Research Article



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Abstract

PolyTile 2.0 interrogates the potential of programmable biofunctionalities in our constructed architectural environments through the development of advanced ceramic bio-tiles. These tiles utilize novel patterning techniques and hydrogel biomaterials to tune surface conditions at the micro- and macroscale. This trans-disciplinary work builds upon recent advancements in the fields of three-dimensional printing, digital ceramics, materials science, bioengineering, chemical biology, and architecture. PolyTile 2.0 enables designers and architects to implement biofunctionality and microscale patterning fittingly and with the ability to continuously adjust design iterations across scales. The refinement utilizes glazing strategies as a directable fluidic device and biocompatible hydrogels as a sensing platform to further developments in responsive built environments. This article outlines methods for the production of bulk-scale hydrogel materials, stereolithography-based three-dimensional printed ceramic tiles, and scalable glazing techniques, which bring building-scale application of this technology to the foreground.

Keywords

Three-dimensional printing, hydrogel materials, digital ceramics, biofunctionality, advanced composite materials, responsive architecture

Background

PolyTile 2.0 looks at applying bioengineering innovations to the building scale by developing a ceramic tile that can be fabricated with micron-scale precision, yet a useful size for architectural projects. The field of three-dimensional (3D) printed digital ceramics emerged in the past decade thanks to the advent of

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Figure 1. PolyBrick series.

From left to right: PolyBrick 1.0 interlocking brick, PolyBrick 2.0 component based on human bone formation, and PolyBrick 3.0 iteration to interface with DNA hydrogel (15-mm diameter). PolyBrick 1.0 and 2.0 iterations are scaled-down versions as shown and all prototypes used the stereolithography technology for printing.

powder-based additive manufacturing. Since 2009, the Sabin Lab, and Sabin + Jones LabStudio before it, has been building a body of ceramic work, focused on the development of nonstandard bricks, tiles, and aggregated assemblies. The PolyBrick series, previously exhibited at the Centre Pompidou in Paris (Imprimer Le Monde 2017) and the Cooper Hewitt Smithsonian Design Museum (Beauty 2016) brought to light questions around ceramic construction and full-scale fabrication through digitally steered material and process-based investigations within the Sabin Lab at Cornell University. PolyBrick 1.0 iteratively investigated the production of nonstandard ceramic brick components with designs for mortarless assembly and interlocking joinery based on traditional wood construction.¹ The next generation of PolyBrick 2.0 advanced computational methodologies in ceramic construction through application of behaviors and principles based on human bone formation to finite element analysis models. By guiding load paths and developing a series of structural rules, the tests generated a controlled gradient of dense structure when needed and light, porous geometries in areas demanding less load.² PolyBrick 3.0 looked at advanced DNA hydrogel research as a source for the development of intelligent and encoded bio-bricks and tiles. Using clay as a substrate for DNA glazing, this series highlighted living signatures on ceramic surfaces, which could facilitate data storage in and around our constructed environments.³ Designed in collaboration with biological engineers in Luo Labs at Cornell University, this research proved the compatibility with clay as a material host for life and DNA hydrogel materials.⁴ Most recently, the PolyTile series began investigating the development of biofunctional elements within our hybrid tiles. Previous work that utilizes the combination of hydrogels with ceramics mainly resides in bone and tissue engineering research. Hydrogels and ceramics are used as scaffolds to regenerate bone/ tissue through stem cell growth,⁵ composite hydrogel/ceramic (often hydroxyapatite) micellular aqueous solutions used for bone grafting,⁶ and hydrogels have been heavily used in the robocasting of ceramic parts.⁷ In continuing collaboration with Luo Labs, we refined technologies critical to digital fabrication of 3D printed ceramics and bioengineering. This included a microscale method for spatially controlling DNA materials and developing hydrogels with bespoke functionalities (see Figure 1).

Introduction

PolyTile 2.0 translates research within the subjects of hydrogels and 3D printed ceramics into a microtextured architectural tile series with stimulus response capacity, which aims to imagine future application at full building scale. Whereas PolyTile 1.0 primarily depended upon 15-mm disks for prototyping due to the time

and cost intensive process of producing protein-expressing hydrogels, this new investigation pushes geometries, materials, design, and larger-scope architectural parameters into its fabrication. Fluidic behaviors and knowledge gleaned from PolyBrick H2.0 contribute to a dynamic design approach related to surface patterning.⁸ By developing bulk-scale hydrogel materials (producing 100+ mL volumes inexpensively) and opportunistically investigating functionality across a range of these hydrogels, it became possible to design component-based, aggregated tiles suited to answer independent inquiries. PolyTile 2.0 uses pattern design and fine-tuned pixel geometries as a hydrogel-functionalization driver. Patterning and surface conditions (including glaze techniques) directly influence the direction of material flow, hydrophobic/hydrophilic response, and light responsiveness.

Through expanding to more adaptable hydrogels, such as poly(ethylene glycol) (PEG) and PEGcomposites, from the previous DNA hydrogels used in PolyBrick 3.0 and PolyTile 1.0 such as Meta P-gel,⁴ we can use PolyTile 2.0 to expand the breadth, implementation, and approachability of biofunctionality within the context of architectural environments. Hydrogels are often biocompatible^{9–12} and are thus suitable for architectural use due to its engagement with the human body. Extensive research has gone into inventing strong and resilient hydrogels with different monomers, initiators, and cross-linkers.^{8,9} Various research groups are synthesizing these distinct hydrogels through introducing functional groups with different electroactivities, bonding interactions, chemical and physical properties, coordination, and responses to external stimuli. Precedents have shown response to pH, glucose, oxidants, antigens, enzymes, ligands, temperature, pressure, and light. Although hydrogels have mostly been seen used in drug-delivery and biodevices, their versatility and tailorability show hydrogel-ceramic composites as having the capacity to respond at humanscale. The PolyTile 2.0 series encapsulates the development of numerous functionalized ceramic-hydrogel materials.

Coupled with ceramic substrates, biological matter in the form of biofunctional hydrogels exemplifies the future of ceramic materials research. Building upon DNA-glazing strategies from PolyTile 1.0 and micro-textured surface research from PolyBrick H2.0, this article explores dynamic glazing strategies that exhibit novel environmental responsiveness and digitally designed patterning techniques. PolyTile 2.0 pushes func-tionalities beyond previous iterations by challenging design questions at building scale and testing bulk-scale hydrogel materials for varying types of environmental responsiveness. This article looks into numerous formal tile-printing techniques, expanding scales, parameters dealing with nuances in patterning, and hydrogel responsiveness based upon these changes in the ceramic substrate's surface conditions. In interrogating the movement and application of gel or other fluidic material on numerous traditionally or novel glazed surfaces, this article achieves unprecedented control and refinement of ceramic parts, giving way to a new scope of research on the subject of living matter and architectural ceramics.

Materials + technology

PolyTile 2.0 functions via the foundation of three distinct technologies and fields of study: 3D printing, ceramics, and bioengineering. Ceramic as a time-tested and biologically compatible material creates a physical platform from which to build upon. Three-dimensional printing transforms a generally plastic material (clay with "plastic" referring to elasticity) into a highly precise and tunable one, with digital tooling and modeling of design details to the micron scale. Bioengineering in conjunction with traditional glazing and modern material science showcase novel applications of responsive and encoded functionalities.

Ceramic materials undergo three distinct phases: greenware, bisque firing, and glaze firing. Greenware describes any ceramic material or form prior to firing in a kiln. While in the greenware state, other materials can be added into the clay recipe.¹ In the context of contemporary 3D printing techniques, additives are often part of a clay body, with the expectation that these additives are burned-out during bisque firing (the first firing). Stereolithography-based printing requires a resin matrix, which cures under UV light within the 3D

printer. Clay particles are suspended within a specially formulated resin-ceramic matrix to produce a ceramic resin. When in the greenware state, the resin's UV curing allows forms to be fixed whereas only the ceramic remains later in the process. Bisque-ware describes the state of a ceramic part after the first firing at a lower temperature, whereas glaze-fired parts undergo two firings typically and are often glazed as well.

Early examples of ceramic 3D printing depended upon binder-jetting powder machines such as Z-Corporation (ZCorp) printers, which used a glue or alcohol-based solution to fuse clay powder to their corresponding layers and geometries. This technology was closely whetted to laser printing, as the early machines used laser printer cartridges to dispel binder onto the powder bed. Printed parts often benefited from powder beds or chambers, in that the parts required no support material. While the resolution was tunable to millimeter-scale details and available bed sizes were large, these machines did not allow for scalar control appropriate for bioengineering-focused investigations.

Stereolithography printing, however, allows for increased scale of printable parts, while also improving resolution. Autodesk's Ember stereolithography printer brought to light further potentials for 3D printing research thanks to the reported control of print quality down to 10 microns in the Z-axis. The build plate of these machines was limiting (60 mm \times 34 mm on the XY plane), making the machine challenging for projects that envisioned full architectural scale production.¹³ With Formlabs Form 2 Printer and their Ceramic Resin developed in-house, printable scale increased dramatically and reliability of these prints improved. As of 2020, the Form 3 L by Formlabs presents the most potential for the continuation of biologically steered ceramic research at the architectural scale, though proven compatibility with ceramic resin has not yet been confirmed.

Methods

The ceramic tile serves as the interface to extend the functionalities of hydrogel research into a material fit for the built environment. Current developments in PolyTile 2.0 specifically utilize the PEG hydrogel and three glazing types as a fluidic device for gel-functioning the ceramic tile.

Fluidics

Ceramic porosity on the macroscale and microscale provides a scaffolding to form the ceramic-hydrogel composite structure.⁴ Because glaze chemistry crucially determines the formation of the composite, it serves as a limit on manufacturability and scalability. PolyBrick H2.0¹⁴ investigated glaze chemistries and microtexturing to successfully control the flow of water across a ceramic surface. With the addition of a fluidic system based on these principles and its transferability to PolyTile's hydrogel presolution, the manufacturing process becomes increasingly automatic and scalable. For application, the hydrogels are washed over the tiles in a water-based solution and since hydrogels, generally, are greater than 90-wt.% water,⁹ the work from PolyBrick H2.0 translates opportunely to allow for control of hydrogel placement. PolyBrick H2.0 was highly dynamic in that it controlled the flow and directionality of water in the x-, y-, and z-directions as well as against gravitational force. This specificity is highly desirable as it is necessary for addressing the optimization problems that will arise as the designs are implemented for the local environment.

PolyBrick H2.0 gave us three variations of glazes with a range of hydrophobic/hydrophilic behaviors that are used for PolyTile 2.0. The glazes are Opulence 346 White Glaze (hydrophilic), Opulence 346 White Glaze + ZnO (superhydrophilic), and NanoSlic[®] NS-110 coating (hydrophobic and oleophobic). The ZnO was acquired from Pure Organics and is of non-nano, uncoated grade. The glaze is applied through a sponging and brush-on procedure. For the white glaze and white + ZnO glaze, the bisque-fired (at cone 06, 1828°F) clean ceramic surface was cooled and submerged in water for 2–3 s. Brushing over the piece with a water-soaked brush is also viable. It is air dried for another 2 s. The glaze is lightly soaked onto a sponge and applied



Figure 2. Glaze types.

This figure shows a sample piece with three different glazes, White Glaze, White + ZnO, NS-110, from top to bottom, respectively.

to the surface and repeated to ensure sufficient glaze while preserving texturing. The recipe consists of a 7:3 ratio white glaze to water. The ZnO glaze is 4:3:4 white glaze to ZnO powder to water. The ceramic piece is then glaze fired at cone 4 (2086°F). Pieces were fired on the slow setting to prevent cracking and set directly on the kiln shelf. The NS-110 coat is applied after the ceramic is bisque fired (cone 06) and glaze fired (cone 4). In total, 2 mL of NS-110 is added for every square centimeter of surface. It is then air dried overnight to harden to 9 H (hardness) (see Figure 2).

Glaze may be applied at the discretion of the designer. This study often utilized glaze scraping to maintain pattern resolution. Regions intended for hydrogel application were extruded from the surface. Glazes were flooded onto the surface, and the extrusions scraped with a blade revealing unglazed, hydrogel-accepting areas. The combinations of the varying properties that come with surface texturing (such as intrusions, extrusions, curvatures, etc.), texture sizing, and glazing provide a fluidic system to create a designer-specific hydrogel-ceramic tile (see Figure 3).

Hydrogel

Extending from the DNA hydrogel considered in PolyTile 1.0, PolyTile 2.0 looked at a similarly functional hydrogel that has a more accessible preparation protocol. The transition to a PEG hydrogel aimed to expand the prospects of ceramic-hydrogel composites as a general category, which would open the conversation to the breadth of functionalized hydrogel research already existing as well as research yet to be developed across disciplines. Furthermore, this transition would advance preparation methodologies suitable for large-scale hydrogel production.

The PEG hydrogel synthesized for use in this experiment has basis in Zhang et al.'s¹⁵ procedure. However, the poly(ethylene glycol) diacrylate (PEG-DA), 2-hydroxy-4'-(1-hydroxyethoxy)-2-methyl (i2959), and



Figure 3. Glazing method.

water which forms the presolution for hydrogel synthesis were mixed at a ratio of 1 mL of PEG-DA (molecular weight 700) to 0.1 g of i2959 to 3 mL of water. In addition, for every 1 mL of PEG-DA, 1 drop of GLO Effex concentrated UV water dye additive was added to the solution. The solution is UV-cured overnight (average cure time of 15 h) in a sterilizer to crosslink and form the hydrogel. Depending on the wavelength and intensity of UV light, the cure time may vary. A UV-cured hydrogel allows for more intensive testing with ceramic porosity and the glazes as its behavior is similar to water until it is ready to be UV cured. It also provides greater control during the gelation process since it gives the designer freedom to

revise and edit even after the ceramic piece gets washed with the pre-hydrogel solution. Rapid photopolymerization is readily achieved through curing in stronger UV ovens, using a more concentrated photocrosslinker, and/or more easily radicalized photoinitiator.

Texturing

Informed by developments from PolyTile 1.0 and PolyBrick H2.0, the breadth of texturing negotiated fineness of 3D print resolution, visible patterning, fluidic functionality, hydrogel compatibility, and material feasibility as a few primary drivers. Due to water properties such as surface tension, polarity, and capillary action, double-digit and triple-digit micron-sized patterning affects water flow most significantly, considering the range of contact angles water makes with such textured surfaces when under material controls.⁹

Texturing is influential where the pre-hydrogel solution can spread to, but regarding its specific use in hydrogel synthesis, it provides the framework necessary for the global form of cured hydrogels. There are advances in hydrogel "inks" and 3D printed hydrogels, but for widespread and large-scale use, hydrogels customarily receive their cured geometrical structures entirely in the mold it is cured in. Tile functionality utilizes texturing for selective hydrogel distribution and as a means to realize various geometrical forms.

3D Printing + fabrication

The production of 3D printed ceramic tiles utilized Formlabs Form 2 printer and its compatible Ceramic Resin.¹⁶ As mentioned in the PolyTile 1.0 paper, other ceramic resins such as Porcelite were found to be less suitable in stereolithography machines due to lesser density of the clay body after firing.¹⁷ The Form 2 printer has a build size of 145 mm \times 145 mm \times 175 mm. As a result, the determined scale for full-size, aggregable parts was 127 mm \times 127 mm, allowing for space within the bounding condition and with an interest of reducing potential errors around the build plate.

Full-scale tiles were first printed flat and parallel to the surface of the build plate. The aim was to minimize print time and reduce the necessary amount of support material. This printing process involved trials with no rafts, mini rafts, and with a full surface raft as well as other parameters such as touchpoint size, density, raft thickness, and heights.

Improving upon this technique (see "Results" section), the next sequence of tiles was printed at 12° , 22.5° , 30° , 45° , and 60° angles with varying success. Tiles parallel to the surface of the build plate caused jamming errors in the Form 2 Printer, as the friction created from surface adhesion was too great during the course of printing. By increasing the angle in both X- and Y-axis (therefore increasing height in the Z-axis), more support material was added, though it results in less surface area on each sliced layer, which produced improved prints with greater reliability.

With reference to tile geometries, a selection of $127 \text{ mm} \times 127 \text{ mm}$ tiles was printed with varying patterns and textures to evaluate process feasibility. Parameters varied and testing included patterned extrusions and intrusions of varying dimensions, geometries, densities, curvatures, and Fourier frequency-based digital visualization (see Appendix). These variable experiments helped to further understand ceramic curling behavior, appropriate glazing strategies, size and shape in which to induce specific hydrogel behavior, geometrically induced light perception, and control over the piece. Experimentation included tests to investigate the following:

- 1. Global geometries of the tile in relation to tile translucency, glazing options, curling, contribution to fluidics, firing ability, and structural integrity;
- 2. Texture and microtexture morphology, dimension, and placement for feasibility of glaze removal, gelling ability and behavior, and fluorescent behavior;
- 3. Glazing for fluidic response analysis and hydrogel scaffolding;

- 4. Methods to wash the tile with the hydrogel presolution, and with what quantity, for varied effects such as submerged bathing, poured wash, or more localized washing using droppers and pipettes;
- 5. Procedures to create consistent outcomes.

Please refer to the Appendix for detailed procedures of the tile variations used for testing.

Functionality investigation: light response

The UV fluorescent PEG hydrogel developed through the use of dye additives functionalized the tiles to take in short, medically harmful, UV wavelengths and emit the longer visible wavelengths. Furthermore, the developments in PolyTile allow for the design of ceramic-hydrogel surface distribution, hydrogel sizing, hydrogel location, and hydrogel geometry. This manipulability permits the launch into the first functional ceramic-hydrogel response system in which PolyTile utilizes both the light radiating from the environment and the spatial positioning of the observer.

Fluorescence has important biofunctionality in humans and other organisms, mainly serving as signals. Humans are 50 times more sensitive to green/yellow wavelengths making us good fluorescence detectors, and several animals, such as birds, possess corneal or lens filters in their eyes that sharpen their photoreceptors for detecting fluorescent emissions. Fluorescence changes have been shown to change behavior in some organisms.¹⁸ The Fourier raster patterns used in this study were mathematically generated to register a shift in light and our perception of it.

Results

A suite of full-scale tiles is produced, which demonstrates the combinations of hydrogels embedded with functional behavior. The array of size, distribution, consistency, smoothness, and so on are readily programmed onto the designed tiles.

Luminescence

The PEG hydrogel is functionalized for UV fluorescence through a physical, rather than chemical, bonding between fluorescent dye and polymer network. Since the UV fluorescent dye additive is water soluble and polar, this was advantageous for the PEG hydrogel to encapsulate the dye in its physical network when UV cured (see Figure 4).

Testing was done to ensure that the network capture of dye in the hydrogel was able to render it a UV fluorescent, biocompatible hydrogel. The dye used in this specific case is a green fluorescent dye that is neon yellow in the visible light spectrum (several other colors are also offered). When the fully synthesized hydrogel is radiated with UV light, it is being indicated. When the dye is UV cured for the full duration



Figure 4. PEG-hydrogel molecular formula.





Two ceramic test pieces are shown in (a–c). The tile on the left was washed with a hydrogel presolution with fluorescent neon green dye and the one on the right with only the dye in the same ratio of water as the presolution (a). Both pieces were UV cured to observe its fluorescence. In (b), the ceramic pieces were cured for less than the needed cross-linking time to form the hydrogel. It was cured for 6 h and both pieces showed fluorescence—the dye is the cause of this behavior. After the full cross-linking period was completed as shown in (c), only the piece that formed a hydrogel showed fluorescent behavior. The piece with only dye loses its fluorescence as its chromophores degrade. A second test uses two textured pieces. Both pieces were white glazed on its surface other than the square pixels (d). The one with two squares was washed with the fluorescent pre-hydrogel solution (i) and the one square tile with just dye (ii). Both were cured overnight to give sufficient time for cross-linking.

needed to cure the hydrogel, the visible color disappears, and therefore looks clear in visible light. When the dye, however, is encapsulated and protected within the hydrogel network, it retains its fluorescence during the time frame that the hydrogel is cured (see Figure 5).

In addition, microscopy results show the gel, gelled ceramic, and ceramic itself to be distinguishable (see Figure 6). Hydrogels have high refractive properties and fluoresce differently than a dyed ceramic piece.

The ability for PolyTile to be UV fluorescent established a programmable, functionalized architectural tile that responds to radiation and alters the perception of light.



Figure 6. Morphology of a hydrogel-ceramic composite.

For visualization purposes, a visible green dye was used for the demonstration. On the top is a ceramic piece with no glaze resist but glaze fired with a green dye in water solution washed over it and UV cured. On the bottom is a similar piece that is washed with a dyed hydrogel presolution and UV cured.

Building-scale tiles

The set of 127-mm square tiles was functionalized with the UV fluorescent hydrogel material. A critical selection of the aforementioned ceramic tiles is presented to exhibit glazing, texturing, and hydrogel fluorescence.

Glaze-fired regions, which are not glazed, absorb the pre-hydrogel solution and upon gelation, hydrogel forms within the ceramic structure. White glaze resists gelation, but due to its hydrophilicity can allow for hydrogel formation on the ceramic surface as a result of pooling. The ZnO-based glaze is superhydrophilic and while it resists the gel from absorbing into the ceramic piece, it holds tightly to the pre-hydrogel solution that forms a layer of microscale hydrogels on its surface. NanoSlic can be used in two ways. When pre-hydrogel solution is applied to a NanoSlic-treated piece as a wash, the glaze resists hydrogel in the ceramic piece as well as on its surface. Used in conjunction with a microtextured ceramic piece, the NanoSlic-treated regions can form extruded hydrogels from the



Figure 7. Resolution of hydrogel-glazed tile.





A spherical ceramic extrusion on the tile surface was glazed with ZnO + white glaze and resulted in hydrogel pills forming on top of the sphere surface after curing.

surface. It may form hydrogels that form on top of the surface, but unlike the ZnO glaze, the hydrogels are macroscale and may be very smooth. Smooth hydrogels have high refractive properties. The spectrum of hydrogel geometries, sizing, placement, texturing, and so on that can be formed is reliant mainly on the behavior induced through glazing (with help from ceramic texturing). This variation permits the opening of investigations of hydrogel-ceramic composites as sensors and responders to environmental conditions.

The resolution of the hydrogel is seen to be directly affected by texturing and glazing resolution. Through the "JSLab" tile, the letter is unglazed and gelled, while the rest of the tile has a white glaze resist. The hydrogel, seen through its fluorescence, can be seen to fall within the lines of the lettering (see Figure 7).

Tiles with spherically extruded pixels and ZnO glaze coalesced the hydrogel toward the pixels and allowed it to form as pills attached to the glazed surface (see Figure 8).

Similar texturing but with white glaze resisted any hydrogel from forming (see Figure 9).

The small-scale texture sizing and geometry may be configured to increase or decrease the influence of the water-based solution's high surface tension through principles based in capillary action. Tight pixelation not



Figure 9. Effects of ZnO + white glaze versus white glaze.

The figure on the top represents the cured textured tile with white glaze in the top left and bottom right quadrants and ZnO + white glaze on the other. Image on the bottom shows the same tile under UV light. White glaze resists hydrogel formation, while ZnO + white glaze attracts gel formation. The amount and type of gel formation on the ZnO glaze quadrants also vary depending on the size and distance between the texturing.



Figure 10. Z-directional flow.

The above tile was fully coated with a thick ZnO glaze and glaze fired. After cooling, the tile was submerged with fluorescent hydrogel presolution at level with its surface height. The glaze pulled the solution into the center of the tile as well as up the textured spheres. From the tile's design, larger spheres had tighter spacing between them, increasing their density, than smaller spheres. Tight spacing resulted in greater capillary action which, after UV curing, produced greater hydrogel density. Dense texturing directly correlated to dense gelling behavior and increased gelling in the z-direction.

only generates pooling or directed flow on the surface of the tile (x- and y-direction), but it also causes the upward climb (z-direction) of the hydrogel solution (see Figure 10).

Adding curvature or altering the global geometry of the tile will influence gel formation expectedly through use of gravitational force. When glazing a textured surface, scraping pixels provided a means to have highly resolved and small unglazed areas.⁴ However, due to the low viscosity of the pre-hydrogel



Figure 11. Tile with multiple examples of programs.

The above 127 mm \times 127 mm composite tile contains 25 subdivisions of designed programs. It provides an example of the programmability of the hydrogel and the methods that provide the designer with the discretion to curate tile functionality.



Figure 12. Composite tile patterning demonstrating glaze-based crazing.

solution, texturing formed channel walls that affected tile fluidics along with its glaze chemistry. Extruded and intruded texturing formed the topography that induces pre-hydrogel flow. These channels guided the water-based solution directionality, eventually informing the location, geometry, and properties of the formed hydrogel (see Figure 11).

The macro-/micro-texturing and glaze chemistry presented here are integrated as design drivers with ceramic behaviors. For instance, an extruded spherical texturing gradient was glazed with ZnO glaze and through ceramic property changes due to varying time and shape patterning, a crazing pattern developed in which a distinctive hydrogel pattern is observed (see Figure 12).

Further results of varying the parameters mentioned in testing can be viewed in Figure 13. For detailed inquiry into the glazes used and surface micro-texturing, please refer to PolyBrick H2.0.¹⁴

Further hydrogel functionalization can be achieved through applications of PolyTile 2.0 with established techniques used in ceramic craftsmanship.

Functionality investigation: light response

Light patterns naturally evolved from the channeling created through glaze and texture-based fluidics. The techniques elucidated here allow for the refinement of patterning for the expression and suggestion of light.

	White Glaze	ZnO Gla	ZnO Glaze		NanoSlic Glaze			No Glaze	
Gelation	No Gelation		Gelation		No Gelatio	on Surface Gelation		Gelation	
Extruded Behavior	Pilling behavi (full wash or localized droppered) (Fig. 10, Fig. 11, F	Can fo	Can form smooth, very sphe (droppered)			al gels too (ful		Smooth Hydrogels II wash or localized washing or droppered) (Fig. 14)	
Hydro -phobic/philic	High capillary (Fig. 10)	+ w crea	+ with high density texturin creased coverage of surfac that's glazed (Fig. 10)			High hydrophobicity - repulsive force (Fig. 11)			
Fluorescence	Brighter fluorescence for	Bright fluorecence for NS surface hydrogels (Fig. 14)							
Gel Distinction	Clean lines from scrap- ing macroscale (local- ized poured wash) (Fig. 7)	Clean gel (Fig.	distinction 11)	Clean gel distinction fo intrusions (droppered (Fig. 14, Fig. 15)		No clean gel distin for surface gelati (washed) (Fig. 1		nction tion 11)	Distinction depends*
Microtextured Effects	When microtextured, can form macroscale, unsmooth spherical gels (droppered)	When micr high z-d gela (Fig. 9 a	When microtextured, high z-direction gelation (Fig. 9 and 10)		Can form smooth, very spherical gels too when microtextured (drop- pered) (Fig. 15)		Can form smooth surface gels conform to surface when mic textured (washed		When microtextured (intrusions), only gelation in wells
Effects of Curvature	Denser gel formation in dip of curve; more prominent for smaller texturing over larger				Curvature: little to no effect			Curvature affects gelation expectedly. Dips increased gel density. High steepness lowered gel density. (Fig. 11)	
* microtextured tiles often did not produce as clear distinct textures from scrape method—when extrusions too short, there is difficulty scraping as glaze would cover too much; when too much height sometimes may pool too much glaze and a large chunk would scrape off in addition to the extruded texture; (Fig. 11)									

Figure 13. Tile variation: results of varying parameters.

Each row represents a distinct behavior exhibited based on each glaze type, listed as the columns (four glaze types color coded). If a glaze type (column) has a corresponding behavior (row), a description is listed.





The progression began with (a) and (d) in which channels were designed using equally spaced extrusions. Light perception changed from glazing each channel with NanoSlic, no glaze, and ZnO in triplets. This initiated the frequency-dependent light perception study. (b) and (e) is the next tile in the progression and fluoresces according to Fourier transform data.¹⁹ The last in the progression, (c) and (f), reflects a more complex Fourier transform data set.

Three-dimensional printing a Fourier transform frequency raster texture on a tile translated the logic of hydrogel fluidics onto light perception.¹⁹ The UV fluorescent hydrogel directly is able to functionalize a ceramic tile to interpret data from other sources such as those captured through Fourier transform (see Figure 14).

Micro-texturing and glazing also allows for the synthesis of smooth microdroplets of hydrogel material. Attachment of this size and distribution of hydrogel droplets onto a ceramic surface provides added light responsiveness. Here, physical distortion of light is achieved using an unfunctionalized hydrogel (see Figure 15).

3D Printing + fabrication

Printing parameters affected tile translucency, curling, firing ability, and structural integrity. In the case of both mini rafts and no raft printing, the column-like support network failed to adhere sufficiently to the build plate surface. Rafts refer to solid flat surfaces of cured 3D printed material, which are used to build structures of support material. Rafts also helped to address imperfect tolerances and inconsistencies on the metal build plate. While some of the support network would remain intact, the absence of other regions would create problematic areas and structure of the printed part. Furthermore, the remnant pieces of these supports would cause disruptions in the printing process by jamming the printer due to foreign bodies falling in the ceramic resin tray (where the resin is located) or result in misaligned printing. The full surface raft adequately printed desired building-scale tiles, but there were some issues. The larger raft is wasteful from the standpoint of material usage. Prints as a result were assigned a 1-mm thick raft to provide ample support, while reducing resin usage. The angled prints (no longer parallel) had far longer predicted print times (16 h on average); however, the unparalleled reliability was found to be more desirable and significantly expedited the excavation process (see Figure 16).

In printing steeply angled pieces, increased forces on the latter layers produced separation of the tiles from their support material. Additional strain from the support columns often led to macroscopic cracks horizontally along the tile. The structure of these parts was weakest in the final layers. To resolve this, printing at 22.5°



Figure 15. Ceramic pieces with smooth hydrogel microdroplets. The piece has intruded hemispheres with a diameter gradient from 600 microns to 1.159 mm.



Figure 16. 3D printing support network: raft types.

balanced the desire to lessen surface adhesion during printing time, while allowing the parts to retain ample structural stability during the printing process. For smaller tiles, a steeper 45° angle was found to be desirable, as these were lighter and more stabilized due to a shortened distance between the supports and the build plate. This steepness in both cases also allowed for easy support material removal. Without access to this material, such as in the case of the parallel prints, the parts became nearly impossible to safely separate from the build plate.

The full-size 127 mm \times 127 mm tiles are printed with the Formlabs Form 2 printer set at its finest resolution (50 microns) with a z-scale factor of 1.000, density of 0.70, touchpoint size of 1.00 mm, 1.00-mm raft thickness, 0.90–1.00 slope multiplier, 4.00-mm height above raft, 2.60-mm flat spacing, 0.75-mm z-compression correction, and 0.30 mm early layer merge.

Discussion

Through the development of patterning designed to specifically guide and join hydrogel and ceramic, PolyTile 2.0 produced an advanced composite material and multi-step process for the production of dynamic, light-responsive ceramic bio-tiles. This proven technique first requires the designer to digitally craft (through design software) the surface design of a ceramic tile. After printing and firing, the surface is glazed as determined by the designer then washed with the pre-hydrogel solution. The ceramic-hydrogel composite automatically forms dynamic structures within and on the surface of the PolyTiles through the fluidic system composed of glaze chemistry, texturing, and geometry.





A single ray of sunlight/white light enters the droplet, refracting according to Snell's law, to split the light into the visible light spectrum. Although red-violet light exits at slightly variant angles, the average, "rainbow angle," is referenced often as 42° .

The implications of this process are extensive, as this novel method may allow for an assortment of bioresponsive behaviors and functionalities embedded within PolyTiles. This is especially promising given the extent of pre-existing methods for functionalized hydrogels that vary in sensing and responsive ability. With this case study on light-responsive hydrogel PolyTiles, it is a compelling beginning to the series because of the impact of light on health, perception, and information.

In the fluorescent-functionalized tiles, the hydrogel and patterned ceramic is shown to store and signal light-based data through a Fourier transform frequency pattern. The patterns can easily be translated to reflect data from human activity to further investigate its biofunctionality. The combination of this novel technology to create these composites along with carefully selected usage is provocative as it serves to address the duality of information communication. The pattern stores information about its environment, but it also uses its fluorescence to communicate back that data to impact the local environment. Introducing Fourier transforms into PolyTile has implications to a wide range of information storage and its visualization due to the applicability of Fourier transforms to temporal data. For instance, Fourier transforms may also hold data on noise, statistical analysis, and even crystallography.

Light responsiveness is dependent on the hydrogel's refractivity and reflectivity which shifts depending on the PEG:H₂O ratio, glaze type, and texturing. Techniques to form smooth hydrogel microdroplets stimulate a different trajectory to light responsiveness. Bio-inspired from raindrops and a simple, yet impactful atmospheric optical phenomenon, current findings from PolyTile 2.0 suggests its further development to selectively scatter and split white light which can potentially affect plant growth, human health, aesthetics, and/or aid in visualized storytelling.

The familiar and magical sight of rainbows is precisely derived from sunlight hitting a cloud of water droplets at the right angle (red emerges at about 42° and blue at 40.5°) to the observer to split white light into the visible light spectrum. The spherical droplet acts as a prism when light refracts twice and reflects once (see Figure 17) according to Snell's law.²⁰

On account of hydrogels being composed of almost all water, the rainbow theory is extended to hydrogel droplets formed on a ceramic surface through the advancement of ceramic texturing and glaze vocabulary. With increasing molecular weight, PEG hydrogels become transparent. The refractive index of 100% PEG hydrogels have been recorded to be 1.465,²¹ not far from that of water, which is commonly known to be 1.33. The diameter of the droplet is crucial as a diameter of 1-2 mm causes bright violet and green visibility as well

as red, but no blues. A diameter of 0.5 mm refracts weak red, greens, and violets. As the diameter gets to the microscale, 200–300 microns, the rainbow is broad and well developed, but even smaller diameters barely refract white light into its colors.

Ability to redistribute color encourages more efficient use of our most sustainable energy source since each color has a different role in natural processes. PolyTile 2.0 may further allow for functionalization that will redistribute the visible light spectrum to more efficiently use sunlight in this way. Both its fluorescence or color-change may indicate warning patterns or showcase other environmental factors depending upon the conditions at any given time. In terms of scalar potential, the outlined methods and processes exemplify the increasing feasibility of intelligent 3D printed componentry becoming an integral part of the future constructed environment at a full architectural scale (using component-based logics). With readily available equipment such as 3D Systems SLA Pro X 950 (build volume of $1500 \times 750 \times 550$ mm) and larger industrial machines already in use, the next generation and scaling of this component-scale research can leverage challenges related to industrial 3D printed ceramic production and the application of traditional and biologically steered glazing techniques within this larger-scale context. PolyTile 2.0's approach is distinctive because of this building-scale feasibility, translatable applicability in architectural design (i.e. façades), and its potentially expansive environmental and biological functionality as it enables incorporation of functionality existing and being researched in the hydrogel field. These tiles allow for programmability by the designer. Programmability is seen to be possible within one tile as well as through a collective suite which may produce compounded responses (e.g. environmental/biological input \rightarrow tile \rightarrow tile \rightarrow ... \rightarrow tile \rightarrow output). With this new generation of PolyTiles, 3D printing of dynamically responsive full-scale parts reaches an unprecedented closeness to feasible application within the built environment.

Conclusion

This newest series of PolyTile 2.0 prototypes proved the feasibility of developing full-scale 3D printed ceramic tiles with programmable bio-glazing strategies. Whereas earlier work explored hydrogel compatibility with DNA-based hydrogels, PolyTile 2.0 emphasized scalability and multi-pronged functionalities that could be more quickly implemented.

Upon success of this suite of tiles, which focused on visually distinct design phenomena and data-embedded designs, additional implementations of these biocomposite tiles will be further explored, which incorporate hydrogels that respond to other environmental factors critical for design concerns such as temperature fluctuations and its energy distribution, concentration of dissolved atmospheric gases, and so on. Research will look to develop PolyTiles that provide a direct response to the stimulus, and further, secondary response systems. Such secondary responses may look to hydrogels that reversibly switch between hard and soft states when heated or cooled.²² The versatility and composite properties will provide new agency to architects.

Through additional case studies into specific environmental contexts along with the use of the patterning and embedment process presented here, the PolyTile series intends to expand into multi-tile façade systems capable of architectural-scale environmental response.

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Appendix

Tile variations

- The tile was divided into a 30 × 30 grid with the center of the extruded (1 mm) rectangular texturing at the intersection points of the grid. Tile contained a single convex curvature. The back of the tile has a 4 × 4 grid frame of 3-mm thickness behind the piece for structural support. The thickness of the tile is 2.5 mm at its thinnest and 10 mm at its thickest. The texturing dimension decreases along the vertical, perpendicular to the directure of the curvature as a gradient with the smallest side length of 500 microns and largest of 3.3 mm. The tile was glazed as a matrix of varying glaze types, and the hydrogel presolution was washed over the entire tile.
- 2. Identical to Tile 1, but with texturing dimensions decreasing down the axis perpendicular to the direction of curvature ($y = 3.0004 + 0.4421233x 0.01406171x^2 + 0.0001321406x^3 3.729295e 7x^4$). The thickness of the tile is 5 mm at its thinnest and 11 mm at its thickest. Glazing alternated between strips (6) of fully white glazed or ZnO glaze and scraped to expose the non-glazed pattern. The hydrogel presolution was washed over the entire tile.
- The tile (4.5-mm thick) was divided into a 12 × 12 grid with the center of the hemisphere texturing at the intersection points of the grid. The texturing dimension decreases along the diagonal as a gradient with the smallest diameter of 2.828 mm to largest of 8.486 mm.

Two versions using these dimensions are as follows:

- a. *Extruded partial spheres, 90% extruded out.* Fully coated with ZnO glaze with full tile washed with presolution.
- b. *Intruded hemispheres*. White glaze applied over all tile avoiding wells. Wells coated in NS glaze. Presolution applied to wells using dropper to well's maximum fill (presolution spherical droplets sit on wells).
- 4. The tile (4.5-mm thick) was divided into a 24×24 grid with the center of the extruded hemisphere texturing at the intersection points of the grid. The texturing dimension decreases along the diagonal as a gradient with the smallest diameter of 850 microns and largest of 5.656 mm. The tile was divided into four quadrants for glazing: opposite quadrants were either fully coated in white glaze or ZnO glaze. All fully washed with hydrogel presolution.
- 5. The tile (4.5-mm thick) was divided into a 60×60 grid with the center of the extruded (2 mm) rectangular texturing at the intersection points of the grid. The texturing dimension decreases along the diagonal as a gradient with the smallest side length of 100 microns and largest of 1.2 mm. Entire tile was white glazed and scraped to expose non-glazed texturing. The hydrogel presolution was washed over the entire tile.
- 6. The "JSLab" lettering was extruded 1.5 mm from the surface. Entire tile (4.5-mm thick) was white glazed and scraped to expose the non-glazed texturing. The hydrogel presolution was washed over the entire tile.

- 7. *The negative (black) pattern was extruded 2 mm from the surface of the tile. The entire tile (4.5-mm thick) was white glazed and then scraped to expose the non-glazed pattern where it was extruded. The hydrogel presolution was washed over the entire tile.
- 8. *The negative (black) pattern was extruded 2 mm from the surface of the tile. The entire tile (4.5-mm thick) was white glazed and then scraped to expose the non-glazed pattern where it was extruded. The hydrogel presolution was washed over the entire tile.

*The frequency-based tiles are extruded based on its respective Fourier transform data (pictured below either tile).

