Molecular Design Interactions:
Material Synthesis for Human Interaction with Fluids

by

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Abstract

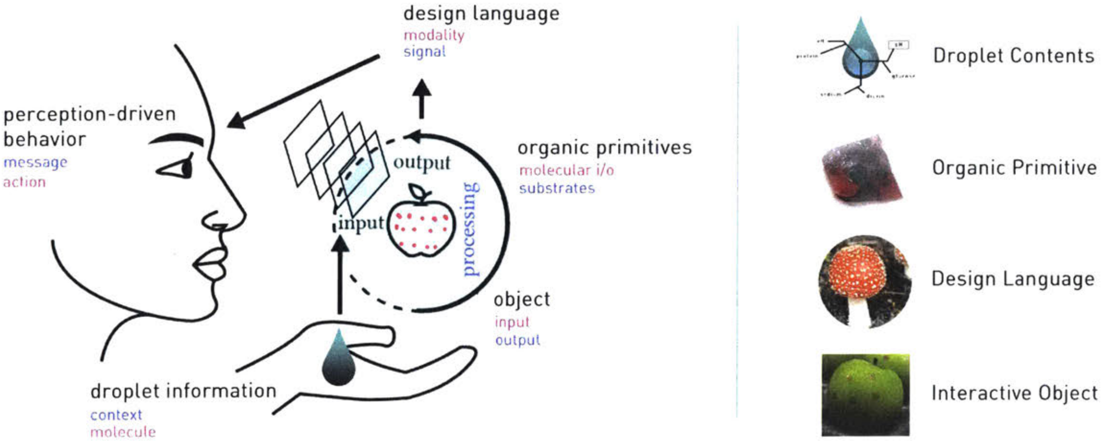

Figure 0-1. Key elements within a *Molecular Design Interactions* interaction loop

Be it information embodied within rain, the oceans, a dinner plate, or human tears; the flow of information through fluids provides insights into the biological and chemical states of systems. Yet a large portion of our everyday experience with these systems remain inaccessible to users, designers and engineers whom operate outside the context of chemical disciplines. This thesis introduces a design framework coined *Molecular Design Interactions*, along with a toolbox of material based input-output devices termed *Organic Primitives* to facilitate the design of interactions with organic, fluid-based systems. The design methodology utilizes organic compounds from food for the development of color, odor and shape changing information displays. Activated by units of fluid information called *droplets*, this thesis focuses on pH signals in fluid as a model to demonstrate how molecular scale phenomena can be brought from materials into applications for interaction with a range of organic systems. A design language and vocabulary, drawing from signaling theory and molecular associations, offer designers a method with which to translate sensor-display output into meaningful experience designs for human perception. The design space showcases techniques for how the *Organic Primitives* can transcend beyond mere input-output devices to achieve higher order complexity. Passive and computational methods are presented to enable designers to control material interface output behaviors. An evaluation of the individual output properties of the sensors-actuators is presented to assess the rate, range, and reversibility of the changes as a function of pH 2-10. Strategies for how the materiality of objects can be augmented using *Organic Primitives* are investigated through several applications under four contexts: environmental, on-body, food, and interspecies. *Molecular Design Interactions* offers a process and toolbox to create interfaces between humans and molecules in fluids, across scales, from the nano to the macro systems.

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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 Master of Science at the Massachusetts Institute of Technology

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1. Introduction

1.1 Enabling Tools Blur Disciplines

A large part of our everyday experience with natural systems is inaccessible to designers and engineers from the field and purpose of user interaction design. The challenge of programming interactive experiences within the realm of food, personal care, and other organic systems for designers lies largely in the limitations of conventional tools for prototyping, as they are incompatible with these soft, fluid-based systems. The dialects required to manipulate such systems are mediated by chemical codes, which control the most complex machine we know —life. In order to open up possibilities for interacting with organic systems, an accessible, biocompatible toolbox and design process is essential.

The complexity of biomolecules makes detection using modern electronic chemical sensors a challenge. Researchers in chemical and biological engineering fields often rely on chemical mechanisms and reactions for sensing and analysis. These processes are typically executed by molecular sensors, in liquid medium or immobilized on material substrates. For example, bicinchoninic acid assay is an analytical procedure utilizing molecules copper(II) sulfate pentahydrate and bicinchoninic acid to form reactions to determine the concentration of proteins in solution [1]. In-home pregnancy tests utilize immobilized antibodies with conjugated enzymes on a substrate to detect biomarkers such as the human chorionic gonadotropin (hCG) hormone in urine [2]. While many methods have been developed by chemical and biological disciplines for molecular sensing and analysis, there has been little to no emphasis within these fields to develop these mechanisms for user interaction.

Disciplines whose primary focus is designing for the human experience, such as industrial design, human-computer interaction, interaction design and architecture, have yet to incorporate molecular design approaches into their practice. This is due to historical precedence for how bodies of knowledge were cataloged and archived during the beginning of the 1800's [3]. The widespread adoption of design tools have enabled the historically disparate realms of the arts and sciences to blur through shared technology and discourse. Software tools such as Solidworks is one such example - where the logic of mechanical design is offloaded into the workflow of how a user interacts with the program [4]. Mechanical engineers utilize computer-aided design (CAD) tools such as Solidworks to engineer mechanisms. As industrial designers adopt CAD tools into their practice, mechanical design considerations and principles are expanded upon in their discipline [5]. Hardware tools such as Arduino micro controller boards and its integrated development environment enabled interaction designers as well as hobbyists to adopt electrical engineering capabilities into their workflows [6]. While it is unclear whether such approaches to do-it-yourself (DIY) learning has the ability to replace formal education to achieve professional proficiency, these tools are embodiments of knowledge for what was once bounded by discipline, but now readily transmitted through the use of tools. Enabling technologies offer opportunities for disciplinary boundaries to be broken down, as they empower broadened audiences to engage with its practices.

1.2 Food as Medium for Interface Design

Tools for enabling non-chemists to interface with chemical and biological systems face challenges with cost, accessibility, safety, and expertise by its users. The expertise required to manipulate these systems make it difficult for individuals without a chemistry background to engage in practice. Often requiring advance equipment and expensive reagents to conduct experiments, novices face operational as well as instructional challenges. Yet food based materials and kitchens are commonplace, possessing a range of equipment that can be leveraged as an accessible laboratory with diverse chemical reagents.

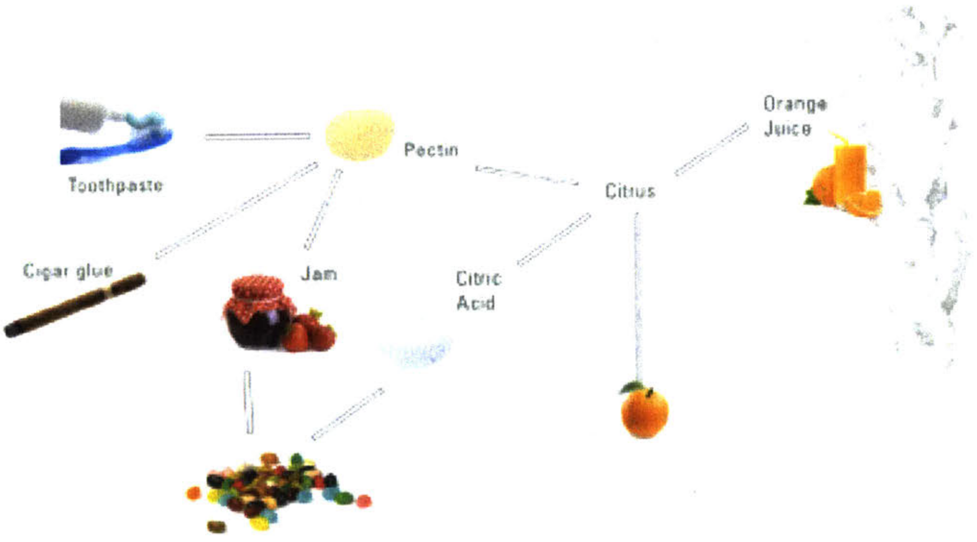

Figure 1-1. Extracted components of citrus fruits are used to produce a range of applications beyond consumption

The process of deconstructing food touches upon a range of domains such as health, agriculture, biotechnology, sociocultural conditions, governance and public policy [7] [8] [9] [10] [11]. What we often think of as food are also organisms that have evolved structures and compounds with sophisticated properties. From stimuli-responsive molecules to biocompatible substrates, edible materials have been exploited for a range of functions, across different industries. As an example, pectin derivatives from citrus fruits are extracted to produce toothpaste, cigar glue, as well as citric acid food additive, as shown in figure 1-1. Erb et. al have utilized gelatin and alginate (seaweed) to create shape changing bio-inspired composites activated by water [12] [13]. Leveraging existing kitchen infrastructure and food materials makes tools for interfacing with chemical and biological systems a feasible, scalable, and accessible endeavor.

Beyond physical properties, food experiences serve as some of the most sensory rich interactions humans have on a daily basis. These multimodal interactions communicate a breadth of information to us through our senses as they encode meaning across social, cultural, and behavioral dimensions [14] [15] [16]. For humans, food extends beyond its consumption for sustenance, but also possesses a power to direct perception and behavior. This makes it a rich medium for communication and messaging.

1.3 Human Interaction and Communication with Molecules

As humans, we gather information about the material world through our sensory systems with what we taste, smell, see, hear, and touch. Perception of chemical signals are encoded through sensory associations with molecules that give rise to certain odor, flavor, texture or color attributes [17]. Take for example the notion of a rotten egg from a refrigerator, where a sulphuric compounds communicates a 'bad' odor and therefore a 'toxic' state in the egg. Attributes of sulphuric compounds (bad smell) become conceptually coupled with objects (egg) in a given context (food from fridge) to form meaning (toxic if eaten). The modality in which these attributes are transmitted is dictated by the medium [1]. Here the medium is the composition of the egg - allowing it to transition from a neutral state into a 'toxic' state to emit a 'bad' odor by sulphuric compounds. The sulphuric odorants function as signals and messages that suggests particular affordances [2] provided by the egg. Drawing from the composition of food items, the mechanisms behind these odor, color, taste and form changing properties can be utilized as semiotic devices for interface design.

1.4 Materials as Translators

"The most profound technologies are those that disappear. They weave themselves into the fabric of everyday life until they are indistinguishable from it." - Mark Weiser [18]

Media technologies such as printed media, radio, television and internet, primarily utilize visual and/or auditory modalities as a means for transmitting information to people. Yet increasingly through the rise of physical computing, Internet of Things (IoT) and wearable technologies, these self-contained mediums are beginning to shift from principal devices into contextualized everyday objects. For example, Philips Hue bulbs enable users to control their lighting environment through their smart phone [19]. Vessyl a cup linked to mobile devices, detects and displays dietary content in beverages including sugar, protein, calories and caffeine [20]. Google ATAP's Project Jacquard utilize conductive thread to weave circuitry into clothing to create seamless interactive clothing [21]. Everyday objects from home appliances to cars, cups and watches are incorporating sensing and actuation to enable user interaction. As media technologies shift from flat screen displays to incorporate into everyday

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[1] Oxford dictionary defines medium as a means by which something is communicated or expressed. Marshall Mcluhan expands upon the notion of medium as the message.

[2] The term affordance was first coined by psychologist James J. Gibson as "action possibilities" for how one can engage with an object or environment.

objects, interface designs are moving from graphical user interfaces (GUI) into a range of form factors and modalities.

Recent developments in human-computer interaction (HCI) and robotics have sought to achieve seamless devices by incorporating sensing, information processing, and actuation within a unitary material [22]. Researchers have investigated shape changing capabilities using a range of techniques such as thin-film electronics, pneumatic soft elastomers, and particle jamming to achieve interfaces that can be deformed by the user as input and provide alternative functionality and haptic feedback as outputs [23] [24] [25]. The notion of interfaces which can deliver color change [26], odor release [27] and shape transformation [28] have been individually explored. However, material interfaces that offer sensing and multiple output functionality remains a challenge.

1.5 Addressable Units of Information

Molecules are the language that systems from the environment to our bodies and food use to communicate. Information such as pH, eH, glucose, and potassium in the natural world are often exchanged through the flow of fluids as they encode structures and properties that are essential for life. They can be transferred through water cycles with rain water, circulate through crops which intake fluid for growth, and then consumed by organisms as they are metabolized and excreted as semiochemical trails.

Pixels are considered to be the smallest addressable unit in a two-dimensional digital image system. Similarly, *voxels* are its three-dimensional counterpart. Physical manifestations of the *pixel* and *voxel* is the *dot* and *maxel*. *Dots* per inch describes the resolution of a printed hard copy. *Maxels*, commonly used in additive manufacturing, is a material element which describes a physical voxel after being 3D printed. These units represent elements for how information is represented both digital and physically. While molecules and information in fluid based systems are inherently physical, they are not volumetric nor can they be considered a solid-state material as described by maxels from additive manufacturing [29] [30] [31]. Pixels and voxels serve as information outputs generated by digital display. Dots and maxels are the physical outcomes of the information represented by the computational systems. These units are important for software and hardware domains, but are not compatible with chemical and biological realms. Adding a fifth element to serve the physical representation of biological and chemical systems is the *droplet*.

Droplets are the smallest addressable unit for fluid based information in a physical biological and chemical system. It represents the bulk, aggregate content and compositions of an unspecified volume of fluid. The term *droplet* is commonly used to refer to liquid particles in fluid mechanics and liquid handling events in chemistry and biology. The precise manipulation of liquids and droplets is essential for our ability to sense, measure and analyze molecular information. However, the capability to do so at precise microliter volumes is a fairly contemporary phenomenon. Prior to the modern micropipette, the Carlsberg pipette was used as the conventional liquid handling device, where adept scientists would use their mouths to suck fluid up in order to displace liquid for experiments [32]. It wasn't until the 1960's when the commercial production of piston-driven micropipettes first began by Eppendorf, that chemists and biologists had precise control of every droplet of reagent they interacted with. The spread of microelectromechanical systems (MEMS) in the 1980's paved way for the development of similarly small devices called microfluidics, as a liquid handling tool. Microfluidics and lab-on-a-chip technologies enable small (10-9 to 10-18 litres) of fluid be manipulated in channels up to tens of micrometers [33]. These devices have enabled researchers to create highly controlled emulsions encapsulating multiple drops within drops [34] [35]. These advancements in droplet control have enabled a variety of applications from diagnostics to chemical synthesis, highlighting the integral role droplet manipulation play in these systems. Just as pixels are programmed visual representations from the information output of computers, droplets can be generated from the information output of molecules. In this thesis, I demonstrate how droplets can be utilized as units for interacting with information in fluids from biological and chemical systems, through a multi-modal semiotic sign system outputted by information displays on everyday objects.

1.6 Research Contributions

This thesis presents a theoretical framework for bridging the gaps between molecular scale phenomena to application scale human interactions with fluid based inputs. It establishes a design framework coined *Molecular Design Interactions*, providing a methodology, theoretical foundation, design language, and toolbox for designers. Technical contributions in the synthesis, design and characterization of material based input-output devices called *Organic Primitives* offers non-chemists a toolbox for designing chemical and biological interfaces. Theoretical foundations highlights key components for interface design utilizing *droplets* as fluid information and *Organic Primitives* as information displays. The design language offers a vocabulary for integration of input-output devices into application designs. Integration of this toolbox and process is demonstrated through several application prototypes and user experience scenarios in four interaction contexts: environment, on-body, food and interspecies.

- Theoretical Framework for *Molecular Design Interactions*
- Methodology utilizing edible materials to bridge molecular scale phenomena with application and interface design
- Technical implementation of *Organic Primitives*, a toolbox of material based input-output devices with protocols for material synthesis
- Design space to achieve higher order complexity
- Design language and vocabulary
- Application prototypes

Various parts of this thesis have been published as papers, posters, and articles. Elements of this research are currently patent pending.:

- CHI 2017 **Best Paper Award** - *Organic Primitives: Synthesis and Design of pH-Reactive Materials using Molecular I/O for Sensing, Actuation, and Interaction* - http://dx.doi.org/10.1145/3025453.3025952
- CHI 2017 Video Showcase - *Pixels to Droplets: Multi-Output Display of Color, Odor, and Shape Change with Organic Primitives* - http://dx.doi.org/10.1145/3027063.3049794
- MIT Polymer Symposium 2016 Poster - *Organic Primitives for Sensing and Actuating Biopolymer Composites*
- *Hidden Code of Edible Materials*
- *Edible Media: Beyond Traditional Consumption of Food*

- *The Dermal Abyss: Interfacing with the Skin by Tattooing Biosensors*

1.7 Thesis Overview

This thesis presents a framework and design methodology for how droplets can be utilized as units for interacting with the information content in fluids from biological and chemical systems. This is achieved through a toolbox that serves to enable non-chemists to design for fluid inputs. The toolbox consists of: a set of combinable material-based color, odor and shape changing sensor-actuators coined *Organic Primitives* with instructional protocols for how to synthesize them; characterization and evaluation of the materials based on their rate, range, reversibility and compatibility with one another; design principles and heuristics for the implications of their use; techniques for activation and control of the primitives; a multi-modal design language and vocabulary; design space with strategies to achieve higher order complexity and functions; and a series of applications in the contexts of environment, body, food, and interspecies.

Chapter 2 discusses the background that motivates this work, along with the peripheral fields it builds upon. It discusses approaches to date for how chemical and biological interfacing is accomplished and the challenge for non-scientists in utilizing these tools. This research builds prior work in programmable matter and contributes an alternative approach to existing art.

Chapter 3 provides an overview of the body of research through a framework for *Molecular Design Interactions*. It goes through the theoretical foundations of key interaction elements along with stakeholder roles, responsibilities and workflows, as a future roadmap. It suggests how *Molecular Design Interactions* operate within an interaction loop as each element interacts with one another, juxtaposed with Peirce's semiotic model. A toolbox of material primitives called *Organic Primitives* is introduced, along with a design language for integrating these with objects for application. The design language exhibits how the primitives leverages the multi-modal food materials and biosemiotic signs as a communication tool. Design methodology for bridging molecular scale phenomena into applied artifact, through explorations with food is discussed.

Chapter 4 presents *Organic Primitives* as information displays in a *Molecular Design Interaction*. Color, odor, shape, and multi-modal primitives are presented to serve as an additional layer of information for designing interactions with objects. It introduces the toolbox of input-output devices which facilitates novices to design with fluid based interaction. It also features a collection of protocols and recipes for synthesizing the materials. Each primitive is presented with corresponding characterization "data sheet" based on the evaluation of their rate, range, reversibility and compatibility with one another, giving rise to their design parameters.

Chapter 5 presents droplets as fluid information that serve to initiate material display outputs. The chapter introduces the use of pH signals as a model. It describes how information in fluids excreted through different systems from the environment to the human body and foods can be addressed by *drops* as an addressable unit. It discusses a variety of fluid inputs across contexts and system scales that can be utilized for controlling the output behaviors of the *Organic Primitive* information displays. The control scheme section offers methods for activation, providing computational, coupled versus decoupled and deposition techniques to implement specific outputs.

Chapter 6 is comprised of a collection technical considerations for developing new material primitives. An experimental process for screening potential molecular sensors and actuators is outlined. A catalog of dopants and substrates has been aggregated as building blocks for new primitives. Notes on the compatibility factors of dopant-polymer integration provides hypotheses for how these materials interact with one another as I suggest potential avenues for future investigation.

Chapter 7 showcases a design space for creating higher order functionality with the material primitives. It also presents knobs for tuning the material output behavior through creating primitive subsets and permutations to achieve multi-output actuation. Techniques for synthesizing the materials across different form factors enable information displays to be created for diverse applications and object integration. It highlights how fluid driven computational devices can be created by leveraging material primitives as output devices for mechanisms using fluidic logic. Temporal sequencing and patterning techniques can be utilized to animate the materials.

Chapter 8 presents the range of applications made possible by this research. It demonstrates how integrating *Organic Primitives* onto existing objects or creating new products using material primitives can generate interactive artifacts that function as fluid information displays. The application prototypes note interfacial considerations between the primitives and the application objects to achieve adhesion and integration. Utilizing specific design language and output modalities, the applications communicate to users in affective, functional and interactive ways.

Concluding the thesis, near and far future work is discussed. It provides motivation for the nascent field of *Molecular Design Interactions* and its aims of elucidating the molecular world around us. It offers perspectives on future directions such as expanding the *Organic Primitives* library beyond pH sensing-display and the potential development of primitives beyond the organic realm.

2. Background

This chapter discusses the background that motivates this work, along with the peripheral fields it builds upon.

2.1 Interfacing with Chemical and Biological Systems

In the field of human-computer interaction, sensing and display of fluid information is achieved through the use of chemical sensors with a digital display [36] [37] [38]. These methods are desirable for recording quantitative and machine-readable data regarding the underlying biological and chemical system. However, they require additional processes to translate this information into interfaces, which are human-readable and provide meaningful value for user interaction. While there are efforts in creating electronic sensors or utilizing behavioral data for sensing biological and chemical information, their effectiveness at interfacing with these fluid based systems remains insufficient.

Biocompatible materials for sensing and actuation are becoming increasingly relevant with the rise of research towards soft, low power devices that operate close to and within the human body [39]. Recent developments like bionic contact lenses leverage information excreted from the body such as the composition of tears, for medical diagnosis [40] [41]. Ingestible devices such as Google X's pill-based authentication use stomach acids to power the biodegradable device as a temporary key for unlocking devices [42] [43]. John Rogers group and the Silk Lab have collaborated to develop implants with resorbable electronics which degrade over a defined period of time [39]. Integration of microfluidics with electronics to develop wearable tattoos for health monitoring has also been investigated [44].

These research contributions typically require a background in chemistry or biology. Emerging DIYbio communities have utilized strategies such as adapting household appliances into functional lab equipment in order to hack biology in their kitchens and garages [45]. There have been efforts to increase accessibility of synthetic biology through standardized DNA building blocks called BioBricks [46]. Miniaturized lab equipment and educational kits such as the Amino and Bento serve to lower the barrier to entry for molecular biology [47]. However, genetically engineering organisms for sensing and actuation currently remains difficult in practice, as methods are often irreproducible due to variability across organisms [48] [49] [50]. To facilitate human interaction with biological and chemical systems, an accessible toolbox of input-output devices can be achieved through molecules synthesized into material primitives. As biotechnologies continue to permeate the human experience across systems, questions involving how to enable human scale interaction of such molecular scale information become increasingly important.

2.2 Programmable Matter

A range of fields from biology to robotics have sought to implement a parallel feat - of engineering matter that can be programmed to change its physical properties. Initially introduced as a manifesto by Toffoli and Margolus in 1991, research in reconfigurable modular robotics, material computation, stimuli-responsive materials, self-assembly and synthetic morphogenesis represent a body of approaches varying in scales, with an identical vision as programmable matter [51]. Each approach utilizes different tools and component parts to implement their building blocks, and offer specialized application possibilities that are specific to their systemic regimes. In biology, morphogenic modules have been utilized as parameters in synthetic morphogenesis for the genetic modification of organisms for tuning or functionalization of biomaterials [52]. Self-assembly of DNA biomolecules have been utilized as a method for folding single-stranded DNA molecules into two-dimensional and three-dimensional nanostructures, capable of functioning as biosensors and drug release devices [53] [54] [55]. In robotics, M-Blocks are autonomous, electromechanical cubes that utilize angular momentum to enable the self-reconfigurable modules that move, jump and connect to create a range of forms capable of operating in harsh environments [56], [57]. Aeromorph draws from the field of soft robotics and utilizes heat-sealed inflatables to create pneumatically-drive shape changing materials for applications from haptic gloves to smart packaging [58].

Properties such as color, conductivity, temperature and elasticity can also be changed as a means for achieving specialized functions. In stimuli-responsive materials, Poly(N-isopropylacrylamide) functionalized spiropyran polymers capable of responding to temperature, light and pH were produced to act as logic gates [59] [60]. In applied arts and design, smart materials have begun to influence design practices in architecture, fashion design, industrial design, and interaction design. For example, designer Dahea Sun's Rain Palette utilizes color changing pigments to create textiles that indicate the acidity of the rain through color change[61]. Architect Achim Menges utilizes environmental conditions as parameters to drive material computation in HygroSkin, a wooden climate-responsive pavilion [62] [63].

A key feature of advancing the programmability of matter is to enable the multi-functionality of physical matter. The emergence of smart phones have centralized many single-purpose tools from notepads to calculators into multi-functional devices. Matter which can be programmed to change from round to cubic or rollable to stackable, offer implications for producing a myriad of functionalities in the same material [64] [65] [66]. While these features offer provocative technical challenges, questions involving how programmable matter can be integrated into the production of everyday goods are overlooked. Currently, designers play a large role in the creation of everyday objects from food containers to furniture, cars and clothing. Facilitating these stakeholders in the adoption of programmable matter can play a crucial role in its development, dissemination and future.

3. Framework for *Molecular Design Interactions*

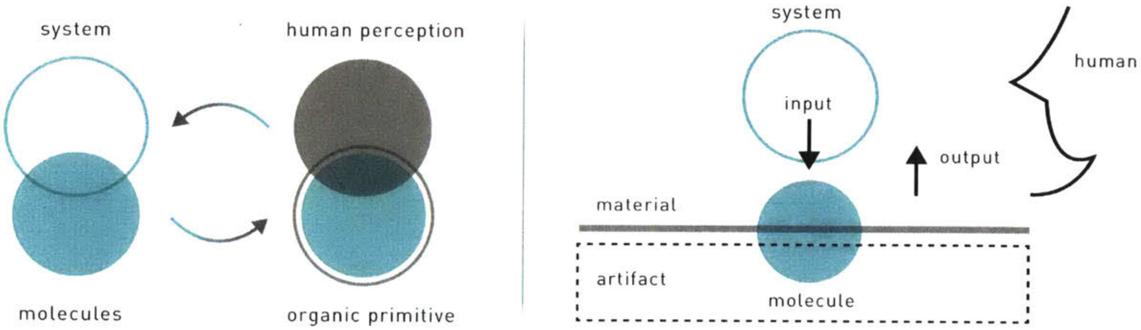

Figure 3-1. (left) High-level diagram describing how ecological systems such as the environment, food, and body produce molecules which interface with *Organic Primitives* to produce human-readable outputs. (right) Detailed diagram of these interactions.

Molecular signals are excreted from different systems in the form of fluids as they provide insight into their states and processes. Figure 3-1 describes how *Organic Primitives* hijack the materiality of artifacts and objects to provide human-perceptible outputs from systems. This allows systems to 'communicate' to humans, altering their perception and transforming their relationships with them.

This thesis investigates methods for developing a system for designing human interactions with molecules. The goal of this work is to:

1. Demonstrate the possibility of user interaction with molecules
2. Establish a reproducible process which can be scaled across a range of molecular signals
3. Enable designers and non-chemists to create molecular design interactions for end users

3.1 Theoretical Foundations

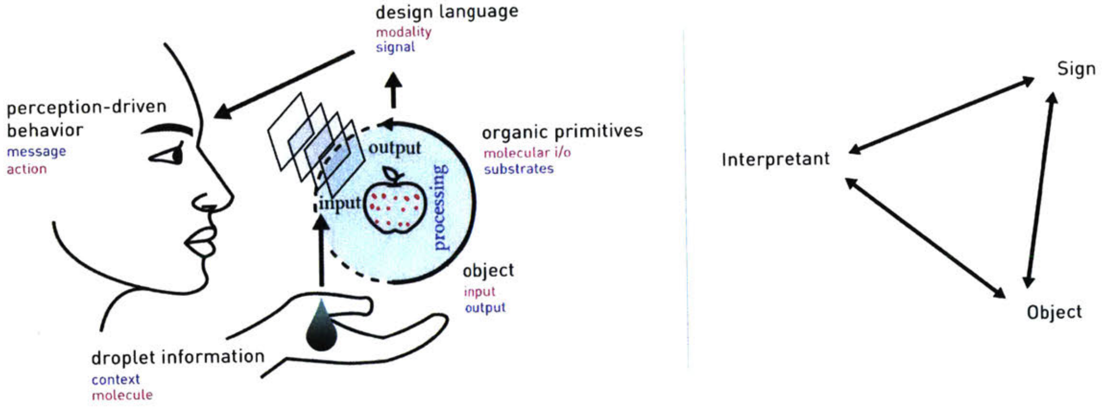

Figure 3-2. Interaction loop for *Molecular Design Interactions*(left) juxtaposed with Peirce's triadic semiotics model (right)

Philosopher Charles Sanders Peirce's work in semiotics denotes that "all this universe is perfused with signs, if it is not composed exclusively of signs" and that logic is a formal semiotic structured with a triadic relation where a sign in relation to an object and interpretant forms meaning [67] [68]. My hypothesis of human interaction with molecules is an implementation of Peirce's semiotic model[1] as shown in the above figure 3-2. The information input of a droplet activates the object and its encoded output properties to give rise to the sign or design language as the user serves as the interpretant to form meaning through the perceived sign and context, as he/she draws from sociocultural associations.

The output pattern generated by the designer of the *Organic Primitive* encodes specific output behavior onto the object to create a sign or design language to be interpreted. The designer does this with the interpretant or user in mind, whom carries prior associations and perceived notions regarding its meaning. For example with odor release, the specific odorant descriptor and quality will dictate how a human observer may interpret the signal. If the smell is a strong sweet odor, the apple is perceived to be a 'sweet apple.' With color change, specific color and patterning will dictate what the sign and interpretation may be. Figure 3-2 shows an apple with red spots, displaying a sign of toxicity as transferred from the spotted patterns of *amanita muscaria* mushrooms. The design language serves to communicate additional layers of information embodied by the object to the user to transform their relationship with the context or system.

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[1] Semiotics is the study of signs and symbols for communication and production of meaning.

3.1.1 Anatomy of a *Molecular Design Interaction*

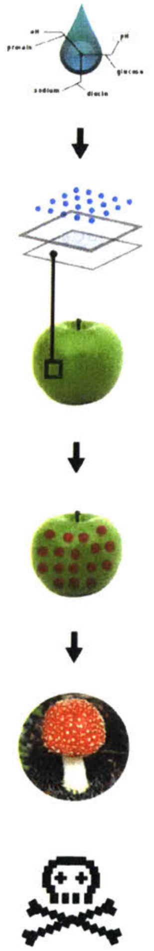

Figure 3-3. The information contents of a liquid becomes human readable upon it's interaction with a material primitive. The primitive can be integrated through the materiality of an object such as an apple, to yield an object based information display and interactive artifact.

A *Molecular Design Interaction* encompasses: the interactive artifact, material primitive, and design language connecting human meaning with molecules in systems. As shown in figure 3-3, the design interaction is activated by a liquid droplet that serves as the input. This *droplet* can possess a number of molecules and chemical contents - from broad signals such as pH or hyper specific signals such as the presence of a particular molecule or functional group such as a dioxin. When the *droplet* makes contact with the material primitive onto an object's surface, it will trigger property changes to produce a human-readable message. The output behavior combined with the object's contextual attributes generates meaning.

Droplets

In *Molecular Design Interactions*, droplets are units of information for interacting with information in fluids from biological and chemical systems. When droplets interact with information displays or *Organic Primitives*, they function as the information output of molecules. Molecular output can be either coupled or decoupled from fluid input signal. Further elaboration and discussion can be found in chapter 5.

Information Display

Organic Primitives input-output devices or material primitives, paired with design language generates an information display. These can be integrated with objects to form an interactive artifact. When a fluid input makes contact with an interactive artifact, it activates a human-readable sign for human perception.

3.1.2 Workflow for Designers and Developers

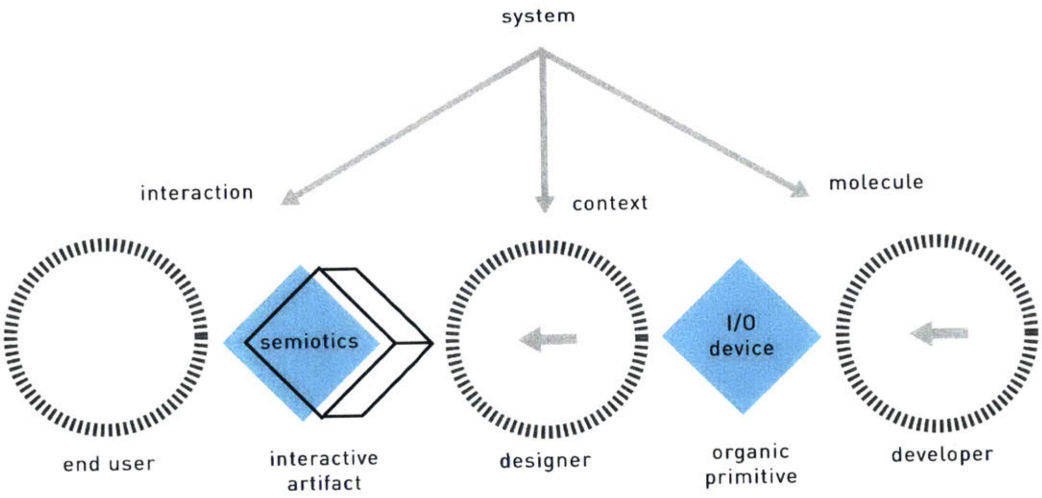

Figure 3-4. Diagram showing relationship of stakeholders and roles contributing to *Molecular Design interactions*.

This process integrates the role of a designer, developer and end user into workflows to form a sustainable design practice (figure 3-4). A developer, possessing a working knowledge in chemistry would formulate new *Organic Primitives* for use by designers. A designer without prior knowledge in chemistry can utilize *Organic Primitives* to create perceptual links and design vocabularies that are applied to objects between a natural system and the end user. The end user in turn forms new relationships with selected systems through their experiences with the molecular design objects.

The designer takes part in dictating the perception of objects by designing signs and symbols. Designers accomplish this by examining layers of information of objects in a given context, in relation to fluid based inputs. Fluid based inputs are information contained within the composition of molecules excreted by natural systems such as the environment, organisms and, foods. Addressable units of fluid information inputs are *droplets*. Droplets activate material based information displays. Designers create sign systems by utilizing the design language and vocabulary provided by this research. Material primitives and output properties are selected based on what a designer is interested in communicating with the user, through the design of the interactive object.

Material primitives are created by developers who synthesize and characterize new *Organic Primitives*. A material primitive is a material based input-output device that produces a human-readable property changes based on fluid inputs. Utilizing a library of dopants and substrates, developers create material based sensor-actuators for designers to use. Developers examine droplets of fluid based information to determine material primitive characteristics. The workflows and products of the designer and developer in this framework will be discussed in the following subsections.

Developers generate Primitives for Design

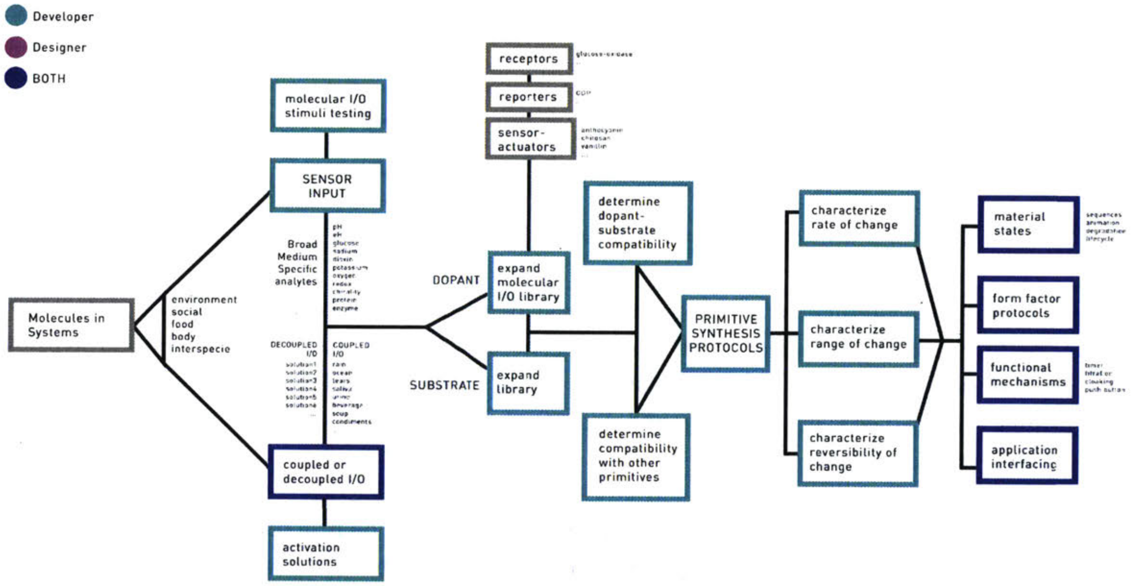

Figure 3-5. Development process of an *Organic Primitive* from a developer's perspective.

While the material primitives are intended as building blocks for use without requiring prior knowledge in chemistry, the development of new primitives will require developers who possess a moderate background in chemistry. Developers generate the recipes for activation solutions as prepared fluid inputs and testing (figure 3-5). In the case of pH solutions, these involve pH 2 through 10 solutions using household consumer reagents such as citric acid, sodium bicarbonate, sodium citrate and sodium hydroxide. New molecular I/O are identified, evaluated and developed into *Organic Primitives* by synthesizing them into solid state materials, where their output parameters are to be characterized.

Designers build interfaces with Material Primitives

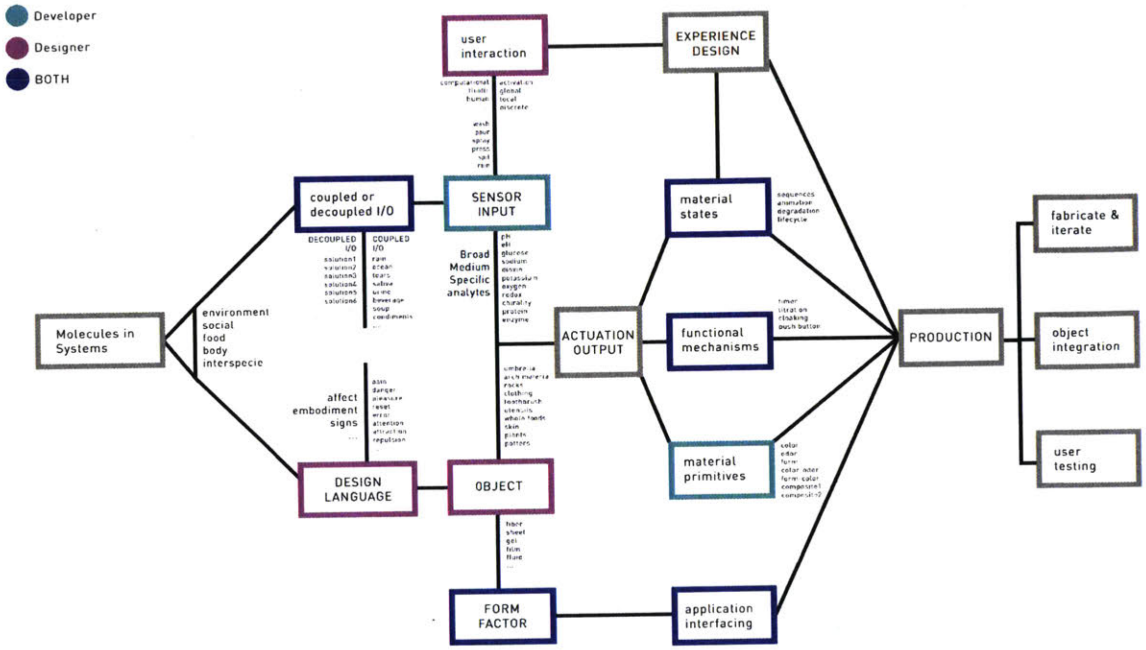

Figure 3-6. Design process and workflow for a Molecular Design Object from a designer's view.

Designers construct interactions between users and their everyday environment (figure 3-6). From utensils to furniture and vehicles, these constructions act as social-cultural artifacts that activate meaning and behavior. By developing a medium to aid designers, everyday object interactions can be transformed into carriers of chemical and biological information from environmental, on-body, organismic, and food systems.

In *Molecular Design Interactions*, designers will be involved in designing how users might experientially engage with an interactive artifact or the chemical signals of a system, to develop social and cultural meaning that contributes to a user's perception and relationship with the system. Whether a designer may begin with project brief from a client or an idea for an object, the design process is a series a decisions and begins by distilling the design objective to determine "what system am I designing for?"

3.2 Design Language for Encoding Perception

3.2.1 Molecular Associations

Figure 3-7. Interpretations of fluids and molecules can serve as a vocabulary for design.

The vocabulary for design can be innate or modifiable interpretations of molecules (figure 3-7). Innate interpretations are typically biological and difficult to change as they are universally shared among humans across prolonged periods of time. Modifiable signs are associations that are shaped by culture, trend and society which can shift through a user's experience. Signs do not require human intent to be created. The "smell of the rain" is encoded to us through rain water falling upon asphalt (in modern roads) to generate the association. Smell of the rain, as associated with asphalt water did not generate out of intent nor instinct, but through a sign (asphalt water molecules) in relation to an object (rain on road), activated by the human interpretant who witnesses this scene through their sensory organs (nose, eyes, ear).

Innate Interpretations of Molecules

Bad smell is toxic

Some chemical experiences give us an innate biological reflex. Sulphuric compounds are associated with a grotesque rotting smell. We carry toxic associations on sulphuric compounds largely due to our manure, a waste product ejected from our bodies, possessing these molecules. These compounds are produced when bacteria decompose organic matter such as food items. The natural gas industry has leveraged this by injecting sulphuric compounds such as mercaptan into their natural gas systems to communicate the toxicity of natural gas, in the event of a leak.

Symbolic Human Self-Perceptions with Body Fluids

Our perceptions of molecules are linked to an embodied perception of these chemicals excreted from our bodies. Symbolically, different body fluids project meaning that may be more universally shared . With tears denoting emotional outcome, sweat in hard work and blood as pain or embodiment. Whether it's culturally constructed, a form of interspecies communication, or primal associations, we live in in a landscape of chemical perceptions that are shaped by both innate and fabricated definitions. We can use this as a principle to create a design language and vocabulary.

- Seeing blood as seeing pain: An embodied sense of pain is experienced when movie characters exhibit blood spill, as a way for filmmakers to sensationalize the physical pain of the characters.

- Tears as emotional outpour: We view sadness and happiness through the physical outpour of tears. These excrements are innate from the moment of birth, despite culture or ethnicity.
- Sweat in hard work: When the body exerts physical activity, sweat is produced. This production of liquid from the body is associated with physical labor.

Encoding Modifiable Chemical Perceptions with Signs

Signs can also be modified, intervened and designed. A prominent example is that of the artificial food flavoring. While real bananas possess hundreds of flavor molecules, isoamyl acetate is the widespread molecule used as the additive to make artificial banana flavored products. Modern commercial banana varieties are derived from a single Cavendish genetic clone. Isoamyl acetate has been said to not resemble the actual flavor of banana. This is because isoamyl acetate is not as prevalent in Cavendish varieties as compared to the historical Gros Michael variety [148]. To modern humans, isoamyl acetate has become the representation for the taste of all bananas through this single molecular abstraction. In actuality, what we are tasting in artificially flavored banana products is more like a banana from the past.

Encode sensory cues to function

The perception of "clean" is shaped by culturally encoded use of certain odorants paired with the functionality such as the odorants used by chemical engineering companies and flavor fragrance houses for chemical solvents to remove stains from surfaces. Lemons are commonly to communicate cleanliness through sensory modes. Household chemicals such as Lysol all-purpose cleaning solution and Mr. Clean Liquid Muscle Multi-Purpose Household Cleaner utilizes yellow coloring and citrus odors to signal associations with lemons. By coupling visual-olfactory cues with the function of chemical solvents, an association of lemons and cleanliness is generated.

3.2.2 Emergence of Meaning

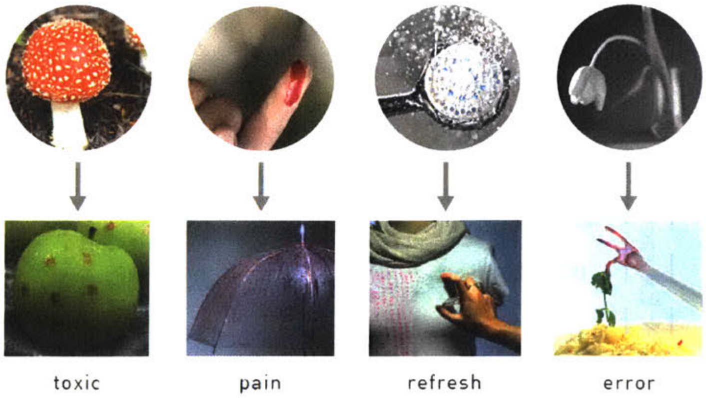

Figure 3-8. Interactive artifacts integrated with *Organic Primitives* (bottom row) utilizes semiotic signs of toxic mushrooms, bleeding, water mist, and wilted flower (top row) to communicate messages of toxicity of toxicity, pain, refreshment, and error.

The design language allows designers to utilize the color, odor and shape changing properties of *Organic Primitives* as a tool for encoding innate or modifiable signs and messages on everyday objects. As shown in figure 3-8, signs from biological and chemical systems are translated as a design language to encode notions toxicity, pain, refreshment, and error. Coated with *Organic Primitives,* the objects display patterns that serve to communicate to users when washed, sprayed, and used. Transformed into a sensor-display, the apple communicates that it has been contaminated during transport by displaying spotted patterns mirroring the toxicity of *amanita muscaria* mushrooms when washed. The umbrella bleeds in the presence of acid rain, to communicate that "the sky is in pain." The shirt designs initiates a color change when it is refreshed by the spray of perfume. The fork wilts, refusing to pick up food when a user has eaten too much. Drawing from the objects' contexts, symbolism of fluids, and signs created through the integration of the *Organic Primitives* property changes, a design vocabulary emerges as a mode of communication.

3.3 Design Methodology: Molecular Phenomena to Applied Artifact

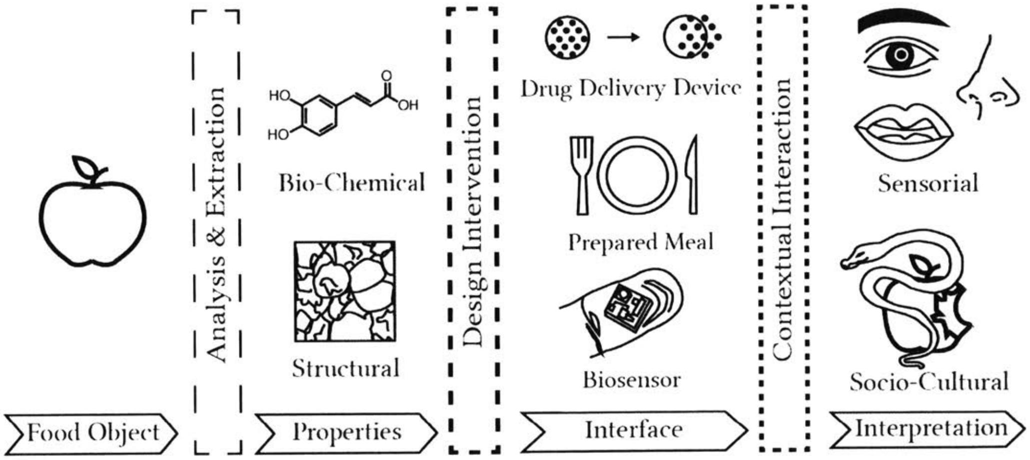

Figure 3-9. Design process for bringing molecular phenomena and properties of edible materials into design language for application interaction. *Illustration courtesy of Vicky Zeamer from our joint paper on "Edible Media."*

The methodology brings together molecular phenomena into application and user interface design. This approach of synthesizing color, odor and shape changing material sensor-actuators with edible materials enable a variety of advantages and design opportunities as these systems: (i) have self-contained functionality as a sensor, actuator and energy source; (ii) can be manifested in different form factors and states of matter, from liquid to solid to vapor; (iii) can be integrated into both biological and electronic systems; (iv) are biocompatible, biodegradable, and edible; (v) are compact, soft, muted, and unobtrusive; (vi) open up additional modalities Including taste and smell.

The process involves utilizing food objects as a medium for interface design (figure 3-9). Upon extraction of biochemical molecules and structural biomaterials from the objects, they are analyzed for their physical and perceptual properties. Interfaces will then be created by exploiting their properties as user interactions shape sensorial and sociocultural perception, enabled by the food materials.

3.3.1 Principles for Designing Interactions with Molecules

The distinct capabilities enabled by this platform includes the ability to "hack" the materiality of objects to transform objects into both a sensor and an information display. Because of the organic nature of these input-output devices, fluid contents as such as raindrops, ocean water, body fluids, food sources, and organismic excretions can be utilized for control. Using these inputs to initiate color, odor and form changes through material primitives, sensory experiences can be augmented between edible, environmental, interspecies and on-body interactions.

Hacking Materiality of Objects

The information displayed on interfaces about our bodies, food, environment and organisms are often divorced from the actual systems themselves. This creates a cognitive dissonance between the information source and the contexts where we engage with them. By utilizing pre-existing objects as sensor-actuator platforms, we can take advantage of the contextual information they embody for designing new relationships with the systems they represent. Figure 2 shows an example of how *Organic Primitives* can enable an additional layer of information on objects - transforming ordinary objects to encode meaning beyond their initial functions.

Utilizing Fluid-based Inputs

Natural inputs from different contexts can be used to activate the material interaction by utilizing information contents such as pH, glucose, and sodium, just to name a few. This includes inputs through rain drops, ocean water, body fluids, food sources, and organismic excretions from plants and microbes. More precise and de-coupled signal outputs can be achieved through the use of prepared solutions. Such solutions can be integrated with mechanical and electronic systems to develop machine-mediated inputs. Selection of chemistry for prepared solutions can enable researchers and designers for an added level of control in the material's reaction time and buffering capabilities when activating an *Organic Primitive*.

Information Representation and Sensory Augmentation

The ability to have a natural object express an additional layer of information about itself through its materiality can afford more expressiveness in everyday objects. *Organic Primitives* enable sensory augmentations through odor, color, shape, and taste change, where novel behaviors can be encoded into every day objects. Figure highlights how odor-changing utensils can be used to simulate a sweet perception of food, instead of adding sugar.

Taste and Odor Affordances

In the case of pH, low pH materials are typically more acidic and tend to taste sour. High pH possesses higher salt content so is considered to be salty in taste. However, further investigation will be required in order to assess the taste parameters of our material primitives. Also, by manipulating odor attributes in a food experience, it effects the way something tastes. A large part of taste perception can be attributed to characteristics in odor through flavor perception.

3.3.2 Properties of Edible Materials

Food materials possess attractive features that can be exploited for interface design such as multi-modality and stimuli-responsiveness. Here I discuss some examples of work from a variety of disciplines which have leveraged edible materials for sensing, actuation and degradation.

Molecular Sensing Capabilities

Often times these chemical mechanisms are derived from organisms that are commonly consumed as food. Plants and animal tissue have been used for developing biosensor electrodes due to their biocatalytic capabilities. For example, Vincke et al utilized cucumber as a biocatalyst to act upon ascorbate (vitamin C) for sensing 02 compounds [69]. Enzymes are also commonly used for biosensing applications. One such example is glucose oxidase enzyme for the detection of glucose (sugar) [70]. Glucose oxidase enzyme is often extracted from *Aspergillus niger*, a fungi found on many food items like apricot and onions.

Passive Actuation

Edible materials can exhibit movement and motility through a range of mechanisms including leveraging surface tension with relative evaporation rates, stimuli responsive properties of particular food materials, and gas generation through C02 reactions. The physical nature of a material's composition can lend itself to complex reactions, enabling researchers to leverage physical phenomena for information processing. Water mixed with food dye have exhibited sensing and motility behaviors termed *artificial chemotaxis* due to the relative evaporation rates and surface tension of two-component fluids. Researchers have exploited these properties to create autonomous fluidic machines including a vertical droplet oscillator, sustained droplet chaser, self-aligner, and self-sorting droplets [71]. The lewis group in Harvard have created passive actuating soft robots through embedding hydrogen peroxide as a liquid fuel [72]. With the same principle, edible materials commonly used in baking often incorporate rising agents like yeast and baking soda to enable dough to rise. As an example, yeast and baking soda (sodium bicarbonate) can be utilized as a food-safe, passive mechanism for building actuation devices. Reaction between baking soda (sodium bicarbonate) and citric acid (food additive derived from citrus fruits) generates Carbon dioxide (C02) gas, akin to carbonated beverages. Yeast requires sugar as an energy source, but as a byproduct of their metabolism they also generate C02. Whether chemically, biologically or physically, there are innumerable methods for creating actuation mechanisms using food.

Biodegradable, Ephemeral, Edible Devices

Many applications for interaction with chemical and biological systems are inherently ephemeral and biodegradable. Electromechanical hardware designs have also begun to exploit these properties. A number of entities such as Bettinger's group at Carnegie Mellon University, Proteus Digital Health, and Google X have also developed ingestible devices [42] [73] [43]. Drone chassis made from mycelium (mushroom) have been developed to create an environmentally friendly UAV [74]. Ephemeral user interactions have leveraged food materials and chemical solvents to create bubbles and smoke based displays [75] [76].

In biomedical engineering, materials which are biocompatible and have the ability of being adopted by cells within body without rejection is an important factor for device development. A number of biopolymers from food have been research for creating devices that can function inside the body. Carrageenan, alginate, gelatin, cellulose and agar have all been researched as substrates for tissue engineering scaffolds and drug delivery [77] [78] [79] [80]. In synthetic biology, organic compounds are used to develop synthetic minimal cells as biosensors, lipsome bioreactors, protocells and chemical messengers with natural cells [81] [82] [83]. These molecular machines are often made from organic compounds - such as phospholipids, cholesterol, and fatty acids to encapsulate transcription/translation machinery and achieve controlled gene expression [84] [85].

As exhibited in this section, edible materials have long been exploited for a range of biocompatible applications beyond sustenance. Many of these examples utilize the physical properties of food materials, but overlook its perceptual and sensorial capabilities.

3.3.3 Mattermath: Repository of Food Experiments

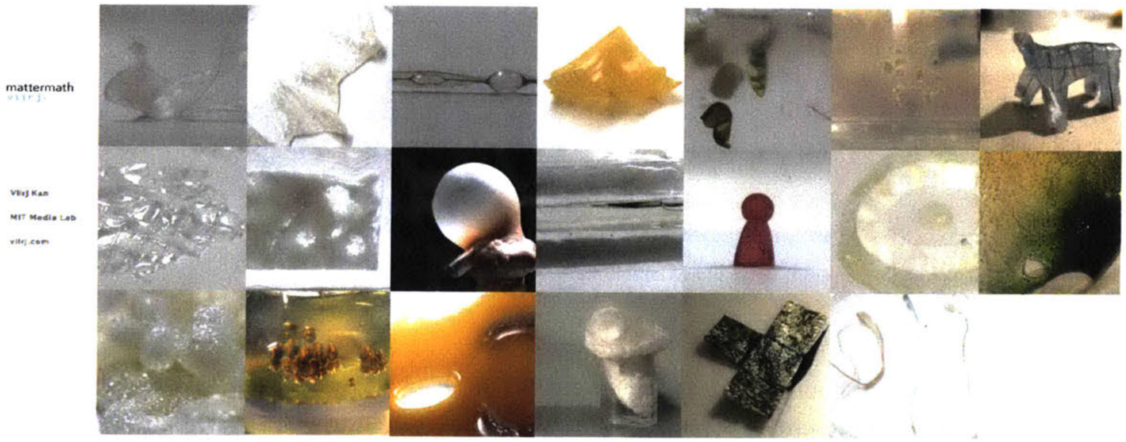

Figure 3-10. Screenshot of the Mattermath website

Initial food hacking experiments were done in 2014 with my colleagues Judith Amores, Dhruv Jain, Christophe Guberan, Penny Webb, Yujie Hong, and Chikara Inamura. At the time, we called these efforts *Computational Food,*a process of *uncovering the hidden code of edible materials*. These experiments ranged from raw material polysaccharides, lipids and proteins to processed food materials such as pastas, seaweeds, and chocolate. Our attempt at probing their properties involved encapsulating, cooking, scorching, microwaving, popping, patterning, mixing, compositing, and electrochemically reacting the edible materials with one another. These are being cataloged and documented in a online web-based repository called Mattermath. Figure 3-10 shows an initial version of the website.

3.4 *Organic Primitives*: Toolbox of Input-Output Devices

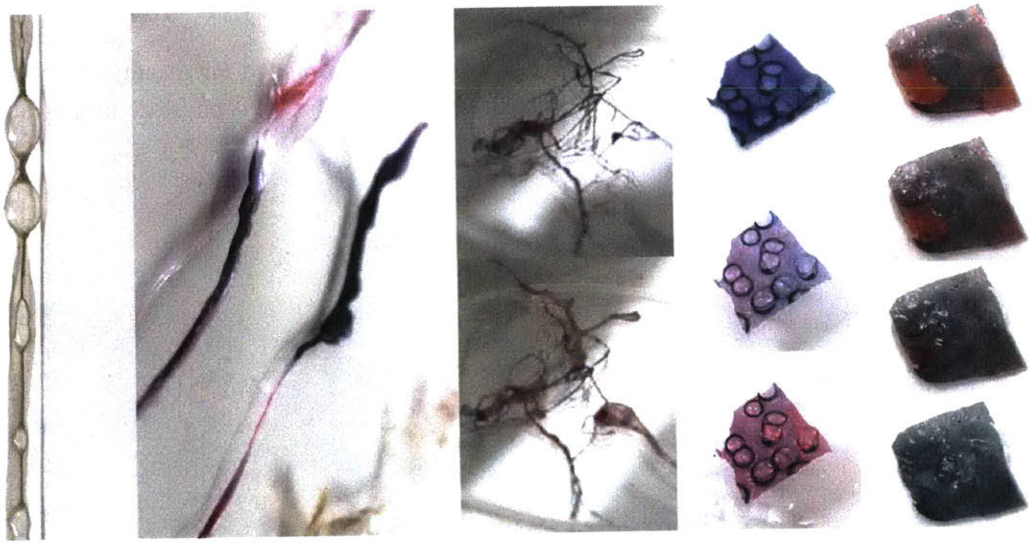

Figure 3-11. Selected material primitives and form factors for use as input-output devices.

These material based input-output devices were designed to enable designers without extensive chemistry backgrounds to create a variety of multi-modal material displays by simply selecting and combining responses of interest (figure 3-11). The design of this toolbox for interface design addresses five key criteria:

- Solid-State: liquid state pH-reactive phenomena of the molecules must be synthesized into solid state materials.
- Flexibility: materials should have the ability to be synthesized into a range of form factors, without drastically differing in their fabrication processes.
- Accessibility: they should be safe for handling, producible in a kitchen environment and utilize reagents that are easy to attain.
- Human-Readable: output properties should be within receptive fields and thresholds of human perception.
- Combinability: material primitives should be compatible with one another to yield multi-property inheritances.

This thesis demonstrates how these criteria are addressed through the development of the material primitives. It offers a set of combinable material-based sensor-actuators coined *Organic Primitives*. It consists of color, odor and shape changing material primitives that sense contents within fluids and convert them into sensory information. The material primitives can output a spectrum of colors, different degrees of shape deformation, and switch between odorous and non-odorous states based on pH value. The scope of this thesis focuses on sensing pH in fluid as a starting point, with the goal of developing a model methodology for future sensor-display development.

4. *Organic Primitives* as Information Displays

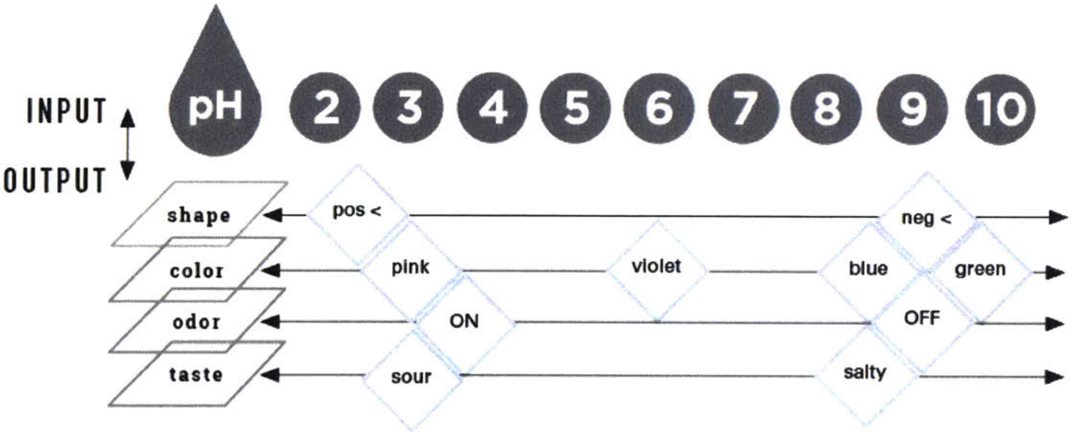

Figure 4-15. Diagram of pH inputs and outputs from pH 2 - 10.

pH value is an intrinsic attribute among all fluids and serves as an important indicator in a broad range of systems, from nanoscale microbial systems to planetary-scale marine ecosystems. For this reason, I begin the development of these material sensor-actuators with pH-reactive primitives.The scope of this thesis focuses on sensing pH in fluid as a starting point, with the goal of developing a model methodology for future sensor-actuator development.

Many organic molecules respond to pH, but only a subset of these are edible and yield human-readable changes like color, odor, shape, and taste (figure 4-15). The material-based sensor-actuators begin with the selection of dopants and biopolymer substrates. Within this approach, I utilize the Brønsted-Lowry acid-base reaction as a core driver of the material transformations due to the rapid, reversible, and bidirectional nature of the reaction.

In this section, color, shape, and odor changing primitives are introduced with protocols for their material synthesis as films. Characterization results for their output as a function of pH are discussed. Proposed output parameters of the *Organic Primitives* are discussed, to inspire users to build upon this logic and contribute future design parameters for this platform. Finally, the capability for multi-modal primitives are demonstrated through mixing, layering and panelling methods.

Output Characterization Methods

The individual outputs of the Organic Primitives were evaluated in the form of films. The test samples were all prepared by pouring the formulations into small polystyrene petri dishes, purchased from VWR. The samples were dried overnight beneath fans before being removed from the dishes. pH solutions were made using food-grade materials to ensure safety and accessibility: citric acid (H3Cit) an acid derived from citrus fruits; sodium citrate (NaHCitrate), a common food additive; sodium bicarbonate (NaHCO) also known as baking soda; sodium hydroxide (NaOH) or lye, commonly used for food preparation; and deionized water (H2O). These materials and concentrations do not require special facilities or tools, as they comply with the Federal Food and Drug Administration's (FDA) generally recognized as safe (GRAS) listing [86]. The solutions were tested and verified using the Oakton pH5+ EW-35613-52 pH meter, as well as pHydrion litmus strips.

4.1 Color Changing Primitive

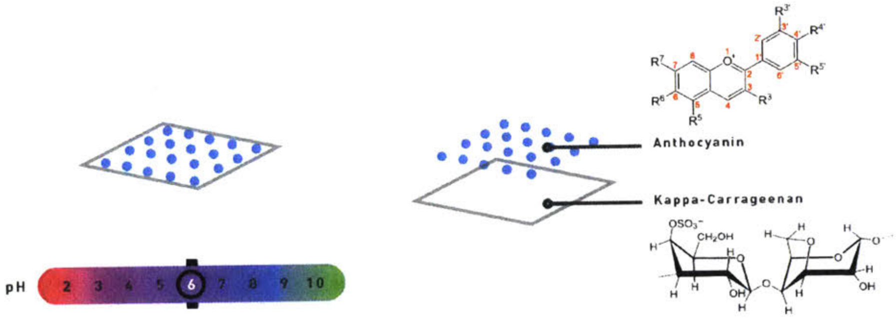

Figure 4-1. Output behavior for color primitives and dopant-biopolymer combination composed of the primitive.

Kappa-carrageenan was used as the base material for the anthocyanin color-changing films (figure 4-1). Electrostatic interactions between the two molecules allow them to create a stable compound; kappa-carrageenan is an anionic polysaccharide, while anthocyanin is cationic. In particular, the bond comes from negatively-charged sulfate groups of kappa-carrageenan interacting with a positively-charged heterocyclic ring of oxygen atoms found in anthocyanin [87].

4.1.1 Color Output Characterization

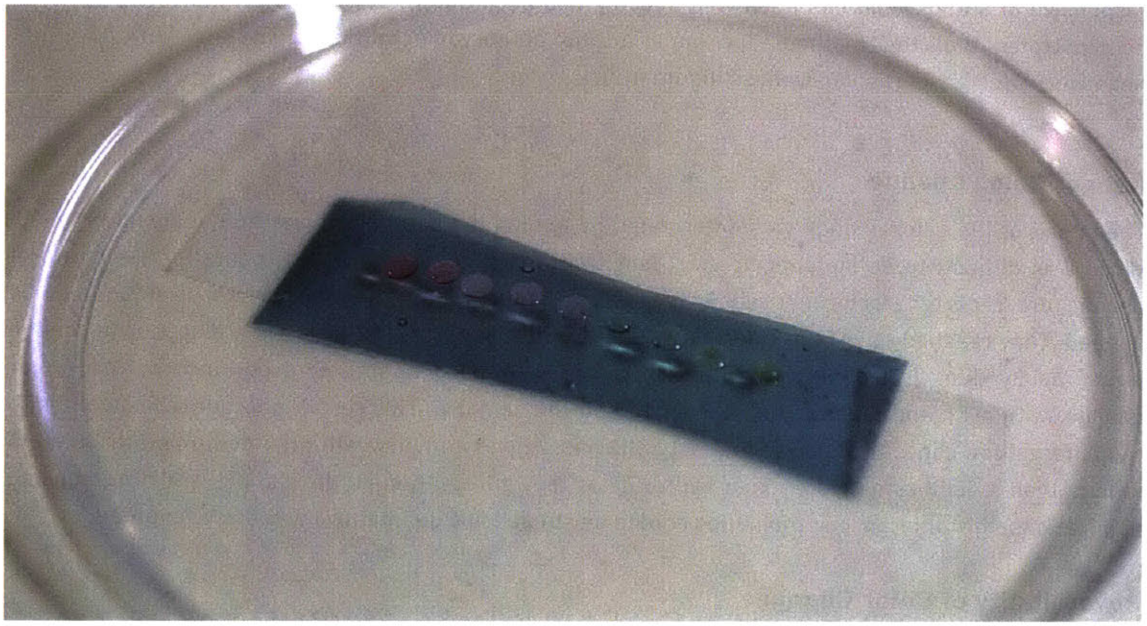

Figure 4-2. Anthocyanin was employed as the color changing dopant with Kappa-Carageenan biopolymer substrate.

The color-changing samples were prepared using 1.5%w/v kappa-carrageenan in deionized water doped with 0.1% w/v anthocyanin, purchased from Modernist Pantry and Enasco respectively. Samples were tested with pH 2-10 solutions. 7.5 mm squares were cut from the film, and sprayed by hand until color change was uniform across the sample. Once uniform, the color did not change, indicating chemical equilibrium. The samples were allowed to dry and then analyzed using an X-Rite CMUNPH spectrophotometer, providing a CIE L*a*b* color value for each sample.

Range and Spectrum of Color Change

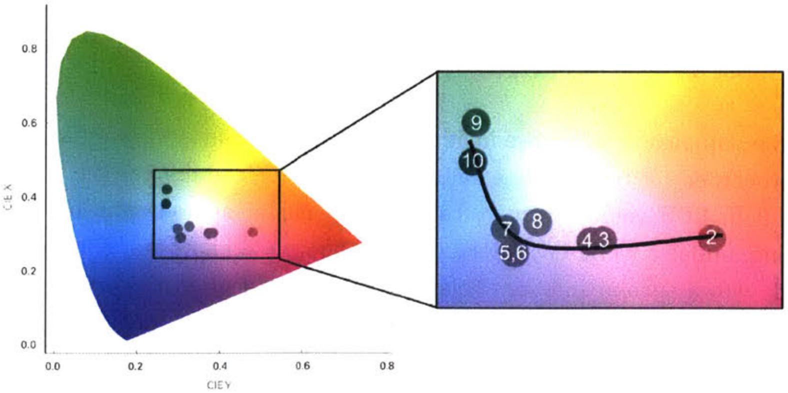

Figure 4-3. CIE L*A*B* color space of color change ranges of *Organic Primitive* sample made with 1.5% w/v kappa-carrageenan doped with 0.1% w/v anthocyanin.

Depending on pH, the sample color changes from redder tones to green (figure 4-2). The visual pH responsive of anthocyanin has been well documented because of its use as a pH indicator; this trial demonstrates that the methods for materializing anthocyanin into a solid-state film have not negatively affected its responsiveness (figure 4-3).

Rate of Color Change

The speed of the color change was determined by filming the color primitives and applying pH 2-10. While it is difficult to define specific start and stop times, qualitative assessments for their rate of change are reported. Color noticeably begins to change within 11 milliseconds after solution is applied. They reached their final color between 50 to 90 seconds. Acidic pH's 2-4 are appear to change more rapidly than 7-10. Samples with pHs closer to 7 didn't noticeably experience a rapid color change, with pH 5 and 6 as the slowest among all the samples. This result is largely consistent with the different ion concentrations in the test solutions. Any asymmetry with pH 7 could be attributed to different ion mobilities of hydrogen or hydroxide as they diffuse through the sample. For time-sensing applications, this difference in mobilities should be studied in a quantitative way and accounted for.

Reversibility of Color Change

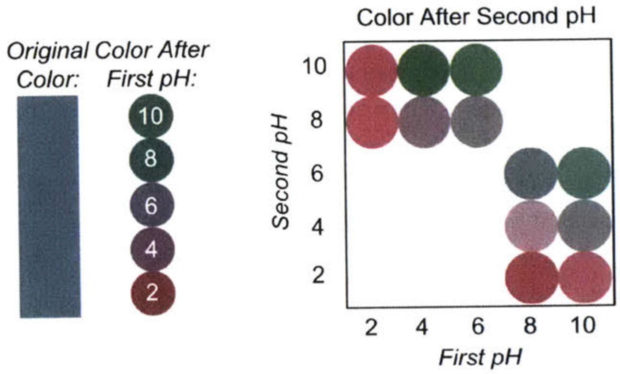

Figure 4-4. Graph showing reversible effects of layering pH.

For this study, the samples were sprayed with the first pH solution, and given an hour to equilibrate. This color was recorded for the baseline value. After an hour, the dry sample was sprayed with the second pH solution. RGB values were extracted from digital photographs of samples taken with controlled lighting, under a white hood. Results from figure 4-4 show that only certain pH responses can be reversed, with a second solution. This study shows that pH 2 was unable to be reversed (the column remains pink, though slightly less vivid). However, every other pH can be influenced by adding a second solution. This is due to the constraints of the substrate biopolymer contained in this primitive, as anthocyanin color changes in fluid are entirely reversible reactions. I hypothesize that this incomplete reversibility is influenced by buffering effects of the polymer and slower diffusion rates in solid materials.

4.1.2 Material Synthesis

Protocol for synthesizing color changing films

Making 0.1% Anthocyanin Solution:

1. Suspend 0.5 g anthocyanin in 500 mL water
2. Mix on magnetic stir plate until fully dissolved.

Making Anthocyanin- 1.5% Kappa Carrageenan Gel Solution

1. Add 7.5 g of kappa carrageenan into the 500 mL anthocyanin liquid.
2. Place the bottle into a pot of hot water.
3. Place pot above the magnetic stir plate to mix until a gel solution is achieved.

Uniform Color Changing Films

1. While warm, pour the Anthocyanin-Kappa Gel Solution into a large petri dish or desired mold.
2. Place the dish in a cool location to gel and under a fan at 4.5" high with uniform air flow to form a film.

4.2 Odor Changing Primitive

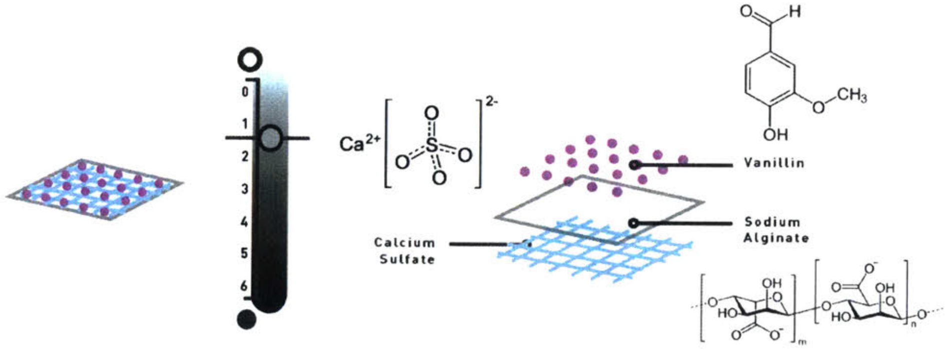

Figure 4-5. Output behavior for odor primitives and dopant-biopolymer-crosslinker combination composed of the primitive.

Vanillin was encapsulated in an alginate gel to form the scent-changing solution (figure 4-5). Sodium alginate was cross-linked with calcium sulfate right before mixing, creating a tangled matrix to hold the vanillin molecules. As discussed in future sections, the alginate gel behaved neutrally through the entire tested pH range, making it an optimal compositing material. The specific interactions between vanillin and its matrix are not yet well-characterized, but the trials suggest the gel has little effect on vanillin volatility and scent release.

4.2.1 Odor Output Characterization

To evaluate the detectability and limitations of human perception upon the odor-changing primitives, I conducted a user study with 14 subjects, 4 male and 10 female. I had participants rearrange nine 1" x 1" samples of pH activated odor material from pH 2-10, based on a categorical scale of odor intensity. The odor-changing samples were prepared by creating a stock solution of 1 g vanillin (Sigma Aldrich) dissolved in 10 mL 200-proof alcohol. We then made a solution of 0.12% v/v vanillin stock and 1.5% w/v sodium alginate in deionized water. Immediately before pouring, the sodium alginate was cross-linked by adding 15 parts calcium sulfate to 100 parts vanillin-sodium alginate solution. Samples were tested with the same pH 2-10 solutions as in previous trials.

Range of Odor Strengths

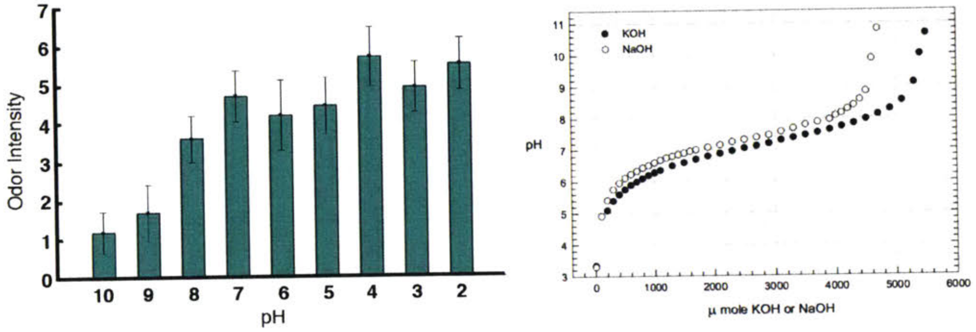

Figure 4-6. Odor rearrangement results (left) where error bar represents standard error. Titration curve (right) by Frenkel [88], showing vanillin molecule response to pH.

In figure 4-6, the results show that users can clearly distinguish the on-off switching behavior of the odor material based on pH. pH 9 and 10 were ranked as odors with no smell or low intensity, whereas samples inputted with low pH generated a stronger odor perception. The user's sample arrangements for odor intensity correlated with the titration curve and response of the vanillin molecule to pH [88].

Rate of Odor Change

Two independent researchers qualitatively assessed the rates of odor change using a timer. Odor changing films as prepared above were cut into nine 1" x 1" samples and 640uL of each pH solution 2-10 were applied to a film. pH 2-8 exhibited a sweet odor released within 1-5 seconds of depositing solution. After 30 or more seconds, the odor intensified over the course of hours and maintained its smell after dried. With pH 9-10, approximately 30 seconds after a pH solution is deposited, users were able to establish that the material had no smell. Within the first 30 seconds, the material possessed a faint sweet smell of vanillin. This preliminary study exhibits that each applied pH results in a different time-intensity relationship. This phenomena will be better examined in future studies, using olfactometry test methods as defined by the American Society of Testing and Materials ASTM E679 and E544 [149] and quantitative analysis with headspace gas chromatography.

Reversibility of Odor Change

Based on qualitative assessments, the odor primitives were able to be activated from a low pH (smell on) to a high pH (smell off). However, after a high pH change, the material was unable to reactivate in its smell. We hypothesize that due to the low concentrations of vanillin in our materials, each time it is activated by pH solution the odor molecules are consumed, by sublimation and evaporation into the air.

4.2.2 Material Synthesis

Protocol for synthesizing odor changing films

Making 10% w/v Vanillin Stock Solution

1. Pipet 10 mL of 200-proof ethanol into a glass vial
2. Dissolve 1 g vanillin into the vial.

Making 1.5% w/v Calcium Sulfate Crosslinking Solution

1. Weigh out 1.5 g calcium sulfate into a glass bottle.
2. Add de-ionized water until 100 mL is reached.
3. Mix the solution until dissolved. Some settling may occur.

Making 0.05% v/v Vanillin-Alginate Gel Solution

1. Weigh out 1.5 g Sodium Alginate into a glass bottle.
2. Add de-ionized water until 100 mL has been reached.
3. Mix with magnetic stir plate until a gel solution is achieved.
4. Pipet 50 uL of Vanillin Solution into the Sodium Alginate gel solution. Mix with magnetic stir plate until the vanillin solution is suspended evenly.

Uniform Odor-changing F ilms

1. Measure out 15 mL of Calcium Sulfate Cross linking Solution into a falcon tube.
2. Place 100 mL of the Alginate-Vanillin Gel Solution in a bottle on a magnetic stir plate.
3. As it is mixing, pour the 15 mL of Calcium Sulfate Cross linking Solution into the bottle. As soon as all of it is poured, close the bottle and mix vigorously by shaking it for about 5 seconds.
4. Pour the solution into a large petri dish immediately.
5. Use a metal utensil to ensure clear clumps are evenly distributed throughout the plate.
6. Place the poured plate under fans with uniform air output, at approximately 4.5" high. Allow to dry.
7. When dried, cut the films to desired dimensions - voiding the edges.

4.3 Shape Changing Primitive

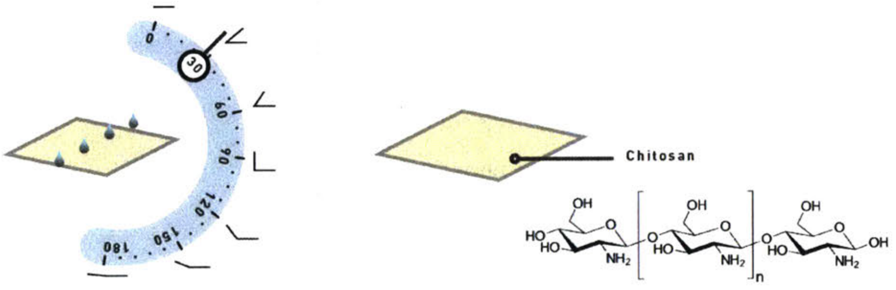

Figure 4-7. Output behavior for shape primitives and its composition.

The shape-changing samples were prepared using 4% w/v chitosan powder (Spectrum) dissolved in 3% v/v acetic acid (Sigma Aldrich). This sample composition was determined from preliminary studies of pH-reactivity across different chitosan percentages. Lower concentrations of chitosan yielded thinner, fast-acting films, while higher resulted in thick, slow-reacting films. The optimal composition was determined to be 4% chitosan, which created films that swelled within a reasonable timescale but were sturdy enough for structural applications.

4.3.1 Shape Output Characterization

Chitosan samples were cut into 20 x 7.5 mm strips, and tested using pH solutions of 2, 4, 6, 8, and 10. To create a hinge, five 0.75 uL droplets of the sample pH solution were applied across the sample's 7.5 mm centerline. The sample then bent along this line over a period of approximately 10 minutes. Samples were filmed and a still image for each sample was taken from the footage 7 minutes after the droplets were first applied. This still image was analyzed using the software ImageJ, which quantified the shape change as the bend angle along the 20 mm side of the sample. Results are the average of two identical trials.

Range of Angle Change

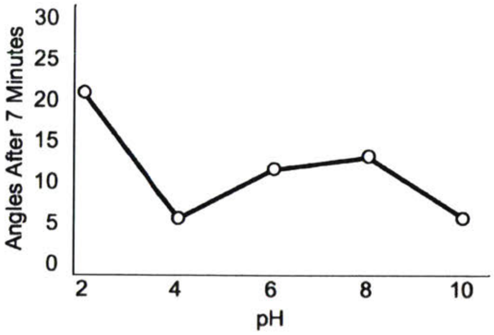

Figure 4-8. Angle change 7 minutes after droplets are applied to a 4% w/v chitosan film. The bimodal trend matches literature results [89].

The results as shown in figure rangeshape reveal a bimodal relationship

between bend angle and pH within the range of pH 2-10. Mahdavinia et al. found a similar relationship in their study, with swelling maxima at pH 3 and 8. Decreased swelling at middle pHs may be attributed to cross-linking [89].

Rate of Angle Change

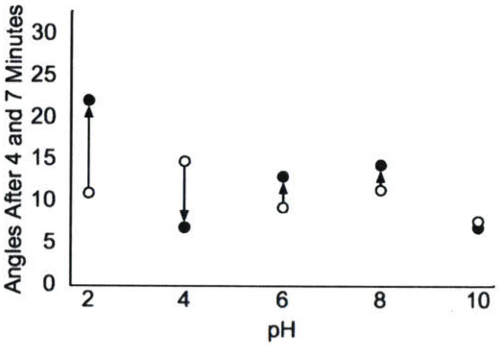

Figure 4-9. Changes in bend angle for 4% w/v chitosan films between 4 and 7 minutes after pH solution is added. Upward arrows indicate an increase in bend angle.

Regarding time evolution, the two pHs corresponding to angle minima, 4 and 10, decreased in angle between 4 and 7 minute measurements (figure 4-9). The other pH solutions had an increase in angle. It may be that interaction with any solution causes the chitosan to swell approximately the same amount, and then after some time the swelling becomes pH-dependent.

Reversibility of Angle Change

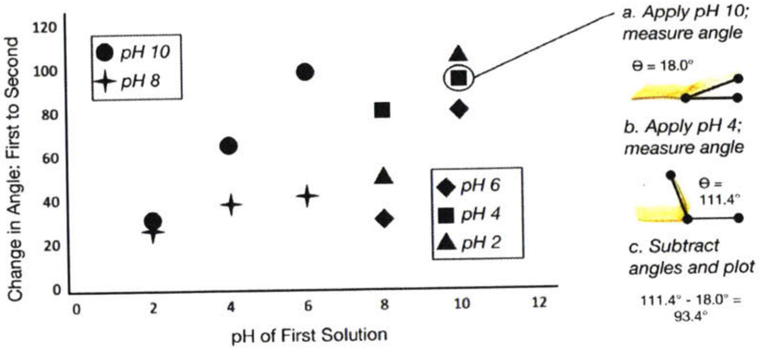

Figure 4-10. Reversibility graph of layering different solutions.

For this study, five 0.75 uL drops of the first pH solution were applied across the center of the sample. The film was given an hour to reach its final angle, measured with ImageJ (a). After the hour, droplets were applied again with a second pH solution on top of the first solution(b). Figure 4-11 shows that the more the second solution differs in strength from the first, the greater the change in angle is going to be. This preliminary study is promising for shape reversibility, as the bend angle of the film can be predictably manipulated by applying high and low pH solutions on the same material. It is interesting to note that pH 2 did not yield as much angle change as the other samples, perhaps due to a buffering effect with the high-strength solution.

4.3.2 Material Synthesis

Protocol for synthesizing shape changing films

Making 3% v/v Acetic Acid Stock Solution

1. Pipet out 15 mL acetic acid
2. Add it to 485 mL de-ionized water

Making 4% w/v Chitosan Gel Solution

1. Weigh out 20 g chitosan powder into a glass bottle
2. Add 3% Acetic Acid Solution (15 mL acetic acid + 485 mL DI water) until 500 mL
3. Using a magnetic stir bar, mix until chitosan powder is fully dissolved to form a yellow-brown gel solution. This may take 2+ hours depending upon volume.

Uniform Shape Changing Films:

1. Pour Chitosan Gel Solution onto an aluminum plate or acrylic plate with sanded edges.
2. Place the poured plate under fans with uniform air output, at approximately 4.5" high. Allow to dry for 5+ hours depending upon area.
3. When dried, cut the films to desired dimensions - voiding the edges.

4.4 Multi-Modal Primitives

In order to produce multi-property outputs within a single material, combinations of the three Primitives (odor, shape and color) were systematically tested to evaluate how they interface with one another.

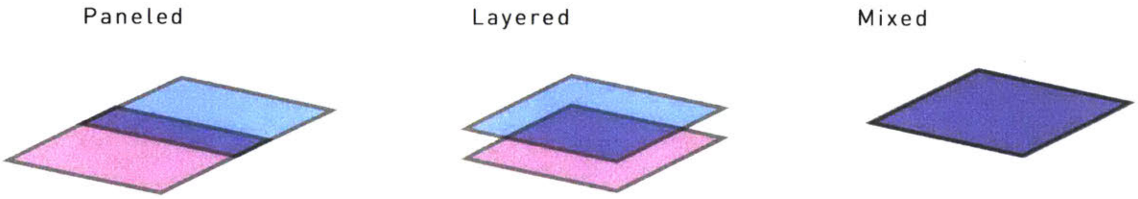

Figure 4-11. (Left to right) Paneled, layered and mixed methods for achieving multi-modal output.

I have classified three different ways the material primitives can be composited (figure 4-11): they can be mixed into a homogeneous solution before pouring, layered on top of each other, or synthesized side-by-side so that only the edges mix.

4.4.1 Multi-Primitive Output Characterization

The full combinatory range of composites made with the three methods were evaluated, to better understand the interactions between the three materials (figure 4-12). The combinations were tested with the same techniques used to characterize the original Primitives.

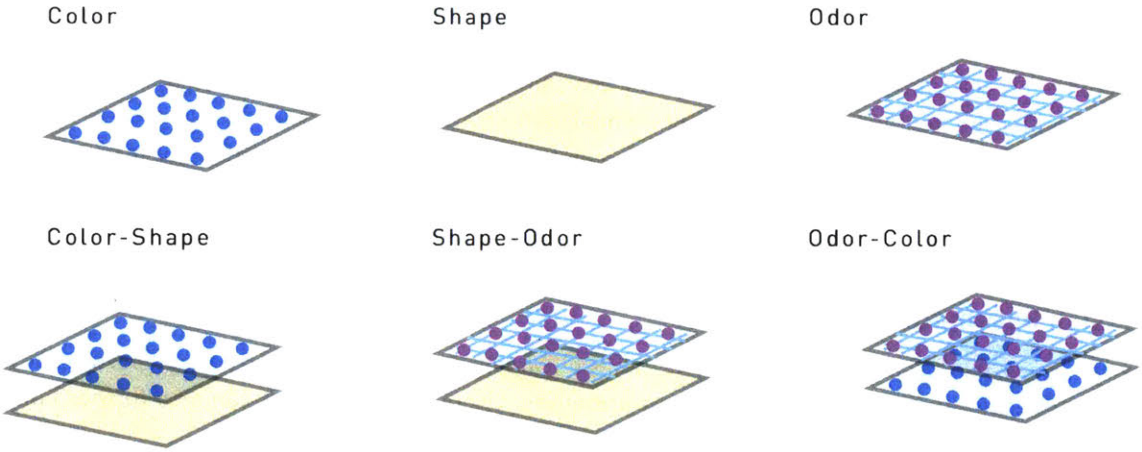

Figure 4-12. Permutations for multi-modal material primitives.

The resulting data showed that layered samples involving shape had one of two different pH-dependencies (figure 4-13). If the combination was scent-shape, the sample bent as predicted by the original shape trials. There was a difference in bend magnitude due to the different film thickness, but the relationship was cubic as expected. However, color-shape layered samples revealed an entirely different pH-dependence, being more quadratic in nature. This is due to buffering effects of the color primitive.

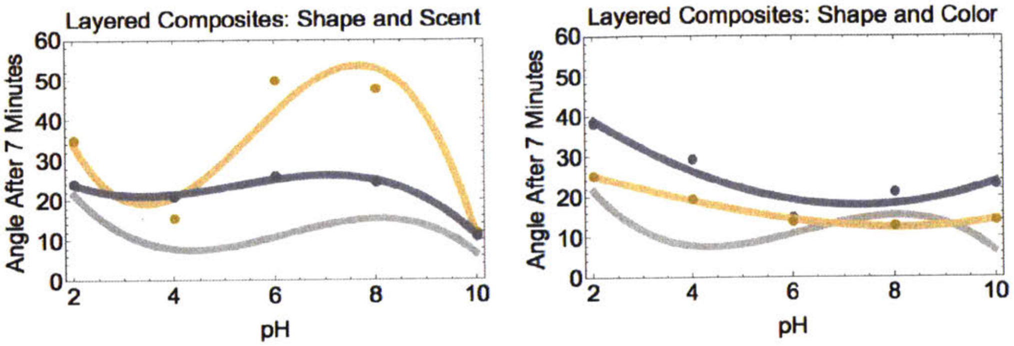

Figure 4-13. Results of shape-change tests on layered composites show that adding color significantly changes the shape dependence on pH. The yellow line is the shape on top sample, the purple line is shape on bottom, and gray is the control.

Generally speaking, the scent primitive rarely affected the material it was mixed with, color had a noticeable but non-destructive effect, and shape had a destructive effect except when paneled. This is due to the pHs of the Primitives themselves - shape is highly acidic, color is moderately basic, and scent is slightly basic. The greater the deviation from neutral, the greater the effect the component will have.

4.4.2 Material Synthesis

To synthesize multi-modal primitives, the *Organic Primitives* can be mixed, layered or paneled. The mixed approach involves mixing primitives in their liquid form to yield a homogenous solution and then poured to create a uniform film. Layered primitives can be achieved by pouring one primitive on top of another after the first had dried for 48 hours. Paneled method involves primitive solutions casted side-by-side where only the edges mix. A section is cut out of a sample dried that has dried for 48 hours and a different primitive is then poured in the void.

4.5 Design Parameters

These proposed design parameters allows users of *Organic Primitives* to leverage its properties for a variety of applications and development new primitives. Users of this platform can also further build upon this logic to expand upon these initial design parameters.

4.5.1 Color, Odor and Shape Primitives

Designers working with the shape primitive must consider its bimodal pH response. Activating the material with pH 2 will create the most pronounced shape change, while pHs 4 and 10 generate more subtle changes. If rate of change is a consideration, more basic pHs result in faster equilibration. The scent-changing material gives a maximum scent response when activated with pH 4, and the scent can be suppressed with pHs 9 or 10. Using pHs greater than 9 to activate the material creates a range of intensities, though perception of these intensities vary between different users. Of the three primitives, the color-changing material had the most linear pH response. pH 2 creates a vivid and instantaneous response. However, the response to very acidic or basic pHs cannot be reversed. If the desired application requires changing color repeatedly, pHs closer to neutral (ie. 6 and 8) should be used.

4.5.2 Multi-Modal Primitives

Figure 4-14 depicts the various combinations of primitive composites that was determined perform most optimally. A composite was considered successful if it was (i) reactive as intended, (ii) structurally sound, and (iii) reactive within a human-perceptible timescale. These materials exhibit different behaviors when patterned by way of layering, paneling, or mixing the *Organic Primitives* together in various combinations.

Figure 4-14. Composites with optimal performance (left to right): layering of color-on-odor; the paneling of shape-in-odor, odor-in-shape, odor-in color; and the mixing of color-odor.

Shape-and-Odor or Color-and-Odor combinations appear to be most compatible with one another, while Shape and Color were not. To make a single material with all three properties, several methods can be used: 1) Chitosan can be paneled with Anthocyanin-Vanillin Alginate Calcium Sulfate; 2) Chitosan can be paneled with Anthocyanin Alginate Calcium and Vanillin Alginate Calcium.

5. Droplets as Fluid Information

Fluid information from natural contexts as well as prepared solutions can be used to activate material outputs (figure 5-4). Contextual fluid inputs include rain drops, ocean water, body fluids, food sources, and organismic excretions from plants and microbes. Prepared inputs using prepared acid-base solutions can offer designers a wider output range while yielding more precise outputs. These can be integrated with mechanical and electronic systems to develop machine-mediated activation methods.

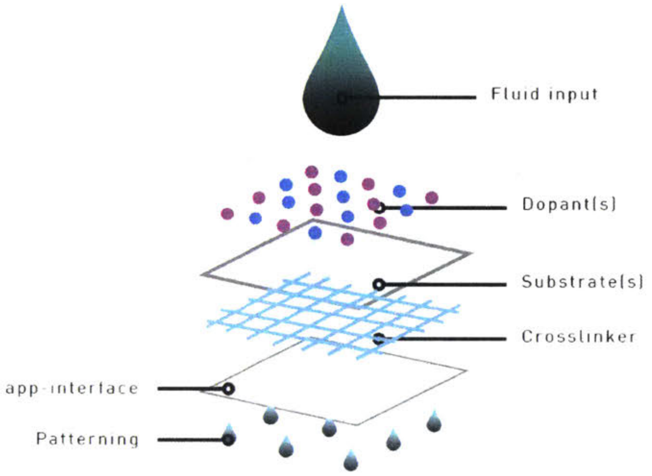

Figure 5-4. Exploded diagram of a material interface as an information display for droplet input.

As noted in the previous chapter, this thesis focuses on sensing pH in fluid as a model for the development of this design methodology, to demonstrate how this molecular scale phenomena can be brought into material, application, interaction and cultural implications, across scales.

5.1 Omnipresent pH Signals

Chemically, pH is the measure of the acidity and alkalinity with the relative amount of free hydrogen and hydroxyl ions in a fluid. It is an important health indicator for many complex chemical and biological processes in, out, and on the human body, from food to personal care. pH also serves as a critical environmental indicator for processes such as ocean acidification and atmospheric contamination.

The flow of molecules moderate the pH values in a variety of natural processes, from microorganisms to the atmosphere. Chemical exchanges represent the communication pathways between micro- and macro- scale ecosystems. On a micro scale, pH values fluctuate as different microbes undergo metabolic processes to generate byproducts that lend to the synthesis of various acids and bases, among other molecules. For example, lactic acid is a common byproduct of *Lactobacillales* from carbohydrate fermentation, commonly found in decomposing plants and milk products [90].

Within the human body, the pH of blood maintains an equilibrium of 7.35 - 7.45 [91] [92]. The pH for human tears differs based on individual, time of day and physiology [93] [94] [95]. Generally, researchers have found that a normal range is between 6.5 - 7.6 [96]. The pH of sweat varies between 4 - 7 [97] [98] [99]. The pH of urine is 5.5 - 7 [100]. The pH of saliva varies between 6.2 - 7.6 [101] [102]. Deviations from the norm can signify health issues and disease.

On a macro scale, the pH of ocean is about 8.2, but has been gradually decreasing as a result of ocean acidification induced by ocean absorption of CO2 emissions [103] [104] [105]. The pH of the rain ranges from place to place [106]. On average at 5.6 in the United States, it can be acidified to less than pH 4.3 due to emissions of sulfur dioxide and nitrogen oxide [106] [107] [108].

Many edible materials and personal care products can be used as pH inputs for Organic Primitives, as the range of many off-the shelf products spans pH 2 - 12 [109]. The pH of food varies across a range of pH 2 - 8 [86]. For example, lemon juice is at a pH 2, soda is at a pH of 2.5. Many fermented foods, such as pickles and cheese, range between pH 3 - 6. Milk and vegetables are more alkaline and range from 6 - 8. Oral supplements can range from pH 2 - 10, while personal care products such as soaps range between pH 8 - 12. For example, a vitamin C oral supplement is pH 2.4, while milk of magnesia is pH 10.5 [110] [111].

5.2 Context-specific Fluid Inputs

The input begins with determining what fluid-based information should be sensed in the desired system. The contents and characteristics of the fluid differs depending upon whether the target system for interaction is environmental, edible, bodily, domestic, social, cultural or interspecies.

Interacting with the environment may entail considering the composition of rainwater, ocean water, pool water, snow, pipe exhaust, faucet water or bath water.

Food may utilize edible compounds from beverages, condiments, additives, and other engineered or whole products. Beverages such as soda, milk, fruit juice, tea, alcohol, cocktails, soups or coffee can be used. Condiments and additives like mixers, jelly, sauces, dressings and other emulsions can be considered. Ingredients like egg whites, lemon juice, and olive oil can be extracted from whole foods into liquid state. Engineered state changing foods like ice-cream and popsicles add another dimension of possibility to interaction.

The human body may involve tears, blood, interstitial fluid, urine, sweat, saliva, or mucus. Some fluids are not innate to the body's chemistry but are culturally adopted and utilized such as skin care regimens such as face wash, cosmetics, soaps, perfumes, toothpaste, mouthwash, hairspray, shampoo, conditioners, moisturizers and masks. Medicinal fluids that are topical or ingestible such as tums, milk of magnesia, creams, epson baths, capsules, and vitamins can also be used.

Fluids that do not pertain to a natural system such as the environment, human body, or food can be prevalent in a domestic, manmade environment like the home, office, or spiritual space. These include household cleaning products like laundry detergents, window cleaning fluid, paint supplies, dish soap, air freshener, and disinfectants.

Ceremonial fluids from social rituals and spiritual practices can also be considered for use. For example, the blessing of holy water is as a form of spiritual cleansing or protection against evil [112] [113] [114]. Fluids such as oil and wine are central components in ritual and worship, as they carry symbolic and spiritual functions [115] [116].

Interspecies interaction can derive from synthetic chemical products like insecticides, pesticides, soil nutrients and minerals. Molecules produced by organisms that exist symbiotically with one another such as pheromones, animal secretions, and metabolic byproducts [117] [118] [119] [120].

5.3 Prepared Fluid Inputs

In a scenario which a designer or engineer is interested in utilizing an arbitrary fluid as an input mechanism, powdered acids and bases or predefined pH solutions can used. Researchers can make their own prepared solutions that are context agnostic and discretized to be embedded within an application experience with no visible fluid interaction with the user.

pH	g citric acid / 100 mL	g sodium citrate / 100 mL	g sodium bicarbonate / 100 mL	g sodium hydroxide / 100 mL
2	9.606	0	0	0
3	1.575	0.286	0	0
4	0.788	1.523	0	0
5	0.75	2	0	0
6	0.221	2.194	0	0
7	0	0	0	0
8	0	1.913	0	0
9	0	9	0	0.012
10	0	0	0.21	0.048

Table 5-1. Prepared solution recipes for pH 2-10. These solutions were used to characterize all of the material primitives within this thesis.

pH solutions can be selected from everyday food and personal care products or prepared through pure formulations. For skin-safe or edible applications, the Federal Food and Drug Administration (FDA) maintains a Generally Recognized as Safe (GRAS) listing where I have based the concentrations of all the prepared solutions for testing and use [86]. As shown in table 5-1, pH solutions I utilized within these systems are all made using food-grade materials: citric acid (derived from citrus fruits), sodium citrate (food additive), sodium bicarbonate (baking soda) and sodium hydroxide / lye (commonly used for food preparation). pH solutions across the safe to handle pH 2 - 10 range can be made using a combination of different acids and bases. Selection of chemistry for prepared solutions can enable researchers and designers for an added level of control in the material's reaction time and buffering capabilities when activating an *Organic Primitive*. Other common acids and bases include lactic acid (byproduct of bacterial food fermentation), ascorbic acid (vitamin C), and sodium tripolyphosphate (used as food emulsifier), among others.

5.4 Coupling and Decoupling Input-Output

Researchers should specify whether they would like coupled input-outputs such as use of natural input of saliva at pH 6, or decoupled input-outputs where prepared pH solutions generate output designs where input value can be arbitrary.

Coupled I/O

The material input of pH is coupled with the output of its color, odor, and shape characteristics. This makes it possible to utilize natural pH fluid inputs from the environment such as rain water. However, this approach limits the possible output states that a material will perform. For a particular color, shape and odor state to be actuated, only particular input values can be used, as these are coupled to specific information states. Utilizing natural inputs at pH 6 to output property ranges that do not correspond to their specific value is possible, but requires researchers to develop new composite structures with the primitives to support this.

Decoupled I/O

Prepared fluid inputs that are computationally activated offer maximum precision in manipulating the output dynamics of *Organic Primitive* information displays. Presented in the prior chapter, prepared inputs involves utilizing predetermined pH solution recipes to activate the display outputs. Rates of change, range of change, buffered effects and reversibility parameters can be manipulated by tuning and selecting specific acids and bases. Further details on computational, machine-mediated methods for activation will be discussed in the following section.

5.5 Activation and Control Schemes

All input methods are spatially defined, whether they are natural inputs such as the rain or prepared and machine-mediated inputs with fluidic logic. These will generally fall into one of three categories of control - global, local or discrete. Temporal dimensions of material behavior can be defined through sequential ordering of specific pH solutions deposited spatially.

5.5.1 Global, Local, Discrete Control

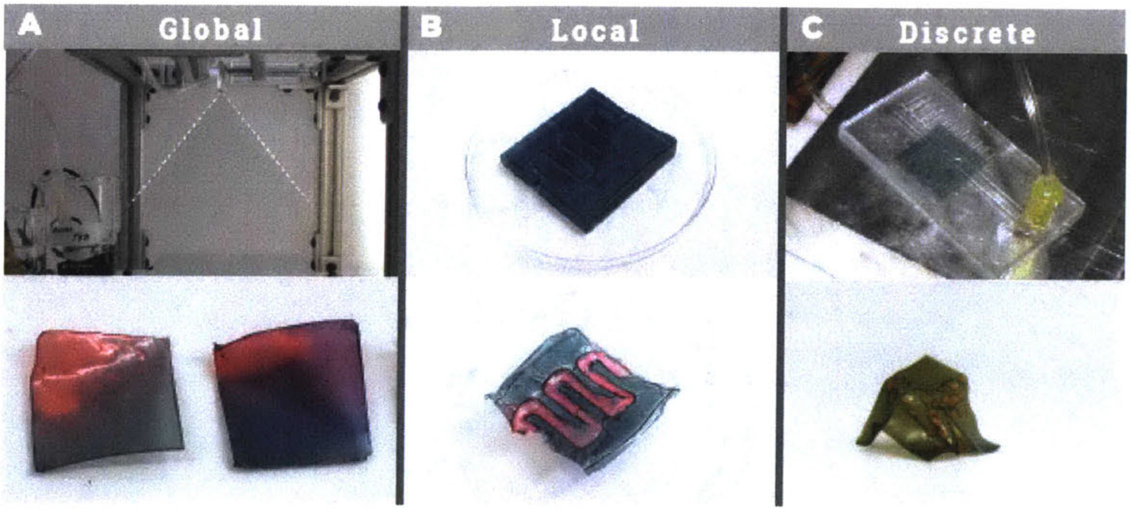

Figure 5-2. Techniques for activation and control of the material primitives: a) Global activation involves pH solution sprayed onto the entirety of a material in an instance b) Local activation actuates a stream of solution in a channel c) Discrete activation triggers a single droplet in a precise position to actuate

Material activation generally fall into one of three categories of control—global, local or discrete, as shown in image a, b, and c of figure 5-2. Different deposition techniques can yield specific material output behavior. Global control utilizing atomizers and nozzles can activate a large to medium size area of the material. Local control can be developed by fabricating pipes and channels into the material structure through molding or compositing with other materials. Discrete control can be implemented by designing specified outlets in fluidic circuits or patterning on defined regions where pH solutions make contact with an *Organic Primitive*. Computational control can employ any of these three control schemes to enable temporal control of a primitive's output by sequentially defining specific pH values deposited, elaborated in the following section.

5.5.2 Computational Deposition

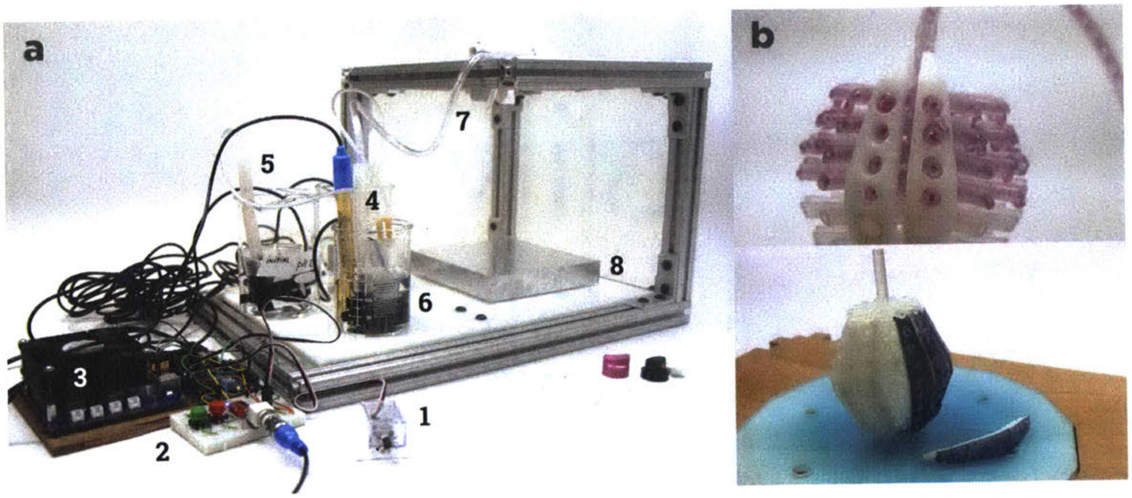

Figure 5-3. (a) Hydraulic system for global and sequential activation (b) Bionic fruit device deposits material primitives for use as edible films.

While it is not required in order to achieve an input-output response, electromechanical systems can be integrated for activation (figure 5-3a) or deposition (figure 5-3b). The apparatus in (a) is a platform which computationally generates a desired pH solution for global activation of an *Organic Primitive* , as discussed in the previous section on figure 5-2a. It can be used to control the outputs in a particular sequence, to enable the material to animate and perform a series of behaviors. It possesses 2 reservoirs of solution - pH 2 and pH 10, and outputs any desired pH these values. Using a series of hydraulic pumps and a pH sensor to complete the closed loop control of the system, I can continually adjust the pH value as I measure with a digital pH meter. A microcontroller with a potentiometer enables a user to select the desired pH by rotating the knob(a-4), initiating the pumps to draw fluids from pH 2 and pH 10 reservoirs(a-5) into a microfluidic mixer(a-4). Once the desired pH value is reached, the user can deposit the pH solution through the nozzle (a-7) to activate a material on the platform (a-8). The nozzles I used were adopted from off-the-shelf spray paint tops, offering a low-cost and versatile device. For deposition, (b) shows a bionic fruit which deposits primitives on the surface of a 3D printed fruit for user interaction. Further elaboration of the bionic fruit is in the application section.

Local control (figure 5-2b) is created by casting hollow channels with the material primitives. A number of fluidic logic can be implemented to transcend the capabilities of the primitives from mere sensor-actuators. Discrete activation (figure 5-2c) can be achieved by adapting the apparatus in figure 5-3a to connect to a microfluidic device instead of the nozzle. The microfluidic device was fabricated from acrylic with a laser cutter to achieve 600 micron channels. It contains outlets which deposit pH solution to the material in small droplets. By depositing discrete droplets of pH solution onto a shape-color changing material, it can output a complex structure, as shown figure 5-2c.

6. Primitive Development

I will explain the approach used to create the current material primitives and discuss the process for developing an *Organic Primitive*.

6.1 Experimental Process

The process for developing primitives is outlined below:

1. Determine what inputs are to be sensed or what outputs are desirable. Identify molecular mechanisms and reactions that can serve as dopants. One option is to select specific molecules that can serve as integrated sensor-actuator mechanisms. A catalog of dopants is available in this chapter as a resource. The other option is to design your own molecular receptor-spacer-reporter mechanism. More advanced approaches with synthetic biology, synthetic and semisynthetic chemistry, and reconfigurable molecules can also be employed.
2. Once molecules and mechanisms are identified, responses should be tested in liquid phase. Proper solvents for the molecules should be identified. Characterization of how varying concentrations of the molecule and chemical input affect different output parameters (range, rate, reversibility) is carried out. This data aids in understanding how the molecular mechanisms change when they are combined with a range of substrates.
3. When there is a promising input-output reaction in liquid form with human-readable changes, the molecular sensors can then be turned into solid-state materials. Solid-state materials can be generated in a number of ways, but the central task is to screen a number of substrates to find the most compatible one(s). Compatibility factors include buffering capacities, diffusion rates, degradation effects, and native pH values. In depth discussion on compatibility will be presented later in this chapter.
4. Based on the screening results, optimal dopant-substrate combinations should emerge from the different combinations. These will serve as the preliminary *Organic Primitives* and primitive subsets. Methods should be developed for creating uniform and consistent materials.
5. A protocol outlining specific instructions and concentrations should be written up when methods have been established.
6. The new primitive(s) should then be characterized based on the chemical stimuli, which is pH in this model example - to identify its output parameters (range, rate, reversibility). This characterization method should be identical to the study done in its liquid phase. Concentrations and chemical input should be correspondent to the liquid phase tests.
7. The characterization data will form the 'data sheet' and the protocol will be the 'recipe' with which will serve as resources to designers.
8. To ensure it's compatibility with the rest of the *Organic Primitives* collection, a combinability characterization study should be conducted. In this examination, the new primitive will be composited with other *Organic Primitives* across three methods (mixing,

layering, panelling), to determine how they interact. Output parameters (range, rate, reversibility) and structural stability, will be measure for its success.

9. To advance the material primitive into application development capabilities, interfacial materials that this primitive is compatible to should be identified. Following the "like-dissolves-like" principle, these are typically similar classes of materials as the substrate of the primitive.

6.2 Catalog of Material Building Blocks

The following molecules and materials are encountered daily in food, but their stimuli-responsive properties are generally overlooked. Such renewable sensor-actuators are inexpensive to produce, safe to ingest, and offer complex multi-property output capabilities.

6.2.1 Molecular I/O Dopants

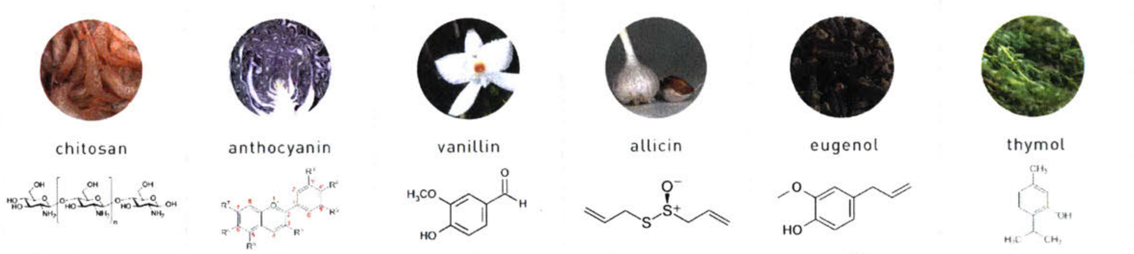

Figure 6-1. Collection of pH-reactive molecules from food, with the first three tested and developed into material primitives in this thesis.

These dopants serve as molecular sensing-actuation building blocks for the development of *Organic Primitives*. In this section, I discuss the chemistry of the pH-reactive compounds explored in this thesis, as shown figure 6-1. These molecules were selected for their safety, accessibility, and biocompatibility, to ensure the possibility for researchers to develop their own pH-responsive material interfaces without special tools or facilities. Future work can explore the potential for other dopants and substrates for creating *Organic Primitives*.

Anthocyanin: Color-Changing Reconfigurable Molecule

All tissues of vascular plants contain the flavonoid anthocyanin, a pigment that changes color under varying pH solutions. In the 1664 book *Experiments and Considerations Touching Colours* by chemist Robert Boyle, various edible plants have been reported for use as visual indicators due to pH-responsive mechanisms in their tissues [121]. Anthocyanin is commonly used as a food colorant in the food and beverage industry, and has been found to possess anti-inflammatory antioxidant properties [122]. It has also been researched for use as an indicator for packaging applications to detect spoilage in pork and fish products [123].

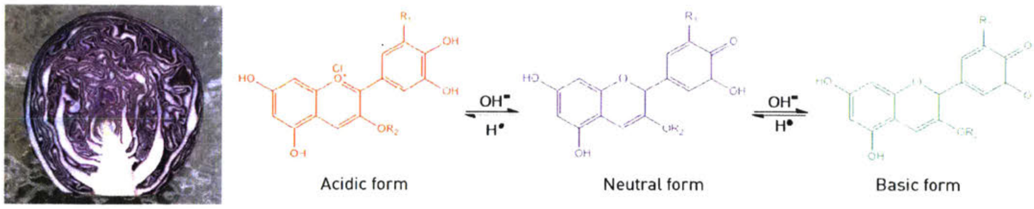

Figure 6-2. Chemical diagram of anthocyanin pH reaction.

Anthocyanins and anthocyanidins (the sugarless form) are a class of molecules that includes cyanidin, delphinidin, pelargonidin, peonidin, and malvidin. Under different pH conditions, the hydroxyl (OH) and/ or methyl ether (O-CH3) groups attached to the carbon rings undergo reversible structural transformations and ionizations (figure 6-2). This restructuring affects which wavelengths of light are absorbed, giving rise to color changes [124] [125].

Vanillin: Odor-Switching Flavor Molecule

Vanillin is a flavor molecule found in vanilla beans, contributing to their characteristic aroma. The smell of vanillin has been found to elicit feelings of relaxation [126], offering potential uses for interface design. The effect of pH on vanillin stability was first characterized by food scientists in the early 1970s [127]. In 1990, Flair and Setzer determined vanillin could be used as an olfactory titration indicator for blind students [128]. Vanillin is prevalent as a flavoring for food products such as chocolate and ice-cream [129].

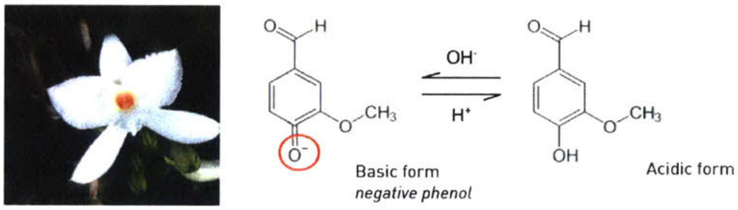

Figure 6-3. Chemical diagram of vanillin pH reaction.

The smell of vanillin is elicited at pH 2-8 and suppressed at pH 9 and 10. Vanillin in ethanol for olfactory titration had been previously investigated, but possess conflicting results in different experiments. Flair and Setzer claim it is unsuitable due to relative lack of odor at the equivalence point [128]. Neppel et al uses vanillin for end-point determinations in olfactory titrations [130]. There is little literature on the mechanisms for odor release of vanillin under different pHs. One possible reason for odor elimination at high pHs is that the phenol group of vanillin possesses a pKa value of 7.38, causing it to become negatively charged, and therefore less volatile [131], under basic conditions as shown in figure 6-3.

Chitosan: Shape-Memory and Swelling Macromolecule

Chitosan is a polysaccharide derived from the exoskeletons of shrimp and crustaceans, widely studied due to its abundant and biocompatible nature [89] [132] [133]. The pH-responsive swelling of chitosan hydrogels was first characterized in publications from the 1980s [134]. It has been researched for drug delivery applications, such as to target tumor tissues through the release of drugs at specific pH [135]. The tunability and shape memory characteristics of chitosan have been studied in tissue engineering to create scaffolds with impregnated bioactive agents for guiding cellular growth towards generation of new tissue [136].

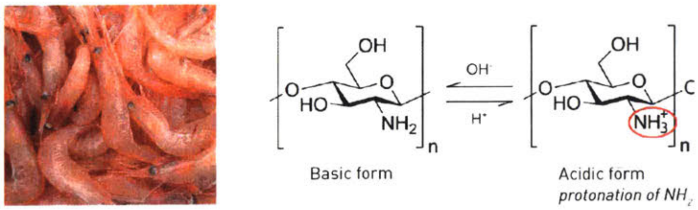

Figure 6-4. Chemical diagram of chitosan pH reaction.

Illustrated in figure 6-4, its NH2 amino functional group is protonated in low pH conditions, causing the polymer to swell due to charge repulsion; if the pH becomes high, chitosan returns to its collapsed state.

6.2.2 Biomaterial Substrates

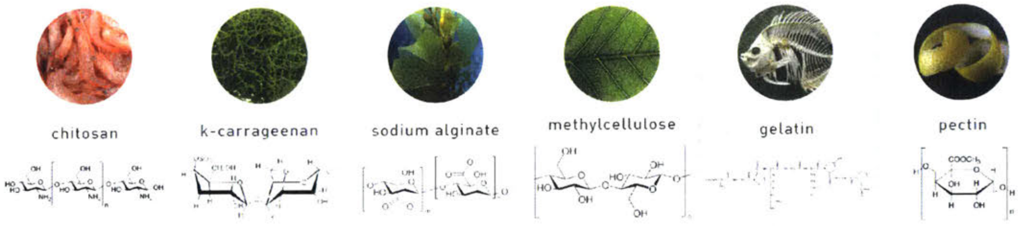

Figure 6-5. Edible biopolymers and materials that can be used as substrates.

In this section I discuss a variety of edible substrates shown in figure 6-5 that can be functionalized by incorporating molecular I/O dopants. Different polymers and cross-linkers can be utilized as matrices for the pH-responsive dopants depending upon the intended application and specific characteristics desired.

Sodium Alginate

Sodium alginates are anionic polysaccharide gums derived from the cell walls of brown seaweeds of the phylum *Phaeophyceae*, which are harvested from seawater. The *Phaeophyceae* or brown algae are also known as kelp, range in size and can span up to 20 meters long. Alginate can be ionically cross linked through the introduction of calcium ions to form a semi-rigid gel [78] [79]. The rate of gellation can be tuned based on specific cross linker. Color-odor changing primitives were made by integrating anthocyanin and vanillin dopants with sodium alginate biopolymer. Depending on the form factor, I used a different cross linker to speed up the reaction. Calcium sulfate cross linked with alginate was used for making films to induce a slower gelling effect, while calcium chloride cross linked with alginate was used to rapidly extrude fibers.

Kappa-Carageenan

Carrageenan are linear sulphated polysaccharides from red seaweeds. There are three variants of carrageenan with varying sulphate groups per disaccharide, with kappa possessing one, iota with two, and lambda with three. Among the three, I used kappa-carrageenan for creating the color primitive as it produced the most optimal films when dried. Other carrageenans can be investigated for use in other form factors [137].

Gelatin

Derived from collagen obtained from different animal body parts such as skin, bones and tissue, it consists of peptides and proteins. It easily dissolves in hot water and gels when cooled. When dried, it becomes a brittle, translucent material. Different blooms or strengths of gelatin are produced [138]. Early experiments of doping gelatin with anthocyanin confirmed it has a slow diffusion rate due its brittleness, making it an ineffective substrate for creating Organic Primitives. However, kappa-carrageenan is very thin when dried, so mixing controlled ratios of gelatin with doped kappa-carrageenan can generate 'primitive subsets' to provide structure and strength, or be used to control an object's rate of change.

Pectin

Found within the cell walls of plants, pectin is a complex set of polysaccharides that are commonly extracted from citrus peels of grapefruits, oranges, and lemons. Similar to gelatin, it forms a brittle, clear material when dried [139] [140]. It can also be used as a primitive subset to provide structure and strength.

6.3 Dopant-Substrate Compatibility Factors

In order to utilize the pH-reactive properties of the molecules, the chemical phenomena must be brought into human-scale interactions by materializing their responsive properties from liquid into a solid state. In my investigation, I have found sodium alginate and kappa-carrageenan to be good biopolymer substrates for the pH-responsive molecules due to their semi-permeable structures, consistent behavior, rapid diffusion rates, and relatively neutral pH. Different polymers and cross-linkers can be utilized to tune a variety of matrices for the pH-responsive dopants, depending upon the intended application and specific characteristics desired. Further investigation is needed to fine-tune adherence, crosslinking, and buffering capacities of different molecule-material combinations.

- Substrate & responsive molecule adherence is dictated by pore size and complexation. I choose a substrate with the least interference.

- Degradability of the substrate across different pH stimuli effects compatibility & reversibility.
- Rate of change and reversibility is dictated by the substrates that are doped.

6.4 Engineering a Library of *Organic Primitives*

This section presents notes and lessons drawn from experiments.

Identifying Dopants

Molecules which are unstable yield the most prominent responses. Stable chemicals are less reactive and prone to state changes. Most chemical products especially with food are engineered to be stable so that their state remains unaltered given a range of environmental conditions. In the case of creating new *Organic Primitives*, identifying edible molecules which are unstable provides an insight towards potential candidates as dopants.

Rate of Change

- Buffering capacity of both the fluid input and the primitives effects the rate of change.
- The composition of the input fluid will likely change the output behaviors in their rate and in some cases the range, from buffering effects.
- Rate of change plays a role in the diffusion rate of the polymer, hydrophobicity of its surface chemistry and the ionic mobility of the fluid in the polymer.
- Size of the molecule effects the diffusion rate and capacity for it to remain within the polymer. Relative pore size of polymer, dopant and pH solution contributes to the rate of change.

Range of Change

- Odor dopants are small molecules with low molecular weight as high vapor pressure allows it to go from liquid to vapor phase.
- Range of change is effected by buffering capacity, functional groups, and titration curves of different substances interacting with one another. The dopant's range of change is based on the reconfigured states of the molecules based on the stimuli. The ability for multiple range of change is due to functional groups interactions with the pH environment.
- The more unstable the molecule is with the fluid input, the easier it is to initiate an output or change.

Reversibility of Change

- When selecting a substrate, ensure it will not degrade from the fluid input.
- Reversibility is also likely due to degradation of molecule with fluid input.
- The dopant's stability at different pH and stimuli also plays a role in reversibility.

- Examining substrate interaction with pH and stimuli is crucial for generating maximally reversible reactions, whereas the dopant in fluid form alone can be reversible for prolonged periods of time.

7. Design Space

7.1 Primitive Subsets

The color, odor and shape changing primitives can be formulated to generate a number of primitive subsets that function to alter output combinations, rates of change, rigidity, and starting states. This section discusses the many different knobs that have been investigated in this work, by techniques such as creating co-polymers, composites, cross linkers and alternative biomaterial substrates.

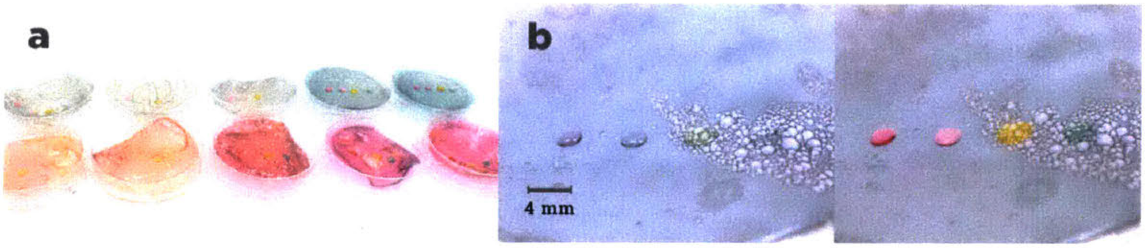

Figure 7-1. (a) Serial dilution of anthocyanin kappa carageenan with structural biopolymers gelatin (top row) and pectin (bottom row). (b) Before and after of pH solutions on anthocyanin doped kappa-gelatin.

For additional structural integrity, other biopolymer substrates can be incorporated with the pH-reactive *Organic Primitives*. I experimented with various ratios of structural polymers and have identified a mixture of kappa carageenan with gelatin or pectin to be most optimal. Pictured in figure 7-1 in image (a) from top left to right, are primitives subset copolymers with ratios of 250 bloom fish gelatin with color primitives at 3:1, 2:1, 1:1, 1:2, 1:3 and rapid set high methoxyl pectin in ratios of 3:1, 2:1, 1:1, 1:2, 1:3. Preliminary pH solution tests as shown in (b) indicates that this approach does not hinder range of change of the color primitive.

7.2 Form Factors

In order to utilize the pH-reactive properties of the molecules, the chemical phenomena must be brought into human-scale interactions. Here, I show several example methods for materializing the molecules.

Functional molecules can be synthesized into different states of matter, from vapor to liquid to solid, yielding primitives that include fibers, films, sheets, liquids, gels, and solids, as shown in figure 7-2. pH-reactive color-odor fiber was fabricated using a syringe tube to extrude a formulation of anthocyanin and vanillin within sodium alginate into a calcium chloride solution. The resulting polymer fiber was ionically cross-linked. Thin color-shape changing films and color-odor-shape changing sheets were synthesized by casting molecule-biopolymer solutions atop a sheet of sanded aluminum under a fan. Molecules can also be suspended and used in liquid form by diluting them in appropriate solvents. Vanillin was suspended in ethanol and anthocyanin in water. Other color and/or odor-changing gels are made by suspending the primitive molecules in polysaccharides such as alginates, agar, pectin, gelatin, and carrageenan. Solid 3D and 2.5D dimensional forms can be casted and molded.

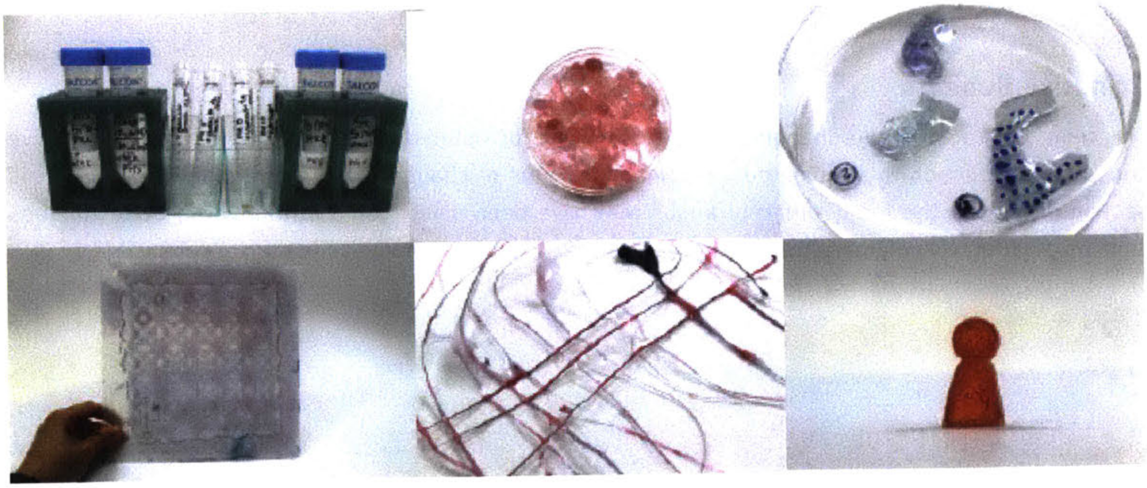

Figure 7-2. (left to right clockwise) liquid based *Organic Primitives*; odor-color changing gel; color-shape changing films; color changing 3D form; odor-color changing fiber; odor-color-shape changing sheets.

Fibers

Figure 7-2 shows a series of *Organic Primitives* with a variety of property and input-output combinations are highlighted. In the (top left) image, a pH-reactive color-odor fiber was fabricated using a syringe tube by extruding a formulation of anthocyanin and vanillin within sodium alginate into a receptacle of calcium chloride to create an ionically cross-linked polymer fiber. The fibers were then dried overnight.

Films

In (top right) image, a pH-reactive color-shape changing film was made by first inoculating anthocyanin with chitosan dissolved in acetic acid, followed by casting the solution atop a sheet of aluminum and finally patterning with a solution of anthocyanin mixed with 0.1mM of pH 10 sodium hydroxide solution (NaOH).

Sheets

In the (middle left) image, a pH-reactive color-odor-shape changing film was made using chitosan dissolved in acetic acid. The material was then pattern using an anthocyanin - pH 10 mixture along with vanillin stock solution.

Liquids

In the (middle right) image, the film switches from strong odor to no odor in liquid form within different pH conditions. The flavor molecule vanillin was dissolved in ethanol to create a stock solution. A portion of the vanillin solution was mixed into pH 2, 4, 6, 8, and 10 solutions.

Gels

In the (bottom left) image, pH-reactive color-changing gels were made using sodium alginate doped with anthocyanin and cross-linked with calcium chloride.

Solids

In the (bottom right) image of figure 7-2, a pH-reactive color-changing form was molded using kappa-carrageenan form doped with anthocyanin. 3D and 2.5D form can also be encoded into flat films where post-activation a dimensional form may emerge.

7.3 Fluidic Logic

Integrating fluidics with Organic Primitives can enable information processing and activation through passive flow or digital control [141] [33] [44]. In this section I highlight different approaches for activation, as shown in the figure 7-3, where different geometry is used to activate and create time-telling and titration devices.

Figure 7-3. Different methods to integrate fluidic logic with *Organic Primitives:* (left to right) closed composite, open composite and molded material logic.

Ticker for Time-Telling

Through diffusion rates of *Organic Primitives* with pH solutions, I developed a material-based "clock" which changes color to mark specific time increments (figure 7-4). The time duration can be tuned by changing the shape, length, or diameter of the channels.

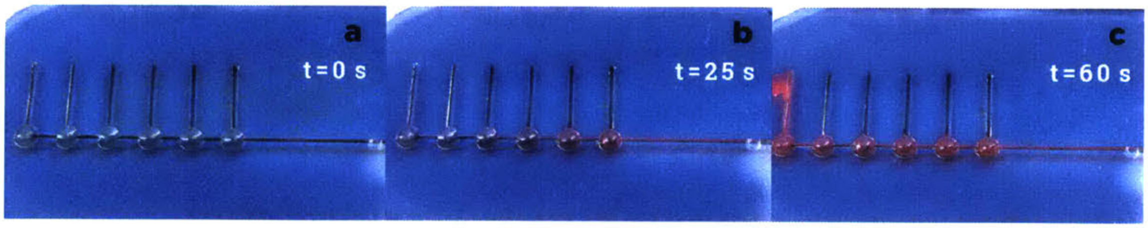

Figure 7-4. Passive diffusion based time-telling device color-odor material composite.

Gradiator for Titrations

Gradiators can be used to titrate solutions by inputting two values, such as pH 2 and pH 10, to create a material with equal values between the endpoints (figure 7-5). This can be used to create arrays of flavor, intensities of color, and/or odor strengths.

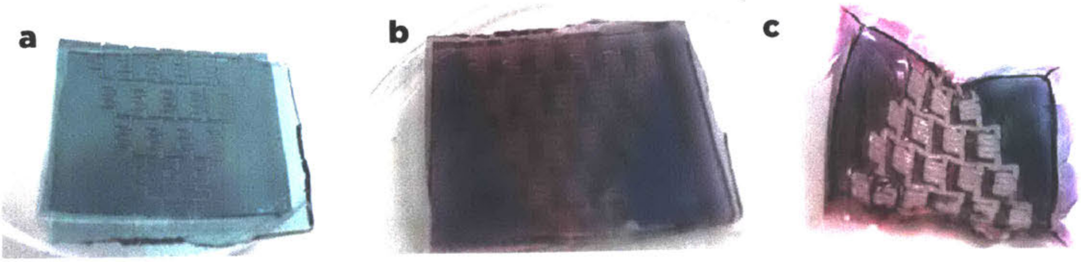

Figure 7-5. Fluidic channels molded then flowed through with pH 2 and pH 10 as inputs to generate a titrated gradient.

7.4 Temporal Sequencing

Sequential and temporal depositions of pH solutions can yield capabilities for physically animating a material's output for color, shape, odor or combinatory changes as demonstrated in figure 7-6.

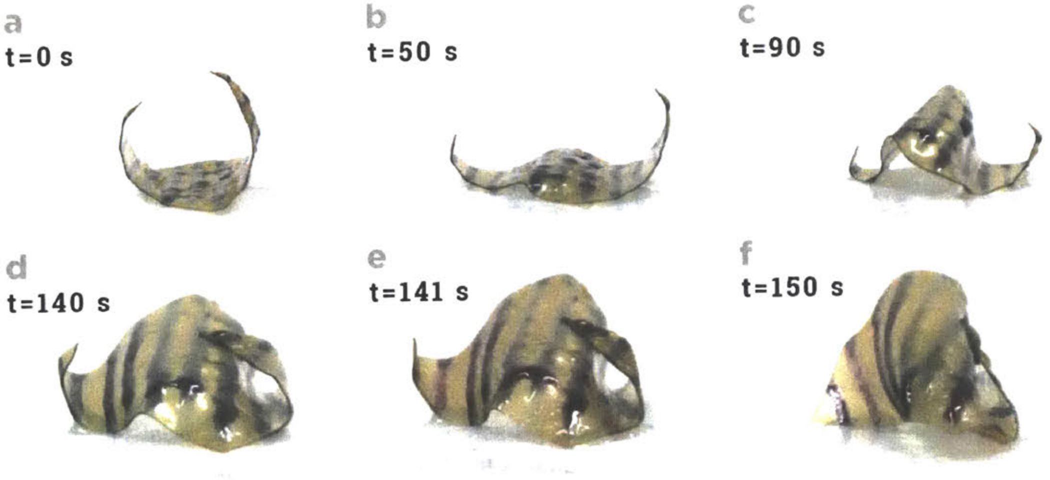

Figure 7-6. Patterned shape changing primitive activated through temporal sequencing of different pH solutions. Top row was globally activated at pH 8 and bottom row at pH 2.

Beyond sequential and temporal methods, form factor can also enable specific animation capabilities through relative diffusion rates of non uniform thickness distributions as exhibited in figure 7-7.

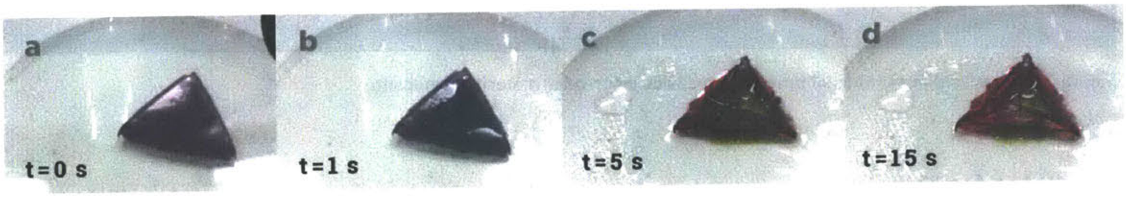

Figure 7-7. Response of color-odor material molded into triangular 2.5D form to pH 2 solution.

7.5 Patterning Techniques

This section describes how multi-property outputs can be achieved through composites and patterning techniques through several examples I have implemented. This technique can be used to encode output behavior of the material primitives.

Patterning with Multiple Organic Primitives

Viscous solutions of color, shape and odor changing polymer can be used to pattern onto the surface of material primitives. This enables designers to determine specific regions that are responsive. Figure 7-8 shows a patterned odor-color primitive enabling a globally activated material to possess discrete output patterns. This process termed *cloaking*, can be used to encode hidden information with multiple output properties. This was achieved by using hydrophobic solutions such as masks. Olive oil was used to define particular areas that should remain unchangeable, when primitive gel solutions were patterned.

Figure 7-8. Example of cloaking and disguising information through patterned Odor-Color Organic Primitive.

Patterning with liquid dopants & pH Solution

Depending upon how a material is synthesized, fabricated and patterned, activation methods can be interchanged. Instead of using pH solutions as an input to activate the material property changes, they can also be used to pattern and spatially define responsive regions in the material. Figure 7-9 illustrates how by patterning a shape changing primitive with liquid dopant at pH 10, a color-shape changing material can be achieved.

Figure 7-9. Example of anthocyanin at pH 10 solution patterned on shape changing material to induce shape-color change

8. Application Integration

This section discusses how the materiality of objects can be augmented using the *Organic Primitives*, to serve as interfaces between humans and molecules in fluids, from macro to nano scale systems. By infiltrating an object's materiality, we can take advantage of its contextual and sensorial associations to transfer biological and chemical information to a human observer more eloquently.

8.1 Food

I explore user scenarios in the context of an eating experience and present a preliminary organic primitive of a material, which is tunable, edible - allowing users to reconfigure the shape of pasta through pH. Dynamic foods can reflect the diversity that people have in their preferences and dietary constraints. Through our programmable food, users can control the shape and appearance of their food to suit their dietary constraints and preferences. During cooking and preparation, chefs can define different sets of changes permitted in the food through patterning for shape and color molecules. When the same dish is served to different users, different operations can be done to output their preference.

8.1.1 Shape Programmable Pasta

Figure 8-1. Prototype of dynamic food that alters in shape based on pH of associated sauces.

Chitosan biopolymer derived from crustacean shells have been explored as dietary fiber and edible coating the food industry [142]. Dynamic food which enables chefs to alter shape to enable different levels of flavor retention and sensory augmentation in pasta was prototyped using chitosan (figure 8-1). The shape-changing pasta was made through patterning specific areas to define shape deformations. Other form factors for dynamic food can be used as on-plate communication and embellishments, utilize condiments as input for simultaneous control of flavor, texture, and appearance. Dynamic, programmable foods can serve to aid in user preferences and dietary constraints in food and dining experiences.

8.1.2 Apple Sensor-Display

An apple can be transformed into a display by patterning color primitives tuned with methylcellulose MethocelF50 deposited onto edible starch paper (figure 8-2). When a user washes the apple, it can display information about itself such as - where to cut for kids; recipe information (ie. required apples for making apple pie); and more advanced application for sensing environmental contaminants during transport for food safety.

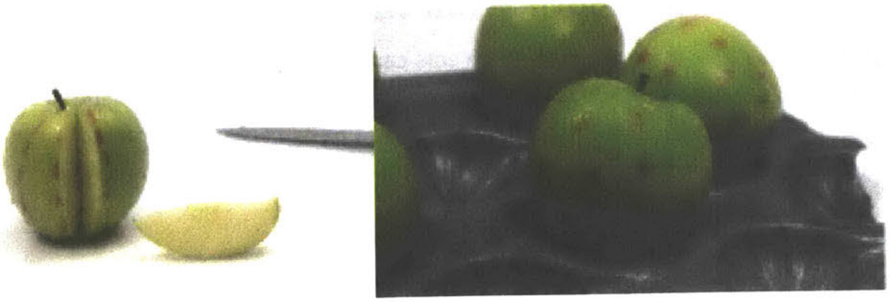

Figure 8-2. Prototype of a patterned edible film coated onto the surface of an apple transforming into a sensor-display. (left) Apple displaying marks showing kids where to cut. (right) Spotted patterns communicate contaminated fruit.

8.1.3 Biodegradable Smart Utensils

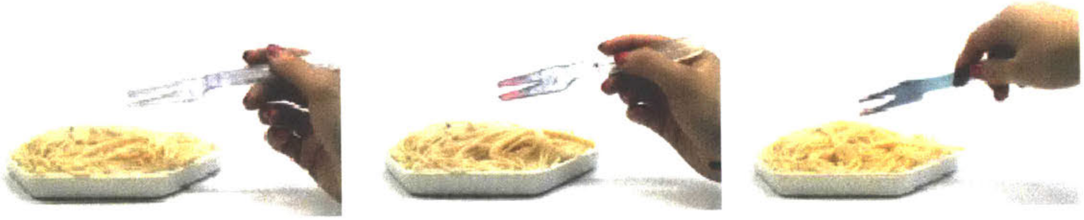

Figure 8-3. Prototype of utensils at varying levels of degradability. (right) Fully edible and biodegradble utensil. (left) Before and after image of dip-coated, 3D-printed fluidic utensil.

Utensils serve as our interfaces with food. Multiple methods of creating responsive utensils at different levels of degradability were explored and pictured in figure 8-3. The first (left) utensil is partially degradable and produced by dip-coated the prong end of a 3D-printed utensil with embedded fluidic channels, into a color primitive gel solution. The fluidic channels serve as a passive time telling mechanism for pacing the food intake for a user, by changing color. The second (right) is a fully biodegradable utensil using odor-color changing material primitives that serve to augment the taste and smell of food by releasing a sweet smell instead of adding sugar. The responsive, biodegradable utensil was created by using color-odor primitive subsets, tuned with added structural stability by mixing 250 bloom fish gelatin with the color-odor gel solution (anthocyanin-kappa carrageenan). A 3D-printed mold was created with Solidworks CAD software, where the tuned color primitive subset was then poured and casted from.

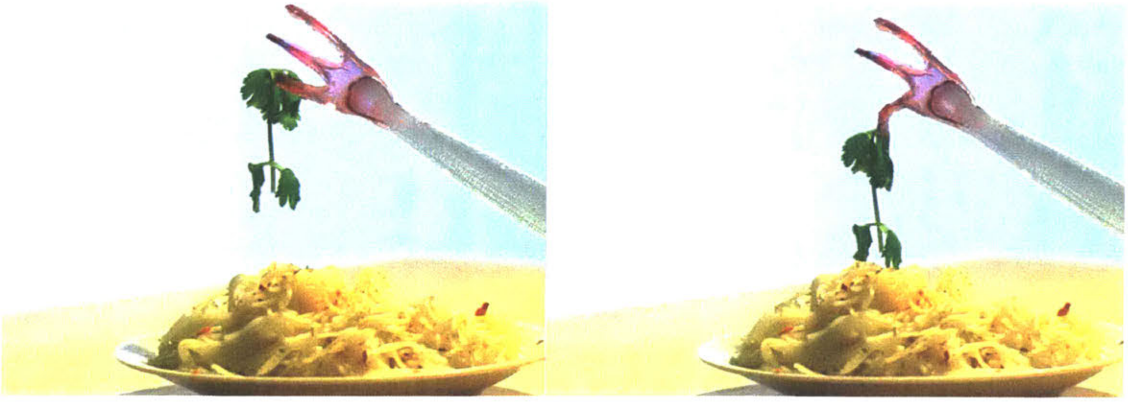

Figure 8-12. A fork communicating error as it wilts and refuses to pick up more food

Figure 8-12 shows an illustrative application prototype of a biodegradable utensil utilizing a biosemiotic sign of a wilting flower to signify the notion of error and disfunction. The shape changing fork recoils its prongs, refusing to pick up more food when you've eaten too much or too unhealthily. This application example was created by layering structural biopolymer with shape and color changing primitives. Shape change was activated passively through molded fluidic channels within the biodegradable utensil.

8.2 Environment

In natural systems, chemical exchanges represent the communication pathways between micro- and macro-scale ecosystems. On a macro scale, pH values can provide an indicator of environmental issues from ocean acidification to acid rain. In this section, I explore how to enable users to form more engaged relationships with the environment by using rain as an input and carrier of environmental information for sensory augmentation and display.

8.2.1 Bloody Umbrella

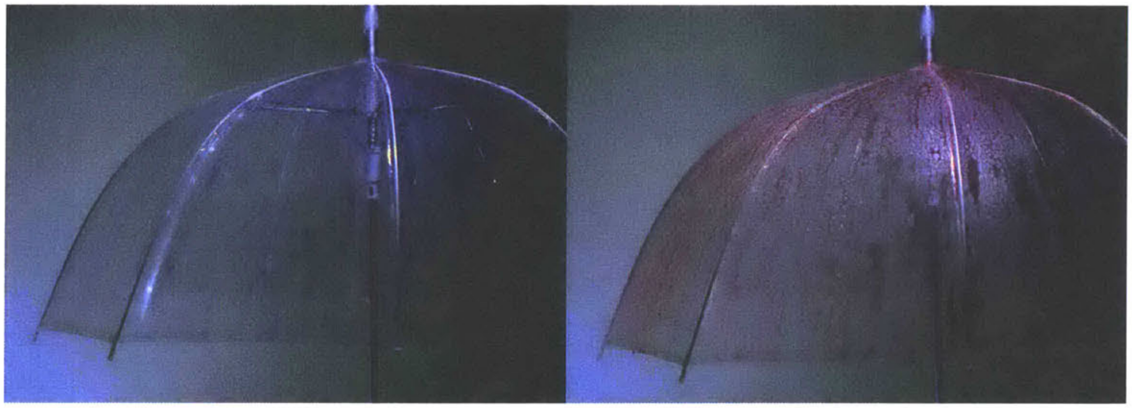

Figure 8-4. Before and after image of umbrella utilizing *Organic Primitives* for acid rain sensing and display.

Fossil fuel combustion from automobile and industrial manufacturing generates emissions of sulfur dioxide and nitrogen oxide which contribute to rainfall acidity and environmental pollution [107] [143]. Nitric and sulfuric acid molecules in acid rain are undetectable by the human eye. In this application prototype (figure 8-4), the umbrella is transformed into an interface for the information in the sky, by integrating color primitives onto the object's surface.

When the umbrella senses acid rain, bright red streaks are displayed on the surface to actuate a bloody umbrella. It utilizes blood as the design language to communicate the pain of the umbrella. The interactive object provides the user with visceral information from the sky, creating a contextual connection between the contents of rain water and environmental contaminants, along with its ramifications.

8.2.2 Architectural Material to Augment Smell of the Rain

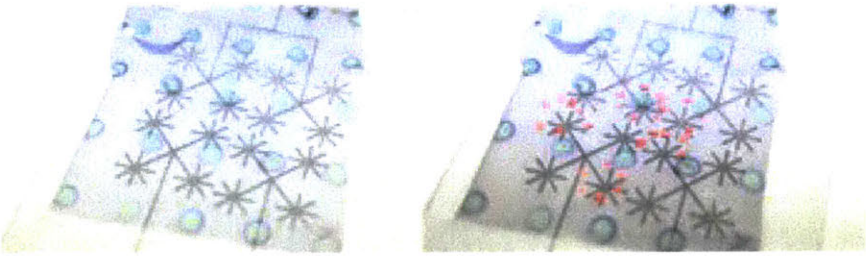

Figure 8-5. Odor and color changing architectural material for rain augmentation.

The smell of rain on a rural landscape is often romanticized by poets and artists. Within an urban environment, the smell of the rain is a byproduct of hydration of materials such as asphalt and concrete. Exhibited in figure 8-5 is an architectural material which outputs odor and color by augmenting the smell of different places. These materials can provide architects an additional sensory medium and experience to design with.

8.3 Interspecies

Plants, microbes, insects and animals utilize chemical signals for communication. In this section, I explore interactions with other species with pH-reactive *Organic Primitives*.

8.3.1 Augmenting Plant Aesthetics & Fragrance

Figure 8-6. Odor-color changing *Organic Primitives* coated on the surfaces of leaves (top row) and flower pedals (bottom row).

Gardens of flowers are often used to enhance the aesthetic environment. Gardeners often monitor and alter the pH of plants to manipulate the way they grow. Figure 8-6 shows an implementation of color and smell-changing leaves and flowers. These can be used for household decor, fostering human-plant and human-microbial interactions when users water the plant using different pH solutions.

8.3.2 Bionic Fruit for Fermentation

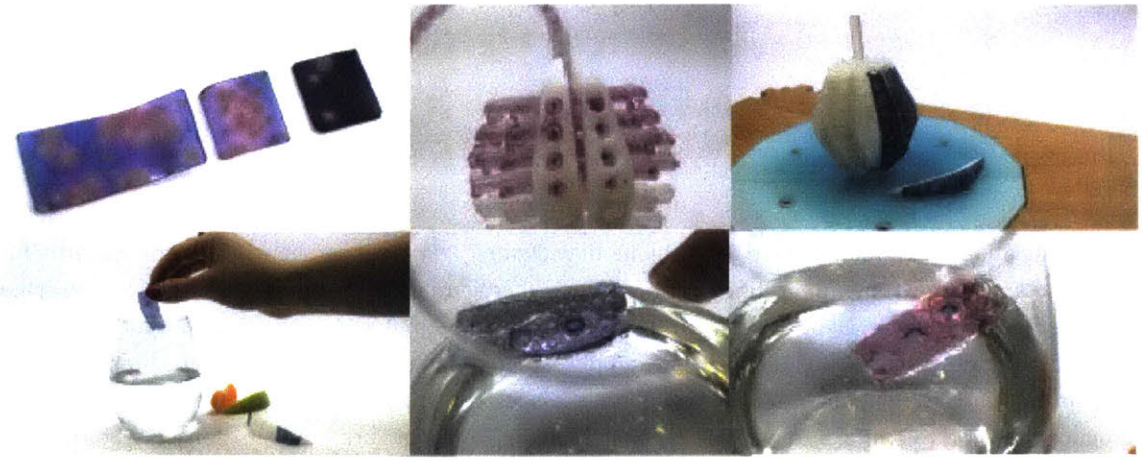

Figure 8-7. Prototype of Fructus, a bionic fruit with a fluidic 3D printed core which deposits edible pH-reactive sensor-actuators onto its surface.

Fructus is a bionic fruit prototype which generates skins of dynamic edible films (figure 8-7). A microcontroller with syringes of the three primitives are set in the table below. The device deposits the materials into the 3D printed fluidic core of the bionic fruit, through a syringe pump mechanism driven by a stepper motor. The outlet of the fluidics draws the materials to coat the surfaces of the "fruit" slices. When dried, users can peel off to use as condiment, flavoring, embellishment, or integrate in non-food applications. Future work can offer these as tools for chefs to communicate to their guests by remotely triggering Fructus to synthesize different flavors, textures, and tastes in the form of dynamic edible films.

8.4 On-Body

8.4.1 Expressive Cosmetic Display

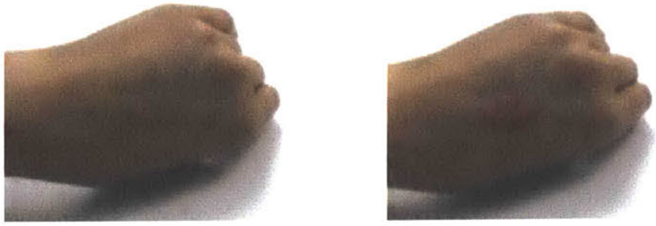

Figure 8-8. Interaction prototype of cosmetics displaying hidden information by tuning color-odor primitive with vegetable glycerin.

This application prototype in figure 8-8 exhibits how *Organic Primitives* can be used as components for cosmetic applications, by integrating primitives for dynamic makeup or for leverage pH data exerted from human body fluids. Depending upon the area applied, the material senses and outputs based on contextual inputs such as body fluid. For example, applied under the eye directs sensing of tears to change color of eye-makeup. Applied throughout the back can implicate sensing of lactic acid in sweat produced by skin microflora to activate an odor release. Personal care products can also be used as inputs for designing interactions activated by household chemicals such as soaps, typically spanning between pH 8-12. Cosmetics have typically been used to accentuate facial features and communicate character. Dynamic cosmetics can be used to alter human appearances for theater and performance since character progressions tend to change rapidly. Further investigation is needed to actualized this application in the wild.

8.4.2 Saliva sensing for oral health

Figure 8-9. Application of functionalized fluidic toothbrush

pH is an important factor for maintaining oral health as certain bacteria contribute to fluctuations. Prolonged periods of acidic oral environment leads to cavities. The oral environment of the mouth maintains an average of pH 6 in the mouth. Figure 8-9 is an illustrative example of material primitives integrated to create a responsive toothbrush where the bristles can sense and survey the pH of a user's oral environment.

8.4.3 Perfume Responsive Textiles

Figure 8-10. Color of clothing changes due to pH of perfume

New experiences with clothing can be created by using perfume contents as an input to create textiles that can change properties. The application prototype shown in figure 8-10 utilizes the modifiable design vocabulary of "refreshment" associated with water mists and shower heads. By using the pH of perfumes as the fluid input to initiate color change in clothing design, the message of "refreshing the way you look by changing how you smell" is mapped to misting of perfume.

8.4.4 Interacting with the Skin by Tattooing Biosensors

The maturation of the field of biotechnology suggests new use cases outside the domain of research. Dermal Abyss (d-abyss), is an approach to bio-interfaces in which the body surface is rendered an interactive display by patterning biosensors into the skin whose colors change in response to variations in the interstitial fluid. This modern interpretation of self-expression blends recent advances in biotechnology with traditional methods in tattoo artistry. d-abyss is designed to use the aesthetic component, the permanence, and the publicly visible nature of tattoos to encode information. As shown in figure 8-11, traditional inks are replaced with biosensors that colormetrically index the concentration of sodium, glucose, and H+ ions (pH) in the interstitial fluid of the skin: the pH sensor changes between purple and pink and the glucose sensor between blue and brown; the sodium and a second pH sensor fluoresce at a higher intensity under UV light. Applications of d-abyss can be used in medical, lifestyle, and security domains. d-abyss describes a platform in which tattoos form wearable displays inside the skin, and the body's metabolism is a semi-deterministic input.

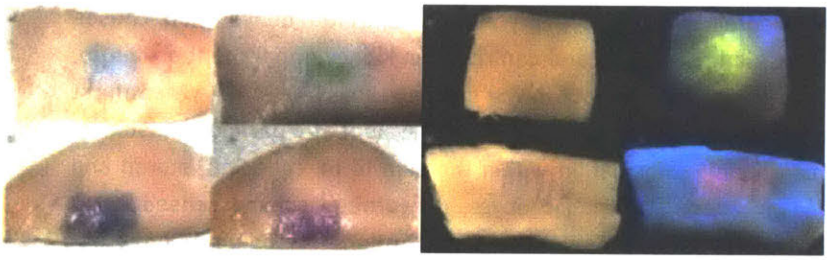

Figure 8-11. pH, sodium and glucose bio-sensors tattooed onto an *ex vivo* pig skin model

The primary function of tattoos are to enable users to embody a sense of identity and expression, however tattoos remain unchanging and passive. Incorporating biosensor tattoos within the skin introduces a dynamic dimension to tattoos. d-abyss offer design possibilities which harness the existing cultural affiliations and functionality of tattoos, while extending its capabilities to encompass health monitoring, wellness advocacy, and data encoding. These examples foresee the potentiality of d-abyss in possible scenarios when biosensor tattoos are ready to implant into humans.

Monitoring Body Fluid Data

Embedded skin biosensing tattoos enable continuous monitoring of interstitial fluid within the skin over prolonged periods of time. This data can be utilized in parallel with other biometric wearables to achieve greater understanding of the chemical and biological conditions of the body.

- **pH Balance**

  The body maintains a homeostatic state where pH is kept at a specific domain for the proper functioning of cells. Blood maintains at a pH of 7.35-7.45 [91] and small deviations away from this range can result in serious health conditions. Within the interstitum of the skin, pH is 7.35. Since interstitial fluid is comprised of a dynamic flux of solutes diffused from plasma within capillaries, pH can be an indicator of bodily responses upon local as well as systemic pathology. One such systemic pathology is diabetic ketoacidosis, a common condition in poorly-controlled diabetes in which a lack of insulin forces cells to rely on the oxidation of fatty acids for fuel [144]. The byproducts of fatty acid metabolism are acidic ketone bodies, which subsequently lowers the pH of blood and interstitial fluid to dangerous levels.

- **Diabetes**

  Diabetics require glucose levels to be monitored by piercing the skin, sometimes 3-10 times per day to draw blood or plasma. Such information reveals glucose changes in the body and aids diabetics with health management tasks from diet adjustments, exercise, to medication intake. Glucose levels in the dermis have shown to be an encouraging sensing sight as it possesses little variation among subjects, is heterogeneously distributed throughout the skin, and dermal glucose concentrations remain promisingly close to values from blood glucose. The volume of glucose in the dermis is typically 0.40 ml/g with a volume density of 44 percent water [145].

- **Dehydration**
- Typically thirst mechanisms are triggered by a decrease in plasma osmolarity which excites the hypothalamic thirst center and induces dry mouth. Extreme dehydration causes hydrostatic pressure in the kidneys to drop and can result in kidney impairment or failure, as buildup of nitrogenous wastes accumulate in the bloodstream [146] [147]. While thirst mechanisms can signal to the body when dehydration is prevalent at a particular instance, long-term monitoring can enable information within the skin to be collected, stored and analyzed over prolonged durations. Utilizing d-abyss, dehydration can be sensed using sodium fluorescent biosensors. Higher monotonic concentrations of sodium solutes are prevalent in tissues when water loss occurs - activating the sodium biosensor.
- **Tattoo with Biometric Wearables**

As an example, different biosensors including sodium, pH and glucose can be incorporated in one d-abyss design on a user's wrist. A camera-enabled smart watch is worn on the same wrist, enabling continuous monitoring of the tattoo. The watch emits a light at 507nm, causing the tattoo to reflect back a tattooed word, which reads "ON" in a fluorescing green color at 532nm - indicating sodium levels. A flash of 530nm light enables the watch to read the fluorescent pH sensor as it reflects across a spectrum. The watch then snaps a photo of the tattoo to collect the sodium and pH level of the user. Over time, data about the user's interstitum is collected, aggregated, and communicated to the user. If implemented at scale, d-abyss has the potential to elucidate how such bodily fluid data in different users are affected by differences in medication intake, mood, lifestyle, or diet.

Maintaining a Culture of Health

d-abyss can enable users to extend the function of self-expression and aesthetics of tattoos beyond personal representation, into a new medium advocating self-care and healthfulness.

- **The Self and I**

  By utilizing the body's metabolic processes as an input for activating d-abyss tattoo displays, this can potentially extend signals of a user's health onto the aesthetic design of the tattoo. This incentivizes users to maintain care for their health and by extension, maintain desirable aesthetics of the tattoo. Chromogenic glucose and pH d-abyss can enable users to scribe symbolic and aesthetic designs on the skin while providing a quick glance to the user about their health.

- **Finding Others**

  There are numerous existent subcultures from bodybuilding to hardline straight-edge culture, which incorporate a notion of identity, belonging and status with health. Biosensor tattoos can facilitate a method of social affordance and display as indicated by health status. A dynamic d-abyss tattoo can enable users to identify other die-heart straight-edge individuals.

Encoding and Encrypting the Skin

Tattoo designs and symbols inscribed onto the body often signal information involving personal identity, tribe belonging and group affiliations. With d-abyss, tattoo designs not only signify an individual's personal expression but can also serve as encrypted health information.

- **Aesthetics as Data**

  As the tattoo design may produce changes over time to reflect different solute concentrations in the interstitium, the dynamics of the tattoo design reflect medically relevant information about the body. The tattoo may appear as aesthetic designs but from the lens of a doctor or health specialist paired with specialized software or decryption device - it may uncover underpinnings for an individual's health progress over time.

Wearable devices and watches equipped with optical tools can also be used in conjunction with d-abyss for medical diagnostics.

- **Unique Identifiers**

  Bar codes and QR codes are computer coded designs which enable software recognition of arbitrary graphic symbols. Such designs can be encoded into tattoos to enable

9. Conclusion

Molecular Design Interactions bridges molecular phenomena with human scale interaction with the information contents of fluids. It provides a framework to facilitate the design of interactions with organic, fluid-based system. This thesis offers designers a toolbox to create multi-modal output to 'hack' the materiality of everyday objects - transforming them into both sensors and information displays. Because of the biocompatible nature of these sensor-actuators, fluids from raindrops, ocean water, body fluids, food sources, and plant/microbe excretions can be utilized as inputs to activate the output properties of the material primitives. This approach of employing organic compounds as molecular I/O offers a method to leverage an object's contextual meaning to enable novel sensory experiences by augmenting its material properties.

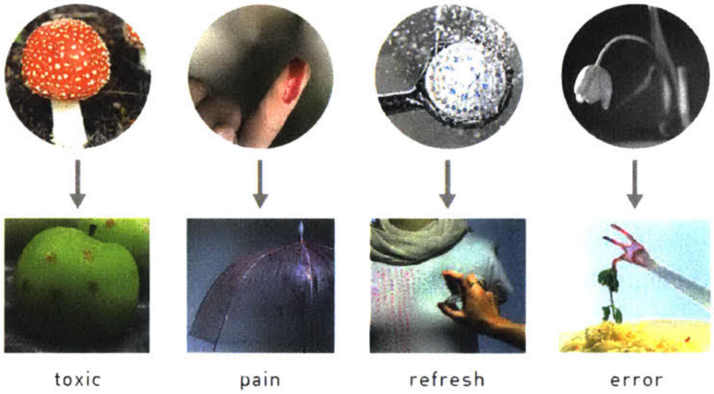

Figure 3-8. Interactive artifacts integrated with *Organic Primitives* (bottom row) utilizes semiotic signs of toxic mushrooms, bleeding, water mist, and wilted flower (top row) to communicate messages of toxicity of toxicity, pain, refreshment, and error.

The information contents encoded within inhalant and ingested goods through

food materials, oral implants, medicines, and perfume are primarily developed by technologists situated in biological and chemical disciplines such as food science, biomedical engineering, synthetic biology, organic chemistry and biomolecular sciences. The tools and languages in these disciplines are largely inaccessible to users of these technologies. These technologies are consumed and embodied every day through the things we eat and the products we use. Yet users themselves have limited abilities to know or control the contents of the things they consume. By empowering people to elucidate the molecular world around them, it is my belief that this can incite a sense of wonder, responsibility, and agency for people to safeguard the natural systems that surround - be it the environment, health, organisms or food. Social dialog and discussion on biological and chemical technologies can be shifted from grave distrust towards science into interventions for progress. When we enable people the tools to take responsibility for the wellness outcomes of complex systems through measurable daily interactions, we can transfer the power to change them to individuals - that when aggregated, form the whole.

Future Roadmap

Offloading Design Parameters to Software Tools

Data generated from material characterization and evaluation serve as design parameters in the *Molecular Design Interactions* framework. This nonlinear and multi-dimensional data can be integrated into software design tools, which incorporate specific material formulations, recipes and fabrication conditions as digital and material design processes. Furthermore, the synthesis and printing of these materials can be automated onto printing platforms.

Expanding the Library of *Organic Primitives*

While pH serves as a starting point in this body of work, I envision a 'library' of material primitives, which can facilitate designers to interact with a variety of organic systems, across a range of signals. Primitives which can transduce molecular signals such as glucose, eH, pCO2 , sodium, magnesium, calcium, potassium, hormones and dioxins, can further enrich possibilities for human scale interaction with organic systems. The methods introduced in this paper can be used to develop sensor-actuators that are responsive to any number of chemical and biological stimuli. As the library of primitives expand, material robotics, which can sense and output a multiplicity of behaviors can be possible. This can be achieved by utilizing the introduced techniques from the design space with multiple molecular I/O dopants, to build complex logic and behavior.

Smarter, Faster, and More Optimized

We can create higher-order functionality by using *Organic Primitives* as building blocks for complex circuits involving logic gates. Further development can combine disparate components into integrated devices. Current rates of change rely on diffusion kinetics for material change, so output speeds are limited by diffusion rates of water. Future work can involve the use of volatile organic compounds (VOCs) as activation mechanisms where gas is the main fluid as opposed to water.

Empowering End Users to become Developers and Designers

Currently, *Organic Primitives* are generated by developers with moderate chemistry knowledge for use by designers with little to no chemistry background to create interactions for end users. As *Molecular Design Interactions* advances, the roles of and between developer, designer and end user can be reformed where the end user can re-inform the system to become developers themselves. This would in effect result in a generative outcome where the practice of *Molecular Design Interactions* produces new developers by refining the process through simplification, ease of use, standardization and educational development. The low-cost nature of developing and utilizing Organic Primitives and accessibility of the components makes this a scalable and feasible outcome.

Beyond the Organic

Can primitives be created that allow us to design interactions beyond fluids and organic systems? While the notion of *Organic Primitives* were based on enabling access to carbon-based systems, there are many inorganic systems that are inaccessible to humans. These systems can be classified in a variety of ways and may be inaccessible for a number of reasons including too large, too small, too dangerous or too far away. The future of *Molecular Design Interactions* may approach these systems by finding the most accessible path or identifying the most crucial. An example of accessible path would be creating "rock" or "geo" primitives from house hold chemicals by way of growing piezoelectric crystals and minerals at home or utilizing rocks from local environments to derive interactive elements from. A crucial path would be in finding ways to create access to the difficult endeavor of sensing small molecules in the vapor phase. As demonstrated by animals with an acute sense of smell, they are able to detect gaseous information to identify patents with cancer.

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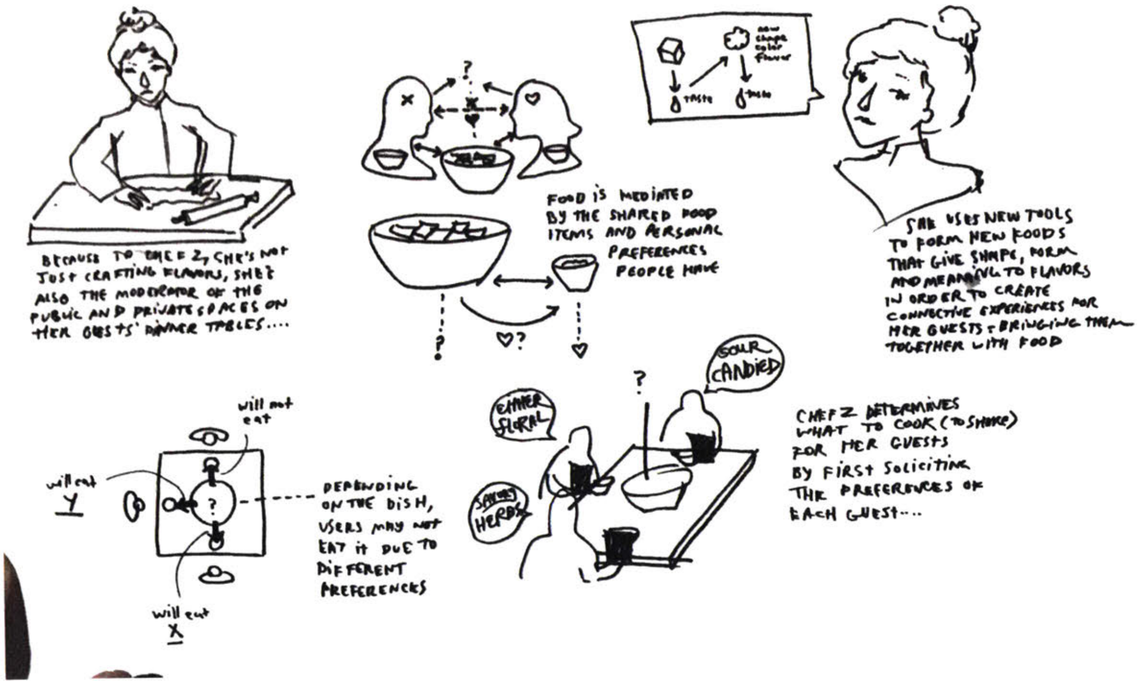
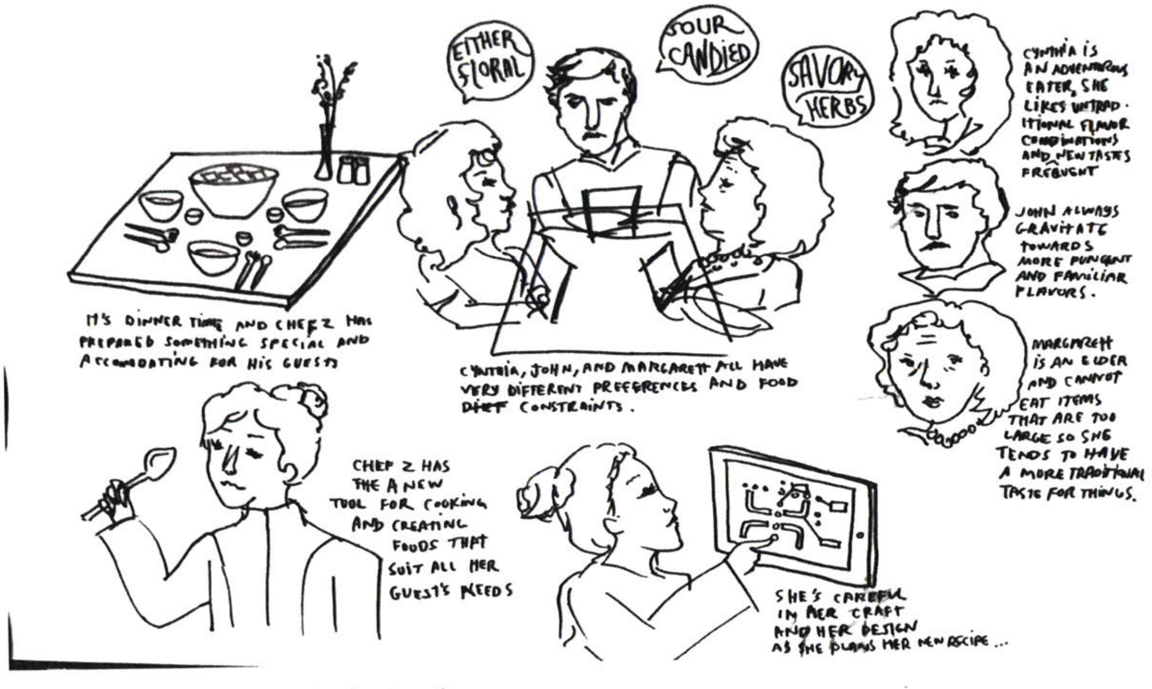
Appendix

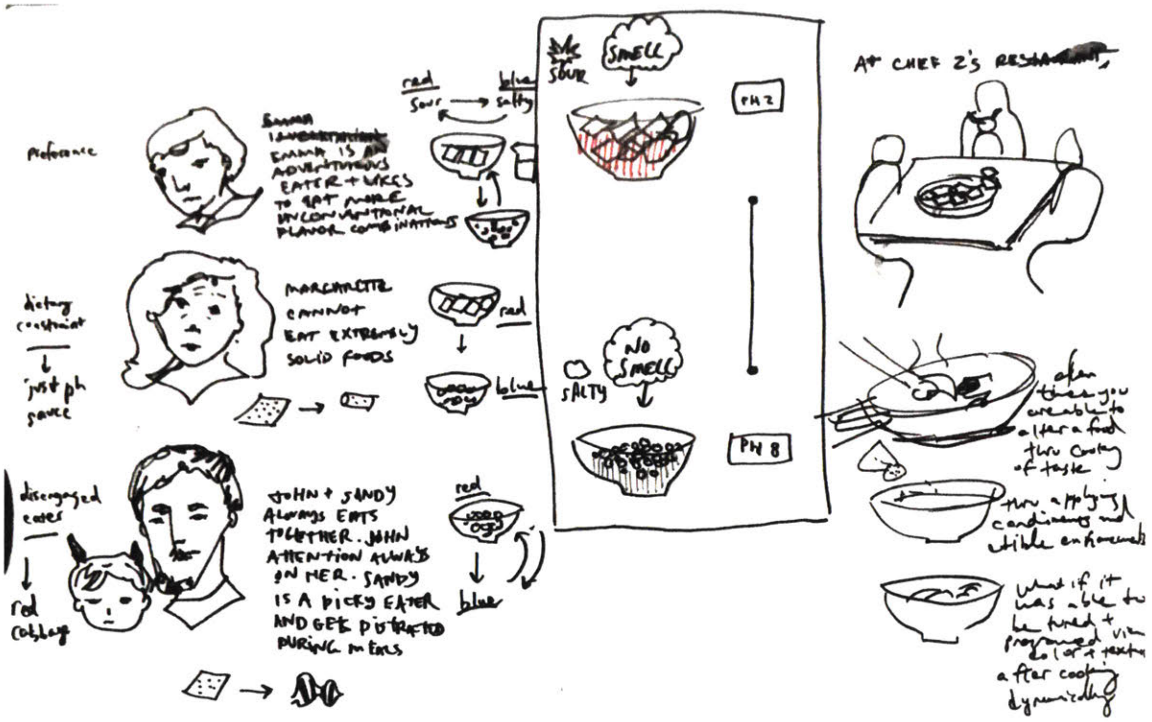
A-1 User experience involving shape and flavor programmable pasta



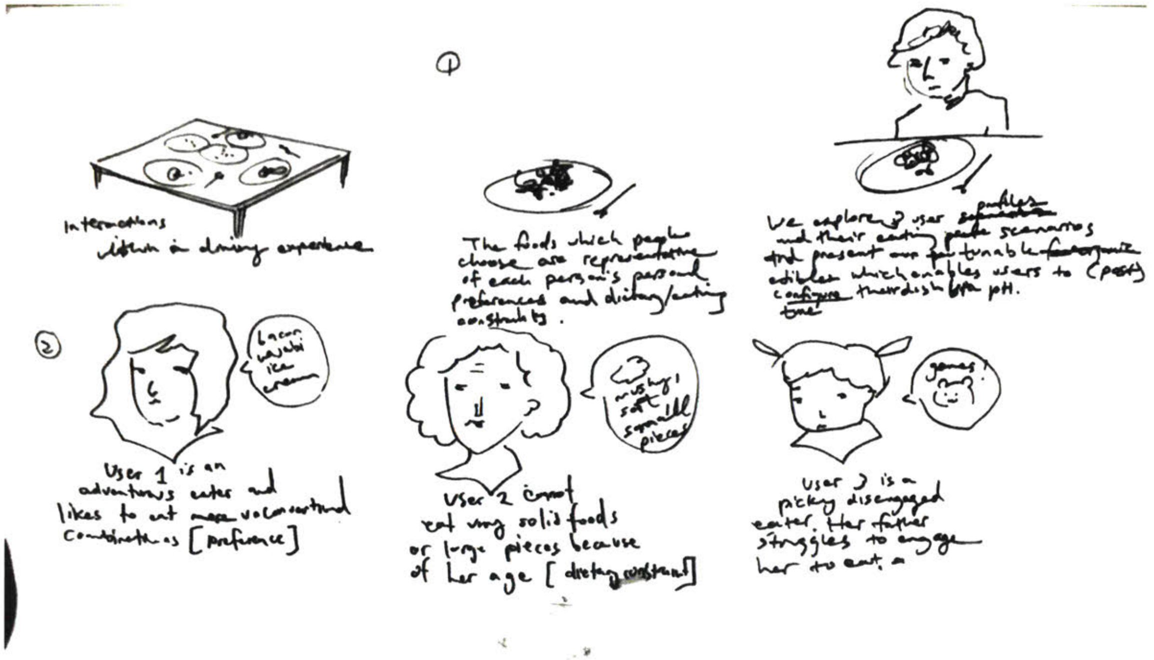
A-2 User experience involving chef considerations



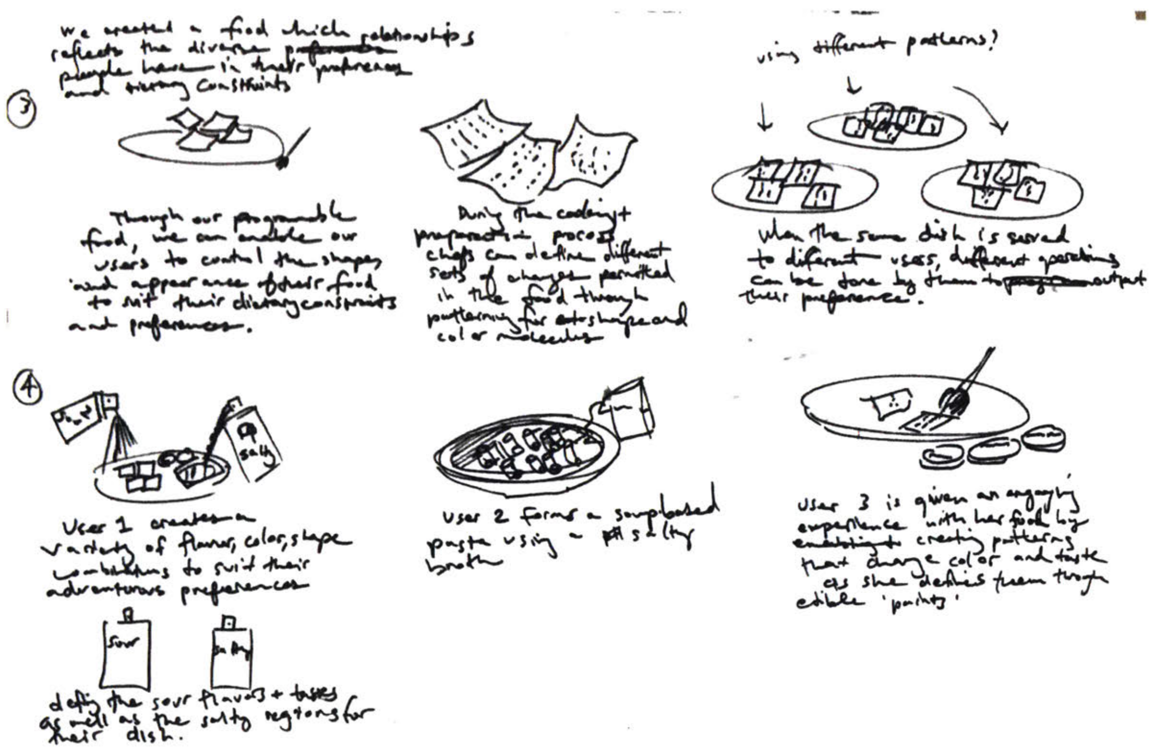
A-3 User experience involving food and taste preferences



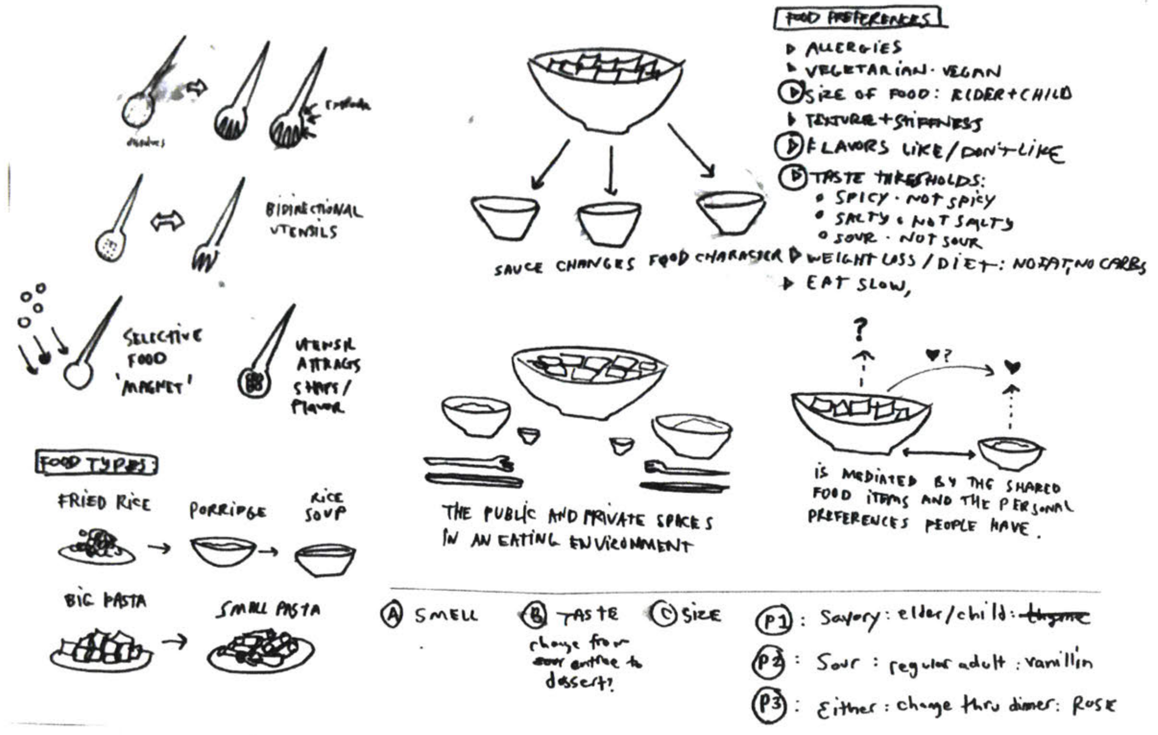
A-4.1 User experience involving dietary requirements



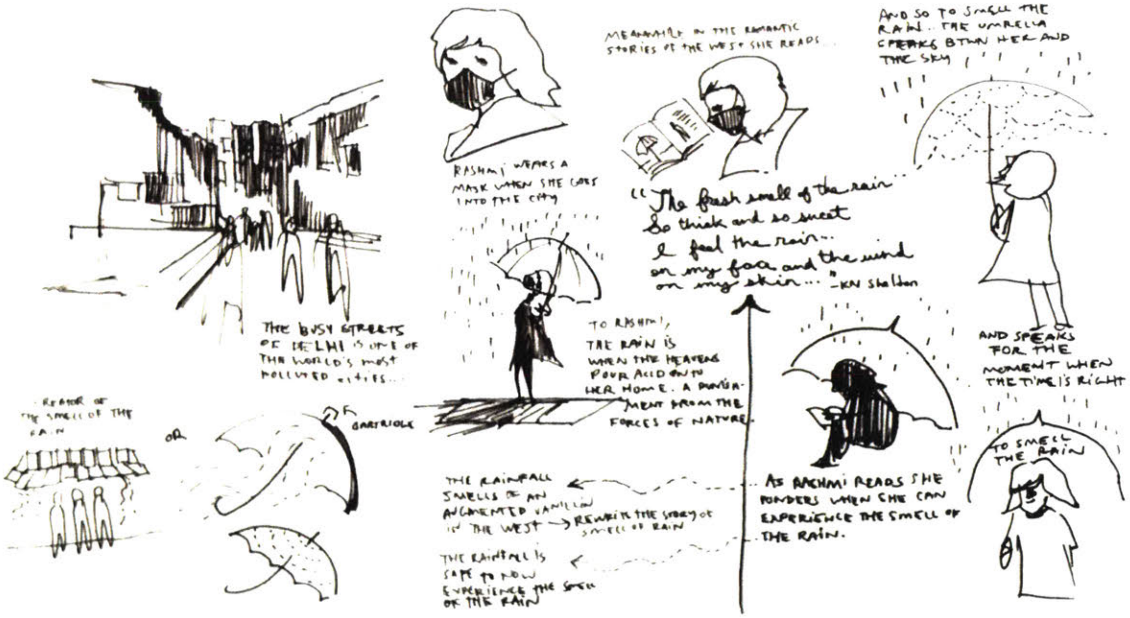
A-4.2 User experience involving dietary requirements



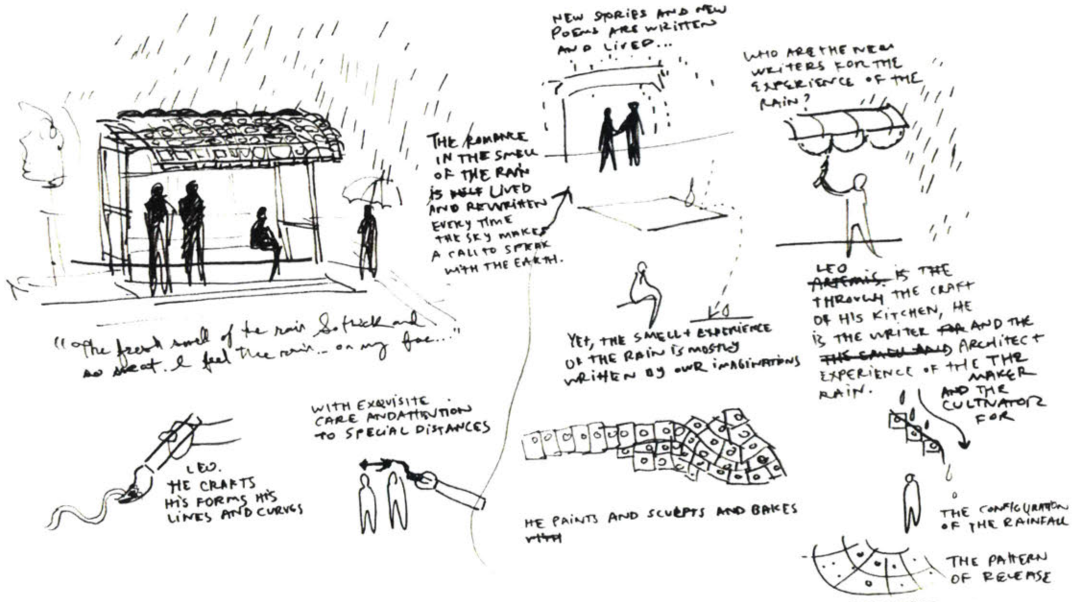
A-4.3 User experience involving dietary requirements



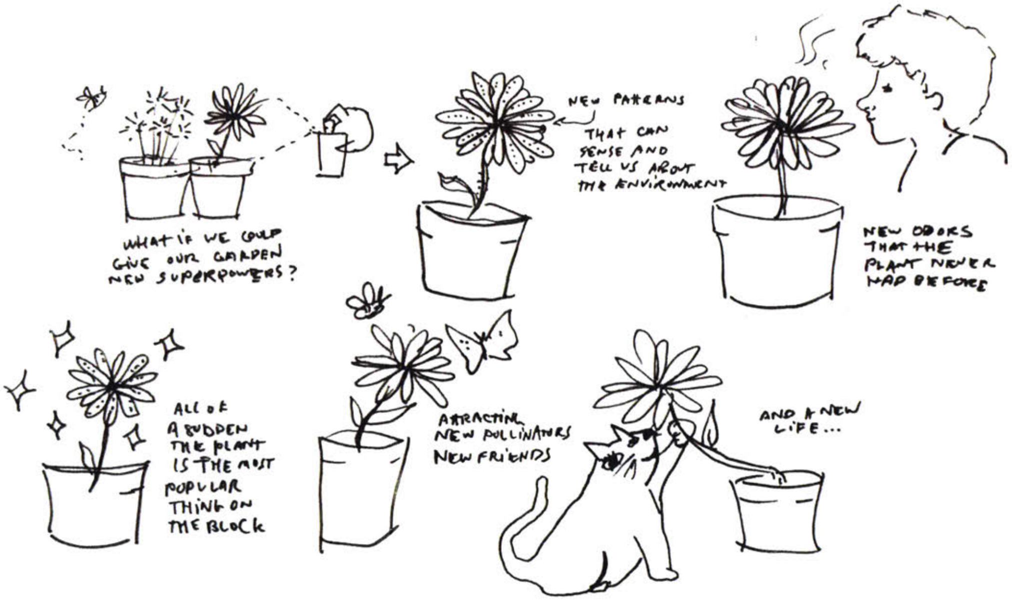
A-5 User experience involving preference parameters and user profiles



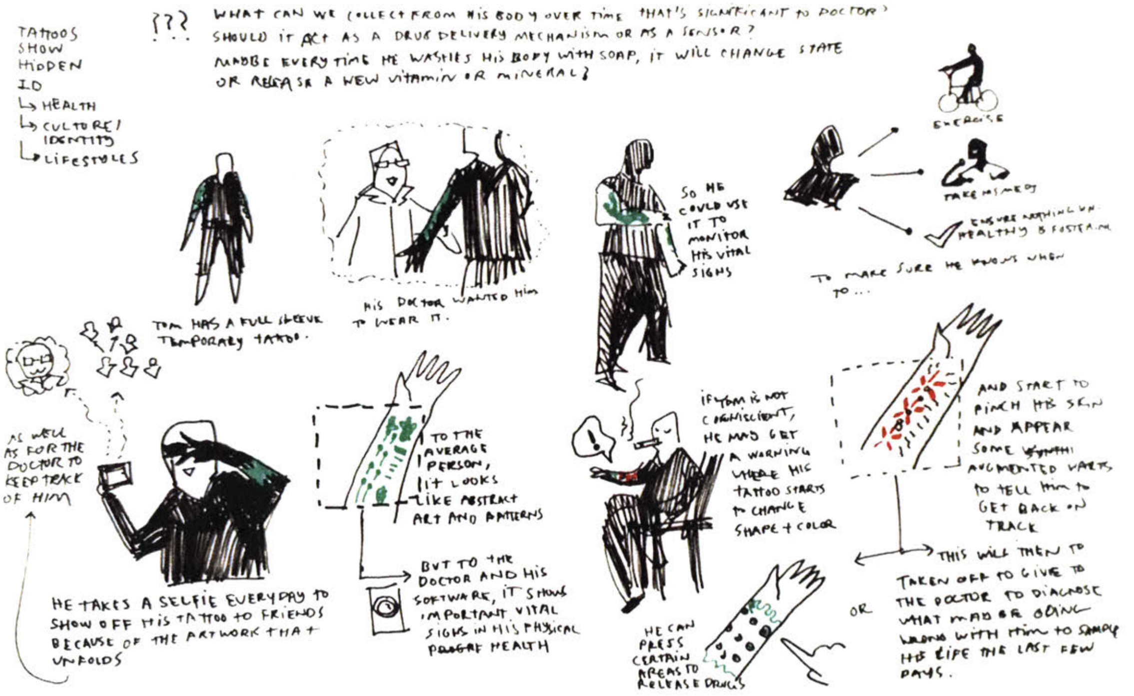
A-6 User experience of rain sensing umbrella



A-7 User experience of the smell of the rain



A-8 User experience of interactive plant



A-9 User experience of smart tattoo for health