Organic Primitives: Synthesis & Design of pH-Reactive Materials with Organic Molecules for Biocompatible I/O

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Figure 1. Organic Primitives adapted onto the surfaces of ordinary objects to transform an umbrella, flower, toothbrush, utensil and apple into information sensors and displays powered by pH in fluids.

ABSTRACT
Whether it be the information embodied within the rain, an aquarium, a dinner plate, or in human tears, pH values constantly fluctuate to reflect chemical processes in natural systems. Utilizing a selection of pH-reactive organic compounds, we have synthesized tunable, material-based sensor-actuators that can output a spectrum of colors, degrees of shape deformation, and the switching of odorous to non-odorous based upon pH value; we call these base sensor-actuators Organic Primitives. In this paper, we present the approach of utilizing the pH-reactive organic molecules vanillin, anthocyanin and chitosan as mechanisms for odor, color and shape actuation. These primitive mechanisms can be used to create responsive, functionalized biopolymers across a variety of form factors, including fluids, fibers, films, sheets, gels and dimensional forms. These biopolymers can be used alone or in combination to form simple but highly functionalized systems. The materials are edible and enables fluid-based sensing-actuation near, on, or even within living systems. In this paper, we demonstrate a design space which can enable higher-order functions in the material system. The design space highlights a variety of techniques to control the material system through defining pH inputs, sequencing techniques, encoding hidden 2D and 3D forms, patterning, and compositing functionalized biopolymers. Through this molecular-scale approach of creating tunable sensors through material synthesis, we explore human-material interaction in four application contexts: edible, cosmetic, environmental, and interspecies. In order to evaluate the functional parameters of our material formulations, we evaluate the output properties of individual pH-reactive molecules in the form of films as a function of pH 2 – 10. Though this work only explores pH response, the methods here could potentially be used to create materials responsive to any number of environmental stimuli, expanding upon the library of input-output devices for HCI.

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

In this paper we present Organic Primitives: biomaterial building blocks that sense information within fluids and convert them into human-readable outputs to enable fluid-based sensing and actuation. We propose an approach of synthesizing material interfaces to deliver dual sensing-actuation, offering novel application domains that serve to bridge the molecular world with human scale interaction.
Beginning with pH, we developed material-based sensor-actuators, which convert biochemical pH signals into odor, color, and shape change. We utilize the organic molecules anthocyanin, vanillin and chitosan as dopants in the functionalization of biopolymers; their molecular reaction to pH enables pH information to be sensed as well as communicated to the user. Within our approach, we utilize the Brønsted–Lowry acid–base reaction as a core driver of our material property transformations due to the rapid, reversible and bidirectional nature of the reaction.

To demonstrate the range of methods for controlling and designing with Organic Primitives, we developed a design space which and a series of application scenarios

We introduce a molecular-scale approach to embedding sensing-actuation within material objects, which offers a variety of distinct advantages: these systems (i) have self-contained functionality as 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 in both biological and electrical systems; (iv) are biocompatible, biodegradable, and edible; (v) are compact, soft, muted, and unobtrusive; (vi) can utilize personal data from body fluids as natural input signals; (vii) open up additional modalities including taste and smell; (viii) are programmable and enable multiple inheritances of output properties such as color, shape and odor change.

Our goal is to enable multiple inheritance of material output properties to broaden the capabilities for user experience design. We demonstrate methods for integrating these Organic Primitives into four application domains through a series of objects functionalized using our materials. The distinct capabilities enabled by this material system include the ability to “hack” the materiality of objects to transform organic objects into both a sensor and an information display. Because of the organic nature of our sensors, we are able to utilize natural inputs as such as raindrops, ocean water, body fluids, food sources, and plant/microbe excretions for control. Using these inputs, we can augment sensory experience between edible, environmental, interspecies and health 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 Natural Inputs

Since pH is an inherent characteristic amongst all fluids, natural inputs from different contexts can be used to activate the material interaction. This includes inputs through rain drops, ocean water, body fluids, food sources, and organismic excretions from plants and microbes.

Information Representation & 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 enables multi-sensory augmentations through odor, color and shape activation where objects inherit intrinsic characteristics.

RELATED WORK

Interfacing with Biological and Chemical systems

Biocompatible materials for sensing and actuation are becoming increasingly relevant in the HCI community with the rise of research towards the development of soft, low power devices that operate close to and even within the human body. Recent developments like bionic contact lenses leverage information excreted from the body such as the composition of tears for medical diagnosis and targeted drug delivery [13]. Google X’s pill-based authentication utilizes stomach acids to power the biodegradable device as a temporary key for unlocking devices [49]. Biopolymers such as silk have been used to develop resorbable electronics as implantable devices that degrade after a surgical procedure [47]. However, the process of developing biocompatible devices with embedded inorganic semiconductor materials for close contact or ingestion with humans remains a challenge and is subject to a great deal of regulatory processes due to safety and privacy concerns. In the interest of enabling user interaction that operates close to and within organic systems, there exists a rich repertoire of stimuli-responsive molecules that can be harnessed by reverse-engineering edible matter and then synthesized into material-based sensor-actuators, yielding safe, ingestible and biocompatible I/O devices.

Information Processing with Fluids

There is a great deal of information encoded within everyday fluids from the environment, our food, the body, and organisms we cohabit with. The density of this information supercedes the capabilities of what our modern electronic chemical sensors can detect. Researchers in chemical and biological engineering fields often rely on chemical mechanisms and reactions for sensing, chemical processing, and analysis. Numerous assays such as enzyme-linked immunosorbent assay (ELISA), Polymerase Chain Reaction (PCR) and Bicichoninic acid assay (BCA) utilizes biomolecules such as enzymes, proteins and antibodies for analyzing chemical and biological samples. In the field of microfluidics, various reactions for chemical synthesis, bioanalysis and nanoparticle production have been carried out through manipulating the flow of fluids in micrometer-scale channels [48][54][56][48][45]. The physical nature of a material’s composition can lend itself to complex
reactions that enable us to leverage physical phenomena for information processing. Water droplets 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 [10]. In synthetic biology, organic compounds are used to develop synthetic minimal cells as biosensors, liposome bioreactors, protocols and chemical messengers with natural cells. These molecular machines are often made from organic compounds such as phospholipids, cholesterol, and fatty acids to encapsulate facilitating transcription/translation machinery to achieve controlled gene expression [4]. However, while many methods for chemical sensing and processing have been developed within chemical and biological engineering disciplines and can be leveraged for user interaction design with organic systems, there has been little emphasis within the field for the development of I/O mechanisms which cater and advance the interests of HCI. Furthermore, many of these existing chemical sensing mechanisms operate and function purely in fluid form. In order to utilize fluid-based data as input and design interactive experiences with natural systems, chemical sensing mechanisms must to be materialized into form, and actuation mechanisms for information representation need to be realized.

**Material-Mediated User Interaction**

“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

Recent developments in HCI have sought to develop material based interfaces incorporating sensing and action processes including sensing, computation, actuation and communication within a unitary material. Approaches for embedding these processes into a single material system are varied from composite-based [21][23], to utilizing biological material for humidity sensing and actuation [57]. Fluid-driven material interfaces. The development of wearable input and output systems [1][29] demonstrate a growing interest in diminishing the scale of sensing and actuation mechanisms within a compact system.

**PH-REACTIVE ORGANIC PRIMITIVES**

Utilizing pH-reactive molecules anthocyanin, chitosan, and vanillin, we introduce our approach to material design from dopant selection to material synthesis, output characterization, and composite evaluation.

pH-reactive Organic Primitives begin with the selection of dopants and structural biopolymers. They can then be synthesized into a variety of form factors and output combinations. Logic and specific activation parameters can be encoded by tuning the material composition to the application.

![Figure 2. Examples of design process: (clockwise from top left) shape-color primitives output different angles based on pH; encoding behaviors through compositing and patterning: global, local and discrete activation with fluidics; application of a smart compostable fork](image)

**Organic Molecules as Dopants for Sensing-Actuation**

Many organic molecules can react in response to pH, but only a subset of these can react to generate products that yield human-readable changes such as color, odor and shape. In this work, central criteria for our selection of these molecules are factors that ensure safety, accessibility and biocompatibility, to encourage researchers to develop their own pH responsive material interfaces without requiring special tools or facilities. In this section, we discuss the chemistry of the pH-reactive molecules anthocyanin, vanillin, and chitosan, and offer mechanisms that facilitate other researchers in identifying new pH-responsive molecules.

**Anthocyanin: Color-Changing Reconfigurable Molecule**

![Figure 3. Chemical structure of anthocyanin](image)

All tissues of higher plants possess the flavonoid anthocyanin, a pigment that changes color when quenched in 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 [5]. Anthocyanins and anthocyanidins (the sugarless form) are a class of molecules that includes cyanidin, delphinidin, pelargonidin, peonidin, and malvidin among others. Under different pH conditions, the hydroxyl and/or methyl ether groups attached on 3, 5, 6, 7, 3’, 4’ and/or 5’ (figure 1)
undergo reversible structural transformations and ionizations. This restructuring affects which spectrums of light are absorbed, giving rise to color changes [8][3].

**Vanillin: Odor-Switching Flavor Molecule**

![Chemical structure of vanillin]

**Figure 4. Chemical structure of vanillin**

Vanillin is a flavor molecule extracted from vanilla beans, which contribute to its characteristic aroma. 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 [1]. Neppel et al uses vanillin for end-point determinations in olfactory titrations [2]. There is little literature on the mechanisms for odor release of vanillin under different pHs. However, it is possible that the 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, under basic conditions[33].

**Chitosan: Swelling & Shape-Memory Macromolecule**

![Chemical structure of chitosan]

**Figure 5. Chemical structure of chitosan.**

Chitosan, a polysaccharide derived from the exoskeleton of shrimp and crustaceans, has been identified as a promising pH-sensitive polymer because of its biocompatible and biodegradable properties [pH sensitive polymers] . Its NH2 amino functional group is protonated in low pH conditions, causing the polymer to swell, while in a high pH condition, chitosan returns to its collapsed state. The tunable and shape memory characteristics of chitosan have attracted great interest among biomedical engineers for use as nanocarriers for targeted bio-sensing and drug delivery applications [chitosan for drug delivery]. Chitosan has also been investigated by food scientists and chemists for use as a dietary fiber and in edible coating to increase shelf life of food [chitosan edible films; edible coatings].

**Material Synthesis**

![Chemical structure of Kappa-Carrageenan (top); Sodium Alginate (bottom)]

**Figure 6. Chemical structure of Kappa-Carrageenan (top); Sodium Alginate (bottom)**

This section we discuss functionalizing base materials with biopolymers. Different polymers and cross-linkers can be utilized as matrices for the pH-responsive dopants depending upon the intended application and specific characteristics desired. In our investigation, we have found sodium alginate and kappa-carrageenan (figure 5) to be good candidates for materializing the pH-responsive molecules due to their semi-permeable structures, consistent behavior, rapid diffusion rates, and relatively neutral pH. However, further investigation will be needed to fine-tune adherence, crosslinking, and buffering capacities of different molecule-material combinations.

We selected kappa-carrageenan to be used as the structural base of anthocyanin. 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. [BPJ]

Vanillin was encapsulated in an alginate gel to form our scent-changing solution. 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 our trials suggest the gel has little effect on vanillin volatility and scent release.

**OUTPUT CHARACTERIZATION**

We evaluated the individual material outputs of our Organic Primitives for odor, color and shape change. The results show the effectiveness of the pH-reactive molecules as tunable sensor-actuators. Our study also offers instructive parameters for how one might synthesize materials with multiple inheritances of output properties for the development of more sophisticated outputs in the Organic Primitives.

Our test samples were all prepared by pouring Primitive solutions (detailed in the following sections) into small
polystyrene petri dishes (VWR). The samples were dried overnight beneath fans before being removed from the dishes. Our stock solutions of pH 2, 4, 6, 8 and 10 were made using citric acid (H3Cit), sodium citrate (NaHCitrate), sodium bicarbonate (NaHCO), and sodium hydroxide (NaOH) and deionized water (H2O), as shown in our supplementary materials. The pH solutions were tested and confirmed using the Oakton pH5+ EW-35613-52 meter, as well as pHdrion litmus strips.

**Color as a Function of pH**

**Figure 7. Material activation across different pH**

**Rate of Change and Spectrum of Colors**

Our 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. We tested samples with our 2, 3, 4, 5, 6, 7, 8, 9, and 10 pH 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 a Colormunki spectrophotometer, providing a CIE L*a*b* color value for each sample (figure 2).

Depending on pH, the sample color changes from redder tones to green. The visual pH responsive of anthocyanin has been well documented because of its use as a pH indicator; this trial demonstrates that embedding anthocyanin in a structural polymer has not negatively affected its responsiveness.

**Figure 8. 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.**

The speed of the color change was difficult to measure quantitatively because the color changed most significantly upon contact with pH solution and then would adjust slightly over a period of a few minutes. Qualitatively, the pH 2 sample changed color significantly faster than any other sample, and samples with pHs closer to 7 didn’t noticeably experience a rapid color change. This is consistent with the range of hydrogen and hydroxide ion concentration in the test solutions. Any asymmetry about 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.

**Shape as a Function of pH**

**Rate of Change and Degree of Shape Deformation**

Our shape-changing samples were prepared using 4% w/v chitosan powder (Spectrum) dissolved in 3% v/v Acetic Acid (Sigma Aldrich). This sample composition came from preliminary studies of pH-reactivity across different chitosan percentages. A smaller percent of chitosan yielded thinner, fast-acting films, while more chitosan 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. Chitosan samples were cut into 20x7.5 mm strips. pHs of 2, 4, 6, 8, and 10 were tested using the same solutions as before. To create a hinge,
five 0.75 uL droplets of the sample pH solution were applied in a line across the sample’s 7.5 mm centerline. The sample then bent along this line over a period of approximately 10 minutes. Samples were videotaped 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.

Our results reveal a somewhat cubic relationship between bend angle and pH within the range of pH 2-10. Mahdavinia et al. published a similar bimodal relationship, with swelling maxima at pH 3 and 8. Decreased swelling at middle pHs may be attributed to cross-linking [30].

Figure 10. Angle change 7 minutes after droplets are applied to a 4% w/v chitosan film. The points were fitted with a cubic regression line.

Regarding time evolution, the two pH’s corresponding to minima, 4 and 10, decreased in angle between the 4 and 7 minute measurements. 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 more specifically pH-dependent.

Figure 11. 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.

**Odor as a function of pH**

While most materials are engineered and optimized for efficiency and performance purposes, material properties serve as important intersections between human perception and physical systems that guide behavior and experience. To evaluate the detectability and limitations of human perception upon our odor changing primitives, we conducted a user study with 10 participants with four male and six female subjects. We had participants rearrange nine 1”x1” samples of pH activated odor material from pH 2-10, based on odor intensity. We had participants fill a series of questionnaires to collect your perspectives of the material’s emotional quality and descriptive characteristics. We also While you are doing so, we will use the FlexComp Infiniti physiological recording device to record your galvanic skin response, heart rate, and respiration.

Our results show that users can clearly distinguish the on-off switching behavior of the odor material based on pH. pH 9 and 10 were not perceivable and ranked as odors with no smell or low intensity, whereas samples inputted with low pH generated a stronger odor. The user’s sample arrangements for odor intensity correlated with the titration curve and response of the vanillin molecule to pH.

Our odor-changing samples for vanillin 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 solutions as in previous trials: 2, 3, 4, 5, 6, 7, 8, 9, 10.
EVALUATION OF COMPOSITE PRIMITIVES

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

We have decided that materials can be composited in three different ways: they can be combined to form a homogeneous solution before pouring, they can be layered on top of each other, or they can be synthesized side-by-side so only the edges touch. We have tested the full combinatorial range of composites made with three different methods. First, we mixed every combination of two Primitives in their liquid form and poured this into a uniform film (“mixed” composites). Next, we created “layered” samples by pouring one primitive on top of another after the first had dried for 48 hours. Finally, we cut a section out of a sample dried for 48 hours and poured a different primitive in the void (“paneled” composites).

We tested these composites using the same techniques used to characterize the original Primitives, and have used these results to better understand the interactions between the three materials. For example, layered samples involving shape had one of two different pH-dependencies. 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- more quadratic in nature. This is due to buffering effects of the color primitive.

Figure 8. 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.

Figure 13. Examples of composite combinations with most optimal performance based on criteria: (left to right) color-on-odor, shape-in-odor, odor-in-shape, odor-in-color, and mixed color-odor.

A composite was considered successful if it was (i) reactive as intended, (ii) structurally sound, and (iii) reactive within a useable timescale. These materials exhibit different behaviors when patterned by way of layering, paneling, or mixing the Organic Primitives together in various combinations. The composites that satisfy all our parameters included: layering of color-on-odor; the paneling of shape-in-odor, odor-in-shape, odor-in-color; and the mixing of color-odor. Figure 13 depicts the various combinations of primitive composites we determined perform most optimally.
ACTIVATION & CONTROL
All input methods whether they be natural inputs such as the rain or geometrically-defined passive inputs through fluidic logic, can be spatially defined. 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.

Defining Spatial Conditions for Activation

Different deposition techniques can yield specific control parameters for the material system. As shown in figure 9 (top row), 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.

Temporal Sequencing for Physical Animation

Sequential and temporal depositions of pH solutions can yield capabilities for physically animating a material’s output behaviors for color, shape, odor or combinatorial changes. Beyond sequential and temporal methods – form factor, discussed in design space section, can also enable specific physical animation capabilities through relative diffusion rates of non-uniform thickness distributions.

Computational Activation

To demonstrate how material control can be computationally activated, we demonstrate a method to integrate Organic Primitives with a digital system. We developed a computationally controlled system, shown in figure 12, for global and discrete deposition of different pH solutions.

We utilized a microcontroller connected to a series of three hydraulic pumps stationed within disparate vessels of liquid solutions, a pH sensing probe, a microfluidic mixing device, two buttons, and a potentiometer. The potentiometer enables a user to define the specific pH desired for deposition, by rotating the knob manually. Once the user defines the pH value of interest, the hydraulic pumps draw fluids from pH 2 and pH 10 vessels through the microfluidic device as shown in left image of figure 12, where the solutions will be mixed until the user initiated pH value is reached. When the pH value is reached the nozzle deposits it onto an Organic primitive. This system can also be utilized to pattern and fabricate specific input-output regions on an inert substrate by replacing pH solutions with pH-reactive molecules in liquid form.

Coupling and Decoupling pH Input - Output
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 inputs of fluids from the
environment. However, 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 be integrated into the system. More examples of such mechanisms to manipulate material interfaces are described in the Organic Primitives biomaterial building blocks section.

Utilizing Natural, Prepared & Machine-Mediated Inputs

In this section, we discuss pH inputs using natural and prepared acid-base solutions to trigger outputs of an Organic Primitive.

Natural pH inputs

The flow of molecules moderate different pH values in variety of natural processes, from microorganisms to atmospheric conditions. In natural systems, chemical exchanges represent the communication pathways between micro- and macro-scale ecosystems.

On a micro scale, pH fluctuates as dynamically produce byproducts that lend to the synthesis of various acids and bases, among other molecules. Within the human body, the pH of blood maintains an equilibrium of 7.385 – 7.437. The pH for human tears differs from person to person. On average, researchers have found that this is between 6.5 and 7.6 [3]. The pH of sweat is 4 – 7. The pH of (normal) urine is 6 – 7. The pH of saliva is 6.5 – 7.5.

On a macro scale, the pH of ocean is about 8, but is gradually decreasing as the ocean absorbs CO2 emissions. The pH of the rain differs from place to place but within the United States is 5.6 on average, though it can be acidified to < pH 4.3 due to emissions of sulfur dioxide and nitrogen oxide.

pH inputs through daily encounters

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. The pH of food varies across a range of pH 2 – 8. 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.

Prepared pH inputs

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 we have based the concentrations of all our prepared solutions for testing and use. The pH solutions we utilized within our 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.

DESIGN SPACE FOR ORGANIC PRIMITIVES

![Diagram](image)

Figure 15. (left) Different layers of control: from a molecular scale to application level; (right) diagram of interaction loop shows input-output are interchangeable.

The level of control the for the designer has over encoding color, odor, and shape changes in the material system can vary across different scales: first-order logic in functionalization and materialization by selecting the appropriate combinations of sensing and structural molecules; second-order logic in intelligently patterning the dopants on a substrate to generate specific combinatorial outputs of color, odor and shape; third-order logic in compositing the Primitive compounds to yield specific changes based on anticipated conditions; and fourth-order logic of adapting the property-changing materials with specific requirements and additional considerations based on desired applications.

Form Factors

In order to utilize the pH-reactive properties of the molecules, the chemical phenomena must be brought into human-scale interactions. Here, we 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.

Fibers

In figure 8, 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.

Figure 16. (left to right) odor-color changing fiber; color-shape changing films; odor-color-shape changing sheets; liquid form of flavor molecules in pH solutions; odor-color changing gel; odor-color changing 3D form.

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, 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.

Encoding Behavior through Composite Patterning
pH solution inputs and pH responsive molecules can both be used interchangeably.

Pattern through Compositing Organic Primitive

Figure 17. Example of cloaking and disguising information through patterned Odor-Color Organic Primitive
Depending upon how an Organic Primitive is fabricated and the specific location different primitives are patterned, spatial control of inputs and outputs can be interchanged. Figure 14 shows a patterned odor-color primitive utilizing hydrophobic fluids such as olive oil for masking particular areas, enabling a globally activated sample with discrete outputs. This process termed cloaking, can be used to encode hidden information and output potentials.

Pattern Organic Primitives in liquid form

Figure 18. Patterned anthocyanin with pH 9 solution on dried chitosan sheet
pH solutions intended for use to activate the molecules can also be used to spatially specify and define responsive regions in a material.

Fluidic Logic

Figure 19. Different methods to integrate fluidic logic with Organic Primitives: (left to right) closed composite, open composite and molded material logic.
Integrating fluidics with Organic Primitives can enable information processing and activation through passive flow or digital control. In this section we highlight different approaches as shown in figure 18. Specific logic can be incorporated to utilize Organic Primitives for time-telling and titrations.

Ticker for Time-telling

Figure 20. Passive diffusion based time-telling device odor-odor material composite.
Through diffusion rates of Organic Primitives with pH solutions, we developed a material-based “clock” which changes color to mark specific time increments.

Gradiator for Titrations

Figure 21. Fluidic channels molded then flowed through with pH 2 and pH 10 as inputs to generate a titrated gradient

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 in between them. This can be used to create an array of flavor, intensities of color and odor strengths.

APPLICATION SCENARIOS

For each application scenario, we tuned the Organic Primitives by utilizing additives for application-specific parameters with adherence on organic and inorganic objects. In addition, pH states of the material interfaces were developed to achieve application specific starting states and output responses through serial dilutions to alter concentrations in figure 20.

Figure 22. Altering concentration of pH reactive molecules can generate different starting and output states.

Through this molecular scale approach to embedding coupled sensing-actuation to material objects, we explore human interaction with the synthesized biomaterials in four application domains: edibles, cosmetics, environments, and interspecies.

Environment

Figure 23. Acid rain sensing and display utilizing color organic primitive

Acid rain is an outcome of nitric oxide and sulfuric acid interacting with the atmosphere. Fossil fuel combustion from automobile and industrial manufacturing generates chemicals which contribute to rainfall acidity and environmental pollution. Molecules of nitric and sulfuric acid are difficult to detect by the human eye. We present an umbrella as an interface to sense acid rain. Droplets from the environment interact with the molecular sensor-actuators on the umbrella to change color. This provides the user with information from the sky, providing users with a visceral connection and relationship with information within rain water.

Architectural Material to Augment Smell of the Rain

Figure 24. Architectural odor-emitting material

The smell of the rain is often a phenomena romanticized by rural poets and artists. Within an urban environment the smell of the rain is a byproduct of hydration of materials such as asphalt and concrete. In figure 25, we developed an architectural material which outputs odor and color for different spaces for augmenting the smell of different places. These materials can enable architects an additional sense and experience to design with.

Personal Care & Cosmetics

Expressive Cosmetic Display

Figure 25. Interaction prototype of cosmetics displaying hidden information by tuning color-odor primitive with vegetable glycerin.

Human interaction is often mediated by physical appearances, which communicate emotion, identity and character. Cosmetics are a medium, which is commonly utilized to alter and augment human appearance for theatre. In this application scenario, we present how our molecular sensors and actuators can be used as components for cosmetic applications by the molecules with integrating other properties suited for this application – enabling us to use data exerted from human body fluids. Depending upon the area, which they determine to be applied, the material
can sense and output based on natural inputs defined by creation of body fluid. The molecule can change color and release pleasant odor when user exerts a lot of sweat from physical activity as pH lowers from the lactic acid produced from their skin microbiome. This can augment dance performances to engage viewers in the dancer’s physical exertion.

**Saliva sensing for oral health**

pH is an important factor for maintaining oral health as certain bacteria contribute to fluctuations. pH responsive materials can be integrated with the bristles of a toothbrush to sense and survey the pH of a user’s oral environment.

**Edible**

We explore user scenarios in the context of an eating experience.

**Shape Programmable Pasta**

Chitosan biopolymer derived from crustacean shells have been thoroughly examined for use in food and biomedical drug delivery applications. In this example, we propose a dynamic food which enables chefs to program shape for different levels of flavor retention and sensory augmentation in food. Dynamic food can be developed by patterning on pasta to define specific shape deformations, used as on-plate communication and embellishments, utilize condiments as input for simultaneous control of flavor, texture, and appearance. Chefs or users can encode pasta shape through pH for pasta sauce retention or to enhance user experience. 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 the cooking and preparation process, 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.

**Apple Sensor-Display**

An apple can be transformed into a display by tuning patterning substrate of color primitives with methylcellulose MethocelF50 on edible starch paper. This can enable an apple to display information about itself such as – where to cut for kids; recipe information (ie. apple pie); and more advanced application for sensing environmental contaminants during transport for food safety.

**Biodegradable Smart Utensils**

We explored multiple methods of creating “smart” utensils that will react to their environment. One method involves mixing kappa carrageenan and anthocyanin with 250 bloom fish gelatin. The gelatin adds more structural integrity to the material. We tried various ratios of concentrations of structural materials (either gelatin or pectin) mixed with kappa carrageenan. We found that a ratio of 1:3 of gelatin to kappa carrageenan worked best for the desired application of the utensil. The pectin was very acidic so the materials were pink. The gelatin did not affect the color of the materials and the ratio of 1:3 allowed for the material to have additional structural integrity while maintaining the color-changing properties of kappa carrageenan with anthocyanin.
(top left to right): 250 Bloom Fish Gelatin mixed with 2% kappa carrageenan with 0.2% anthocyanin in ratios of 3:1, 2:1, 1:1, 1:2, 1:3

(bottom left to right): Rapid Set High Methoxyl Pectin mixed with 2% kappa carrageenan with 0.2% anthocyanin in ratios of 3:1, 2:1, 1:1, 1:2, 1:3

We created a 3D-printed mold which the biopolymers may be cast into. Another method is to composite color-, smell-, or shape-changing primitives on top of gelatin sheets which may be used as the skeletal, structural base. A third method allows the integration of microfluidic logic into the utensils. This method involves 3D-printed utensils with embedded channels that are then dipped in the biomaterials.

Modular utensils also have the potential to augment the user’s experience with food. A smell-changing fork may simulate the experience of eating a sweet food without the addition of any sugar. A utensil with embedded microfluidic logic may allow a user know how long they’ve been eating based on how far liquid has travelled in the channel. This information may then be used to give a user insight into how they can pace their eating.

The organic primitives are particularly intriguing for this application since they are food-safe and also biodegradable.

Interspecies
Augmenting Plant Aesthetics & Fragrance

Figure 29. Example of color changing flower and leaf.

Gardens of flowers are used to often enhance the aesthetic environment. Gardeners often monitor and alter the pH of plants to manipulate the way they grow. In this scenario we show how altering the pH of plant organs can result in different microbial community formation to foster human-plant and human-microbial interactions when users water the plant using different pH solutions. Research in microbiology also suggests this can be used to affect pollination.

Bionic Fruit for Fermentation

Figure 30. Bionic fruit "prints" edible sensors and actuators

Fructus is a bionic fruit which electromechanically regenerates skins of biomaterial capable of attracting microbes in the environment for fermentation. We are interested in providing chefs a tool to communicate to their guests by remotely triggering Fructus to synthesize different flavors, textures, and tastes in the form of dynamic edible films throughout the course of a meal. As these materials are generated using food-grade ingredients, they can also be used to teach chemistry, biology, and food science to kids.

DISCUSSION AND FUTURE WORK

Through this molecular scale approach to embedding coupled sensing-actuation to materials and objects, we uncover some novel properties and design principles. Since pH responsive molecules can function as integrated sensor-actuator-energy source, these components can all be used interchangeably within the system (figure 6). pH solutions can also become outputs when using liquid-form pH molecules as inputs. Likewise, liquid-form anthocyanin can become an “energy source” or trigger for the acid-base reaction on a pH-patterned chitosan sheet. Tools developed for activating the material can also be used for fabricating the material.

The primary factors that yield the rates of change in material property of our materials are based on diffusion rate, buffering capacity and the thickness of the material sample, relative to the concentration of the pH stimuli. By tuning the relative concentrations of each parameter, we can
synthesize biomaterials to respond to pH stimuli under different rates, output modalities, and transition states.

Towards Multiple-Inheritances of Material Properties
Material primitives can be combined in a number of ways to generate desired output combinations whether polymodal combinations such as color-odor or trimodal combinations with shape, odor and color. These properties can be programmed in a number of different combinations, enabling materials to “inherit” certain properties when utilized in combination, similar to genetic traits.

Characterization of reversibility behavior
The degradation life and extent of the material primitive’s capability for reversibility can be tuned based on the specific input and application scenarios. These need to be experimentally examined further.

Offloading Material Design Parameters to Software Tools
Data generated from our material characterization and evaluation serve as our design parameters in our material system. 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 Molecular Sensing Capabilities
While pH is the starting point for input, other molecules can be sensed and utilized as input such as glucose, eH, pCO2, sodium, magnesium, calcium, potassium, dioxins, or even specific hormones. Stimuli can also be physical as opposed to chemical such as temperature or pressure based. The materials can serve as an output platform whereby a sensing molecule is utilized with a pH-reactive organic primitive or it is synthesized anew with our library of biopolymers. Through expansion of sensing capabilities by the testing and identification of additional dopants for input-output, it is possible to synthesize a material which can sense multiple inputs and generate combinatorial outputs based on the information sensed. Additional dopants can be used to expand capabilities for current pH-reactive Organic Primitives or be used towards the development of new primitives with wholly new input output dynamics.

Faster Flows
Relying on diffusion kinetics for material change, the output is typically slow due to the diffusion rates for water. Future work may involve the use of volatile organic compounds (VOCs) as activation mechanisms where gas is the main fluid as opposed to water.

Integrated Organic Primitives & Logic
While only small examples were demonstrated in the application scenarios, circuits can be built using the organic primitives, enabling complexity and logic to be expanded upon.

CONCLUSION
pH can give information and cause change in an astoundingly broad range of systems, from the simplest organic material to the entire global ecosystem. It is an important health indicator of many complex chemical processes within human body, and it is a factor in every material that interacts with humans – from food to personal care. Beyond the personal scale, pH serves as an important environmental indicator, critical to growing pollution concerns in ocean acidification and atmospheric contamination. For this reason, we selected pH-reactive materials to develop into fully functional sensors and actuators.

In this paper, we developed a process for the synthesis of material sensor-actuators, which can be tuned and reconfigured to develop higher order complexity for molecular interface design. We demonstrate through the synthesis of Organic Primitives the possibility for materials with multiple inheritances and functional structures. Through this work, we hope to inspire researchers to develop interfaces that connect the every-day organic objects with the physical chemistries within our environment, our bodies, and the edible materials we consume.

While pH serves as a starting point in this body of work, we hope that the concept of Organic Primitives may be applied to many other sets of stimuli and sensors. The ultimate goal of this project is to develop a full library of Organic Primitives, so that scientists, engineers, students, designers, and hobbyists alike will be able to develop objects that interact with their environment in a direct and natural way.

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