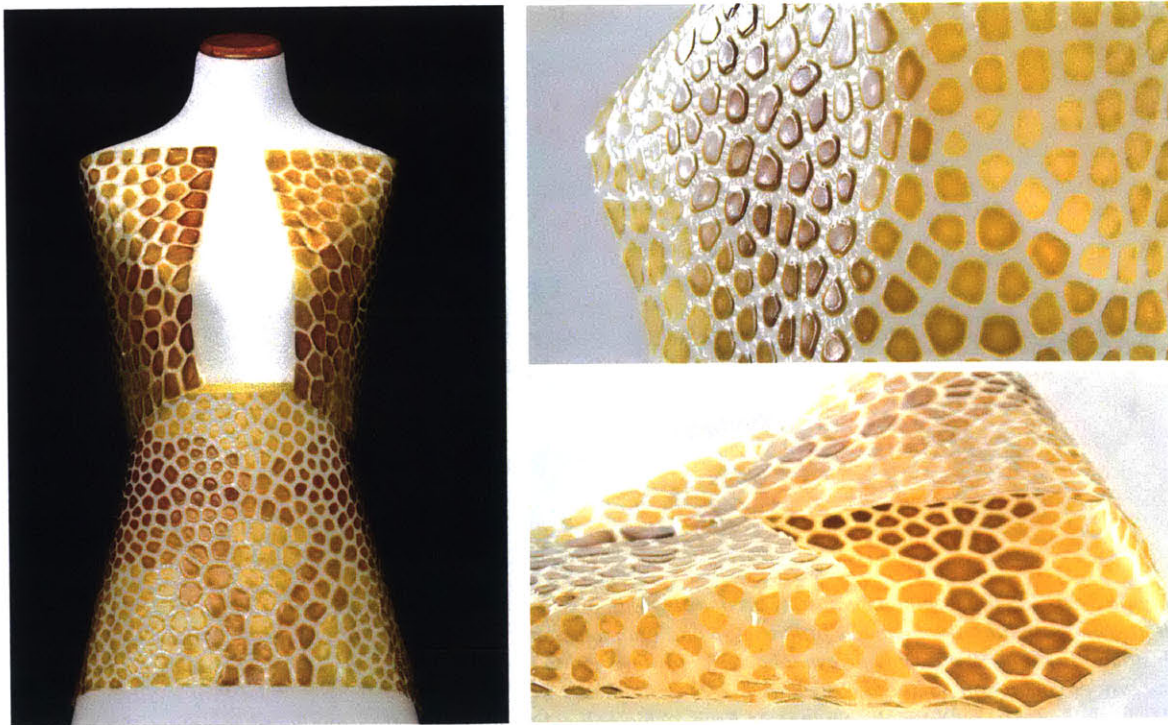


Figure 3-24: Discontinuous robotic deposition of plasticized CHI 6%, 8% and 10%, on top of CHI 3% continuously deposited layer for full-scale jacket and skirt fashion items design displaying flexibility and adaptation to body shape.

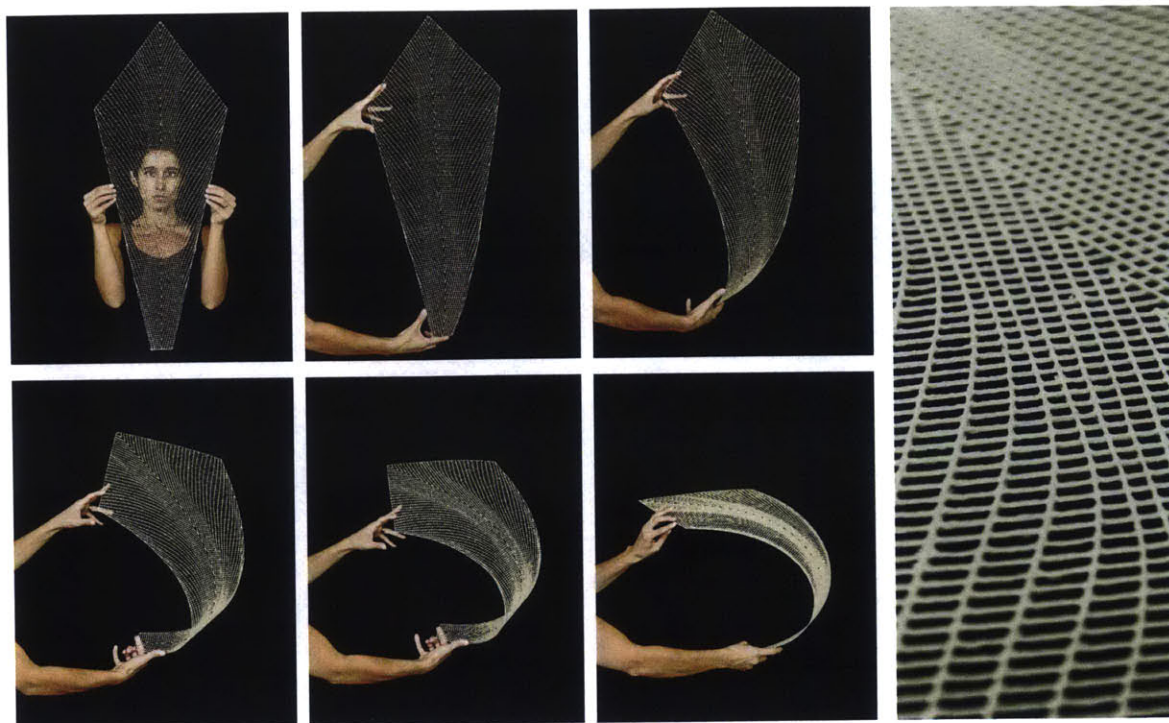


Figure 3-25: Robotically-manufactured facade component design composed of CHI 10% plasticized matrix and cellulose microfibers filler material providing promising bending capabilities.

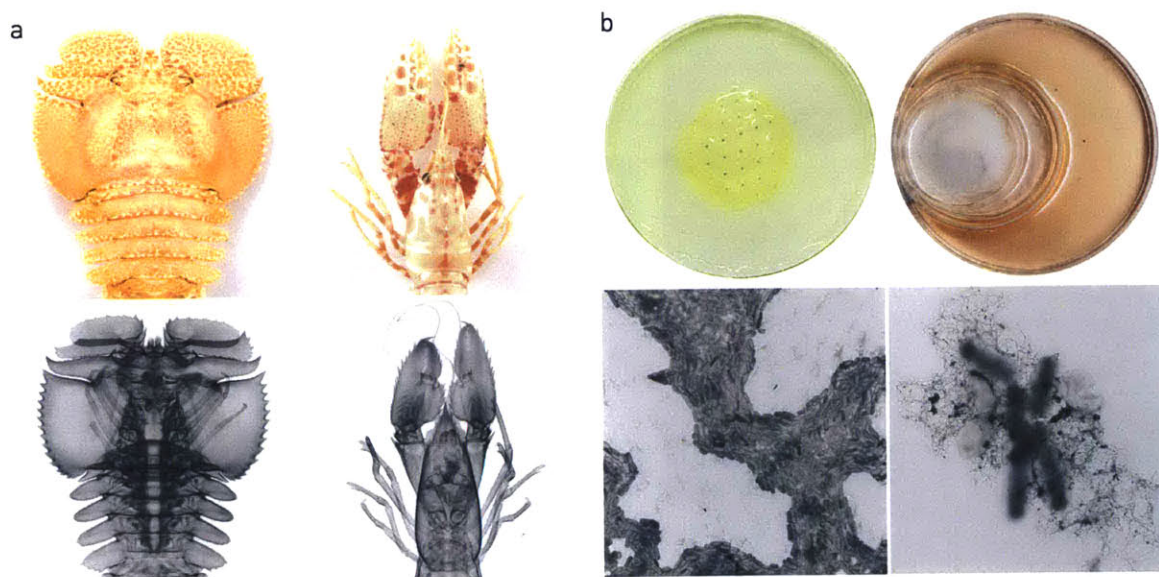


Figure 3-26: (a) Top: Chitin is naturally produced within crustacean shells in combination with minerals and proteins. Bottom: X-ray transmission image of *Ibacus* sp and *Enoplometopus* sp. Image credit: Dr. James Weaver, Harvard Wyss Institute. (b) Top: Photosynthetic cyanobacteria growing within two layers of chitosan. Colorimetric assay quantifying sucrose production in bacterial co-culture (12-24-72h). Image credit: S. Hays, T. Ferrandt and Prof P. Silver, Harvard Medical. Bottom: Synthetically engineered *Escherichia coli* colony (1 μ m length) strain forming biofilm. Synthetically engineered *Escherichia coli* (1 μ m length) with protein fibrils attaching gold nanoparticles. TEM images. Image credit: Elonore Tham, Lu Lab, MIT. Figure from Mogas-Soldevila et al. [2015a].

et al. [2012]. Consequently, in future iterations of the manufacturing platform presented herein, materials that can be used for biocompatible wearable devices in contact with regenerating tissue, or biological microorganisms can be printed or placed using the pneumatic extruder with fine pressure tunability. Bacterial biofuel production pockets can be designed and deposited along with water-storing gels and embedded nutrient and collection devices (Figure 3-23) (Mogas-Soldevila et al. [2015a]).

Figure 3-26 illustrates preliminary results of bacterial culture growth on top of deposited chitosan samples (Figure 3-26b) (Patrick [2015]). Biological-driven design for medical or human-augmentation applications will involve further integration of environmental parameters and biological phenomena such as solar and airflow exposure patterns, temperature gradients, photosynthetic rates, nutrient availability, etc. As in leaf venation patterns, multifunctional aspects of hydraulic transport and mechanical protection (Sack and Scoffoni [2013]) can be integrated into hierarchical structures generating a true material-based ecosystem (Mogas-Soldevila et al. [2015a]).

Chapter 4

Conclusion

Architectural and environmental motivations encourage material-driven research in Water-based Digital Design and Fabrication. The role of water in manufacturing methods has the potential to approach the role of water in the biological world. Water-based Digital Design and Fabrication proposes the use of water as a key ingredient in all phases of the design and production process: initiation, activation, shape formation and biodegradation. The platform and its associated methods serve to advance sustainable digital manufacturing techniques and propel them towards biologically inspired and informed manufacturing. Initial outcomes show the design and production of sustainable and self-supporting structural components across scales. Combining the use of natural regenerated abundant materials with a computer-controlled additive technology, structural and material hierarchies can be designed and manufactured achieving sustainable products and building components characterized by high spatial resolution in manufacturing that are multifunctional and can biodegrade upon contact with water. Moreover, the design of the platform dramatically reduces the need for external energy sources for fabrication other than air-actuated extrusion and displacement of the nozzle head, pointing towards new possibilities for sustainable additive manufacturing.

4.1 Contributions

4.1.1 Material Studies in Practice

Architectural design and building construction can highly benefit from a deeper understanding of the strategies and processes that lie behind formation of non-toxic, robust and efficient, yet chemically simple and fully recyclable, materials and structures in nature. Today's industrial manufacturing processes are generally characterized as wasteful and their products often highly difficult to recycle. Specifically, man-made plastics are the most energy intensive materials to degrade posing global health and resource issues as plastic pollution increases and fuel availability decreases. However, water-based material structures in particular are naturally degradable while displaying exceptional and diverse mechanical properties. The present thesis' case study focuses on low-energy and mild-condition deposition systems for polysaccharide gels in order to design large-scale functionally

graded structures with environmental capabilities for applications in architectural and product design.

4.1.2 Material Studies in Scholarship

It is fundamental that we identify the extraordinary properties of simple assembly, hierarchical structure, and material organization present in biological systems in order to develop efficient strategies for energy and matter conservation in architecture. Architecture and product design scholarship can benefit from; implementation of biological concepts of sustainability, comparison of disadvantages of current sustainable design practices with advantages of naturally formed sustainable systems, bottom-up design exploration strategies based on local material property design and assembly, to global structural system performance, and production of functionally complex and energy efficient designs inspired by biological sustainable strategies. Such integrated knowledge can be achieved by implementing novel architectural education and research practices informed by the disciplines of biology, material science, and advanced manufacturing.

4.2 Perspectives

4.2.1 Research Limitations

In the presented Water-based Fabrication case study; natural materials are combined via bottom-up matter behavior explorations and design of novel digital fabrication systems. The complex chemistry of the material composites studied for direct product fabrication, render the process rich yet unpredictable. However, the ecological implications of using and reusing, in a closed-cycle process, Earth's most abundant natural polymers and ceramics cellulose, chitosan and kaolinite - are vast and exciting. Moreover, such biologically produced materials also hold the potential to sustain life and promote growth of microorganisms allowing "functionalization" of manufactured structures into, for example, biofuel producing factories (Patrick [2015]). Other water-based conventional fabrication materials can be used that are stored in air-tight extruder barrels at room temperature and that cure at room temperature by natural evaporation. For instance, organic resins, polyvinyl alcohols, clay, plaster, calcium sulfate and calcium carbonate compounds in concrete. These materials can also be combined with granular or fibrous fillers proposing further work into the composites possibilities of this research.

4.2.2 Future Directions

Biological structures adapt to external stimuli by water-induced material property variation resulting in hierarchically structured forms (Vincent [2012], Mogas-Soldevila et al. [2014]). This thesis argues that conventional digital design tools and processes, by contrast, lack robust integration of computational design tools, digital fabrication environments and material. Preliminary results

shown here demonstrate chemical tuning of biomaterials with digitally designed and fabricated structured hierarchies informed by load-responsive arrangements as can be found in the biological world. Such case studies have informed my research into form generation and fabrication of optimized load distribution in meso-scale (product) and macro-scale (architectural) components. Importantly, I observe that, in the Water-based Fabrication platform presented here, the shear forces involved in the extrusion process of chitosan solutions induce alignment of the crystalline regions of the polymer in the direction of printing at the nano-scale, which affects the shaping of the cured constructs at the meso-, and macro-scale. Consequently, hydration guided shape-formation studies are currently being conducted by the Mediated Matter group in collaboration with Prof. Katia Bertoldi and doctoral student Johannes Overvelde (Bertoldi Group, Harvard School of Engineering and Applied Sciences) in order to establish informed nano-to-macro scale relationships between shape, material composition, hydration rates, and design-induced deformation.

Our research into Water-based Digital Design and Fabrication engages all aspects of a complex design process - material-driven research, bottom-up experimentation, parallel novel technology development, and file-to-production feedback. Finally, the thesis advocates for the architect to play the role of a cohesive thinker, as well as a rigorous science and aesthetics explorer, able to seed novel processes that emerge from material studies towards digital design and advanced fabrication.

Biography

Laia Mogas Soldevila is an Architect, currently a Research Assistant at the Mediated Matter Group, MIT Media Lab, focusing on design and digital fabrication of natural and biologically inspired material systems. She completed her Architectural MSc. diploma minoring in Visual Arts at the Polytechnic University of Catalonia, School of Architecture (UPC-ETSAV) where she graduated with honors. In 2010, she completed a post-professional degree in Advanced Design and Digital Architecture at the Pompeu Fabra University (UPF). In 2010-2011 she was awarded research grants to pursue the Architecture, Energy and Environment program at UPC-ETSAB and the Master of Science in Architecture Studies at the Massachusetts Institute of Technology, Design Computation Group. Since 2008, she co-leads the DumoLab design research studio with Jorge Duro-Royo at the intersection of architecture, material practices and advanced computation (<http://dumolab.com>). Laia's work has been exhibited at the MIT Museum in 2015, the MIT MediaLab in 2014, the Radcliffe Institute for Advanced Study at Harvard University in 2014, the Lisbon Architecture Triennial 2013, the Istanbul Design Biennial 2012, the Pompidou Center in Paris as part of Advances in Architectural Geometry Exhibit 2012 and the Elisava School of Design in 2010.

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