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In situ Resource Utilization and Reconfiguration of Soils Into Construction Materials for the Additive Manufacturing of Buildings

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The construction industry is being buffeted by winds of change, balancing the urgent need to remedy deteriorating infrastructure in the developed world and the push to build new infrastructure in emerging economies whilst devising means to better its catastrophic carbon footprint. Much of the deleterious environmental impact of construction derives from the utilization of concrete as well as inefficiencies across the construction process that result in considerable waste and energy expenditure. Additive manufacturing methods stand poised to substantially transform the industry by enhancing automation, enabling economy of materials use, and allowing for unprecedented fusion of form and function; however, reliance on concrete as the extrusive material of choice has the potential to greatly compound mounting environmental challenges. In this perspective, we discuss our efforts to develop an altogether new palette of naturally sourced construction materials based on natural soils, which are reconfigured into extrudable formulations compatible with additive manufacturing. We furthermore delineate a roadmap bringing together soil chemistry with composite science, modeling of mesoscale phenomena, rheological studies of extrudable soil "inks," generative design, and the development of robust structure—function correlations relating atomistic and mesoscale structures as well as geometry of the architectures to load-bearing capabilities and mechanical response. We illustrate this approach using a naturally harvested burlewash clay sample crosslinked through formation of a siloxane framework, which has been 3D printed into a load-bearing structure. The need for an integrated life cycle assessment approach is emphasized to ensure development of a new palette of sustainable construction materials.

Keywords: additive manufacturing, clays and clay minerals, structural materials, life cycle (impact) assessment, concrete, rheological performance

INTRODUCTION

The construction industry serves as one of the primary bulwarks of the global economy; global construction spending is estimated to be 13% of GDP, and indeed some estimates suggest that about 7% of the world's working age population is employed in activities related to construction (Barbosa et al., 2017). However, this industry is widely regarded to be deficient in terms of global value addition and has been characterized by relatively sluggish growth in efficiency and productivity in recent years owing to the slow adoption of innovations in materials, automation, and manufacturing that have transformatively altered other industries (Bakshi et al., 2015; The Economist, 2017). The urgent need to remedy deteriorating infrastructure in the developed world and the push to build new infrastructure in emerging economies has provided much recent impetus to efforts to reinvigorate construction through the adoption of new materials (Bechthold and Weaver, 2017), increasing implementation of additive manufacturing methods (Pittsburgh, PA: Daehn and Spanos, 2019), and the widespread use of automation across the construction process. Sustainability and emphasis on energy and resource conservation play a central role in these efforts, which further hold promise for increasing global access to dignified habitats, enabling optimal architecture through generative design, and for enabling construction in inhospitable terrains. In this perspective article, we delineate our ongoing efforts to harvest natural soils and to reconfigure such complex mixtures of clays, rock detritus, and organic matter into extrudable formulations compatible with additive manufacturing processes. We furthermore emphasize the need for multiscale materials design and modification to enable a new paradigm of materials construction utilizing locally sourced soils structured in geometric patterns that provide novel function. Developing extrudable soil formulations that can be cured into viable structures requires the convergence of soil chemistry with composite science, predictive modeling of mesoscale phenomena, control over the rheology of extrudable soil "inks," generative design, post-synthetic modification and cross-linking of printed architectures, and the development of robust structure—function correlations relating atomistic and mesoscale structures as well as geometry of the architectures to load-bearing capabilities and mechanical response.

ADDITIVE MANUFACTURING AND CONSTRUCTION: APPROACHING A TIPPING POINT

3D printing enables the macroscopic structuring of matter in hitherto inaccessible geometries, spanning the range from individual machined components to infrastructure such as bridges and walkways as well as entire habitats, yielding economies of materials use as well as allowing for unprecedented geometric design complexity (Hager et al., 2016; Tay et al., 2017) to be accessed on a routine and repeatable basis. However, materials currently used for 3D printing in construction are neither environmentally sustainable nor tailored to the needs of the 3D-printing process. Low "buy-to-fly" ratios, which are central to the fundamental promise of additive manufacturing to reduce raw material use remain to be realized, and reliance on homogeneous materials compositions squanders the potential benefits of a new construction paradigm that is tailored to the resources and needs of the local environment.

The extrusive material of choice for most current prototypes is currently concrete (Buswell et al., 2018; Flatt and Wangler, 2018; Wangler et al., 2019); this is indeed worrisome since the concrete industry is one of the largest contributors to anthropogenic CO_2 emissions, responsible by some accounts for up to 5—7% of global emissions (World Business Council for Sustainable Development [WBCSD], 2002; Damtoft et al., 2008; IEA, 2008; Allwood et al., 2010; Friedlingstein et al., 2010; UNSTATS, 2010; Hasanbeigi et al., 2012; Andrew, 2018). Some estimates suggest that 85.9 million tons of Ordinary Portland Cement (OPC) were produced in the United States in 2017 (U.S. Geological Survey., 2018). Even with modernization of manufacturing processes, OPC production consumes 4 GJ of energy and releases approximately one ton of CO_2 per ton of OPC (Worrell et al., 2000, 2001; Khurana et al., 2002; Choate, 2003; Thomas and Peethamparan, 2015). As such, a very real concern is that the advent of 3D printed concrete will exacerbate already compounding environmental challenges by promoting unsustainable building practices. Considerable attention has therefore focused on the search for more environmentally sustainable alternatives to concrete, especially naturally sourced clays that can be harvested and deployed without need to transport materials over long distances, enabling a considerably reduced carbon footprint and embedded energy costs. It is further noteworthy that 3D printing methods for extrusion of concrete continue to struggle with incorporation of common aggregates and integration with rebar. As such, the additive manufacturing of load-bearing concrete sections that meet code is fraught with challenges and the primary applications of 3D printing have been limited to architectural facades.

A substantial portion of appeal of 3D printing of large-scale structures is the opportunity to robotically automate construction in extreme or hostile environments (e.g., clinics in times of war or in disease-ridden jungles, extra-terrestrial planetary environments, etc.). Such applications require the utilization of locally sourced materials. Locally sourced materials can furthermore be returned to the natural environment from which they are derived providing an end-of-life scenario that is substantially improved as compared to current building options.

DEVISING A NEW MATERIALS PALETTE: FROM DUST TO STRUCTURES

The inevitable transformation of the construction industry will have widespread resonance for the future of work (Autor, 2015). As 3D-printing technologies transform urban landscapes, the design of a new materials palette has emerged as an urgent imperative and is necessary to mitigate the catastrophic environmental impact of concrete and improve the quality of life of vulnerable populations across the globe. Rather than importing resources, in this paradigm, materials for 3D-printing can be harvested locally from cohesive soils and/or waste plant matter—reducing transportation costs and energy expended, simplifying construction in remote or dangerous locations, and potentially capturing and sequestering carbon.

The goal of fabricating robust, load-bearing architectural forms from naturally sourced structural materials using additive manufacturing (3D printing) processes derives direct inspiration from the impressive structural feats accomplished by ingenious animal species spanning the range from termites to beavers and spiders (Morrison et al., 1977; Napp et al., 2012). Traditional building practices across the world have extensively borrowed from these and other examples, utilizing clays and rocks bonded together with naturally sourced gelling agents to sculpt durable multifunctional structures, generally well-adapted to the needs of local climates. The strength and flexibility observed in such forms of architecture are intrinsic design elements missing in concrete and derive from the simultaneous use of both inorganic and organic components.

The design of a new materials palette spans multiple length scales is schematically illustrated in **Figure 1**. Such design ranges from atomistic considerations such as the specific cross-linking modes and their kinetics of gelation, surface modification or intercalation of soil particles to facilitate modification of properties and imbue compatibility with the continuous matrix, as well as the local structure of interfacial layers and their kinetics of nucleation and growth (Kazemi Najafi, 2013; Meng and Li, 2013; Parija et al., 2018). Such atomistic considerations underpin mesoscale structure and porosity, which is further amenable to modification by inclusion of fibrous reinforcements (as a substitute for rebar) or geometric texturation established during processing. Macroscopic properties in turn derive from the interconnection of mesoscale building blocks and can be controlled with great precision by modern additive manufacturing processes, which assemble components with micron-scale voxel-level control. While the sourcing of components from local soils, native vegetation, and crops impose specific constraints, the versatility of the available materials lends itself to entirely new opportunities and function entirely inaccessible with concrete.

In terms of function, mineral-derived particulate building blocks must be processable and possess tunable surface chemistry such that they can be dispersed within liquid media and extruded using 3D printing methods; such particles furthermore must have intrinsic elastic moduli and hardness such that they are able to withstand mechanical stresses without intragranular cracking. The galleries of layered clays provide a substantial design space for modification through expansion, exfoliation, ion exchange, and insertion of organic cations and monomers (Leroux and Besse, 2001). Interfacial chemistry to link particulates is of particular importance and requires the formation of robust chemically and thermally stable linkages—either periodic inorganic linkages (e.g., oxo, aluminosilicate, and polyborate frameworks) or specific covalent linkages (e.g., oligomeric backbones with one or more modes of covalent or non-covalent cross-linking). Such linkages furthermore must interface with fibrous additives or aggregates and ensure their homogeneous dispersion within the continuous matrix. The interfacial chemistry must allow for load transfer to such reinforcements mitigating damage mechanisms such as pull-out failure and interfacial delamination. The matter of obtaining homogeneous dispersions with bound interfaces of course represents a longstanding challenge in the design of matrix composites, which in this case has to be resolved in order to embed fibers within a soil matrix (Marshall and Evans, 1985; Thouless and Evans, 1988; Sakai et al., 2000). 3D printing requires layers to be robotically deposited and then bound together before hardening, but still harden quickly enough to avoid structural collapse as the next layer is deposited. As such, kinetics of gelation and cross-linking are of vital importance and necessitate the incorporation of catalysts and retardants that enable precise modulation of temporal strengthening profiles. As with the design of functional nanocomposites, embedding additional components that imbue properties such as chemical resistance or protection against UV radiation, deter colonization by microbes, increase thermal insulation, and provide self-cleaning or liquid repellant properties requires introduction of gradients of material inclusions, use of microcapsules and coatings, and anchoring of the inclusions within the continuous matrix (White et al., 2001; Shchukin et al., 2006; Zheludkevich et al., 2007; Dennis et al., 2015; Fleer et al., 2017). The idea of self-healing is one that is particularly important for the design of resilient structures and has been explored by embedding monomeric species within nanoscale containers; the monomers are released in response to mechanical or chemical stimuli and are cross-linked to heal damage (White et al., 2001; Cho et al., 2009; Cotting and Aoki, 2016).

Macroscopic texturation and structuring of naturally sourced composite materials into intricate forms of architecture further provides opportunities for programming mechanical resilience while introducing new function. For instance, triangulation introduces stability, pockmarking a surface modifies acoustical reverberation (Acoustical Society of America [ASA], 2013), whereas shaping openings in a façade streamlines natural ventilation (Zhivov et al., 2001). Some representative vignettes from our current work are discussed in subsequent sections.

GETTING IT RIGHT FROM THE START: EMBEDDING LIFE CYCLE ANALYSIS IN STRUCTURAL MATERIALS DESIGN

The building sector is responsible for over 48% of global energy consumption each year including for building construction and operation, thereby accounting for over 40% of global carbon emissions (Copiello, 2016; Dixit, 2017, 2018). The total energy consumed by a building over its life cycle encompasses embodied and operating energy (Copiello, 2016; Dixit, 2017, 2018). Embodied energy is consumed: (1) directly over the course of construction, fabrication, installation, transportation, and administration processes; and (2) indirectly through the use of construction materials, assemblies, and equipment, which

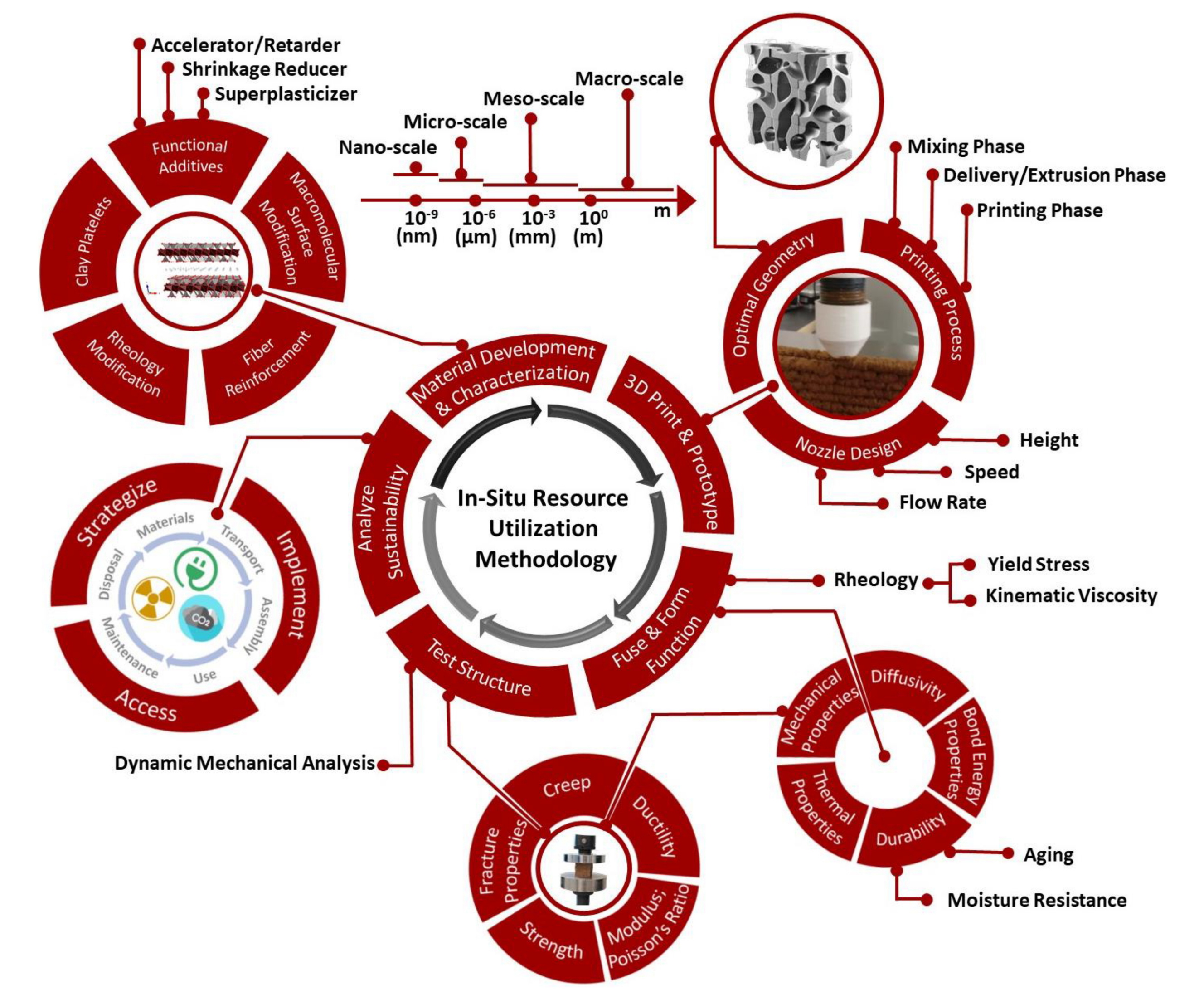

FIGURE 1 | Schematic representation of a proposed process flow for *in situ* resource utilization. The approach depicts the multiscale design of a palette of sustainable 3D-printable construction materials that address a critical imperative for sustainable utilization of the earth's limited resources. This strategy brings together materials development, assessment of printability and buildability, with embedded multifunctionality. Material development efforts are focused on the utilization of naturally sourced clay platelets through surface modification and fiber reinforcement. The modified soil is rendered extrudable by developing hybrid admixtures with the appropriate rheology; 3D printing and prototype development efforts enable exploration of generative design concepts. The fusion of form and function in test structures yields a vastly expanded scope for embedding novel functionality through geometric design whilst achieving economy of materials use. An integrated framework involving evaluation of rheology and printability, assessment of desired function such as load bearing properties and stability, and life cycle assessment enables iteration and successive improvement of all metrics.

incur a variety of energy costs (Copiello, 2016; Dixit, 2017, 2018). In contrast, operational energy includes the energy expenditures associated with air-conditioning, heating, lighting, hot water supply, and powering of building equipment (Copiello, 2016; Dixit, 2017, 2018). Bringing about substantiative reductions in the energy and carbon footprint of buildings requires curtailing the use of both embodied and operating energy (NSTC, 2008; Dixit, 2018). Operating energy costs for modern buildings have been significantly reduced through the incorporation of energy efficient building components (e.g., low-e glass and advanced insulation), adaptive HVAC systems, fixtures such as light-emitting diodes, altogether underpinned by advances in building energy simulations that have allowed for systems level optimization of structures (Copiello, 2016; Dixit, 2017, 2018). However, costs of embodied energy remain substantial and undiminished. Indeed, several studies have emphasized that substantial reductions in operating energy have often come at the cost of utilizing energy-intensive materials, which in turn deleteriously impacts the embodied energy balance sheet (Ghattas et al., 2013; Dixit and Singh, 2018). The direct energy component of embodied energy derives primarily from construction and related processes. The proportion of indirect embodied energy in total embodied energy (>90%) and associated carbon emission outweighs that of direct energy (Dixit, 2017). As such, building materials play a substantial role in the total energy and carbon embodied within a building. Both direct and indirect impacts of construction can be potentially mitigated to a large extent by adopting altogether new structural materials and efficient construction methods that allow for economy of materials use and reduction of waste generation.

Materials that are manufactured using locally extracted natural resources with expenditure of relatively less process energy are typically characterized by a lower embodied energy and relatively reduced carbon footprint (Buyle et al., 2013; Langston, 2015). However, selecting a material based on just lower embodied energy is not a sufficient criterion and requires further consideration of durability, recyclability, and reusability (Langston, 2015; Dixit, 2018). For instance, a material characterized by a higher embodied energy and longer service life can indeed afford greater environmental benefits as compared to one with smaller embodied energy and shorter service life (Langston, 2015). Similarly, using a higher embodied energy material with higher recyclability or reusability may help offset a substantial portion of embodied impacts. As such, there is a growing demand for natural, locally sourced, and durable materials with higher recyclability and/or reusability potential and lower maintenance requirements (Ardente et al., 2005; Wilson and Boehland, 2005; Akadiri et al., 2012; Menzies and Tsolaki, 2016). Harnessing natural materials for construction further holds promise for reducing the toxicity of built environments if traditional materials often associated with harmful indoor emissions can be eliminated (van Wijk and van Wijk, 2015; Ford and Despeisse, 2016; Ghaffar et al., 2018). Local materials are furthermore often adaptable to the local climatic conditions, and thereby can potentially be used to offset heating and cooling loads (Labonnote and Rüther, 2017). Such materials can help maximize the energy and carbon benefits over the whole life cycle of a building. In addition, supplanting concrete with a vastly expanded repertoire of natural materials may also help decrease embodied impacts. For instance, a material with enhanced structural, thermal, and moisture control performance, can replace traditional multi-layered wall assemblies containing drywall, stud core, mineral wool, and a number of cladding materials (Biswas et al., 2017). This not only simplifies the construction process but improves the potential for recycling owing to the utilization of natural materials that can be disassembled into soil and fibrous components through processes such as microbial degradation. An urgent imperative therefore is a rigorous life cycle assessment and embodied energy accounting of natural materials and their reconstitution processes to serve as a blueprint and provide guiding principles for the design of novel structural materials. Such accounting will further enable a clear contrast to be drawn with the utilization of concrete.

The processes of on-site or off-site construction, fabrication, and installation involve the use of a variety of hand and power tools and equipment, which consume energy typically in the form of electricity, diesel, and gasoline (Copiello, 2016; Dixit, 2017, 2018). These processes also include transportation and hoisting of materials, assemblies, labor, and equipment, which further incur substantial energy expenditures (Dixit, 2017, 2018). A construction site furthermore incurs energy expenditures on account of lighting, air-conditioning, and heating. Energy is not only consumed by equipment, vehicles, and tools but also by human labor (Dixit et al., 2015). Although construction-related direct energy consumption is generally lower than the indirect energy of materials and products, digitalization and automation derived from implementation of additive manufacturing methods can potentially bring about substantial reductions in such energy expenditures, generating significant environmental benefits across the industry.

AN EXPANDED PALETTE OF CONSTRUCTION MATERIALS: BUILDING WITH GEOPOLYMER CONCRETE AND MUSKEG SOIL

As noted in the preceding sections, the cement manufacturing industry is a major contributor to anthropogenic CO_2 emissions and as such numerous alternatives have been explored to expand the palette of viable construction materials. Recently, alkali-activated (AA) binders, also known as geopolymers have gained increasing attention as alternatives to OPC and appear to yield mechanical strengths comparable to OPC (Ding et al., 2016, 2018). These materials have the potential to reduce greenhouse gas emissions (Shi et al., 2003; Duxson et al., 2007; Davidovits, 2008; Pacheco-Torgal et al., 2008; Yang et al., 2013) by as much as 25–45% (Stengel et al., 2009; Turner and Collins, 2013) or even up to 70% by some estimates (Weil et al., 2009). Alkali-activated cement further shows better performance than OPC such as early strength development, durability, resistance to chemical attack, hydration heat, and resistance to freeze-thaw conditions (Shi and Fernández-Jiménez, 2006; Ding et al., 2018).

Geopolymer cement generally comprises a combination of bauxite tailings, fumed silica or amorphous alumino-silicate materials, fly ash or natural zeolites, in reactive combination with alkali-based chemicals (alkali activator), for example hydroxides and silicates. This reactive mixture yields a geopolymer cement that can be constituted into a geopolymer concrete (Davidovits, 2008; Provis and van Deventer, 2009, 2014; Provis, 2014; Provis and Bernal, 2014). The underlying chemistry involves the reaction of alumina and silica to form an amorphous aluminosilicate network; the strength of the framework depends on the silica to alumina ratio. The resulting framework's strength is comparable to Portland cement concrete (De Silva et al., 2007; De Silva and Sagoe-Crenstil, 2008; De Vargas et al., 2011; Phoongernkham et al., 2014). The amount of aluminum influences the setting time and reactivity of the blend in such a way that Al-substituted calcium silicate hydrate (C-S-H) or calcium aluminum silicate hydrate C-A-S-H form instantly and continuously, which imbues high early strength to this system (Puligilla and Mondal, 2013).

The viability of a material to be deposited through 3D printing is strongly predicated on its ability to flow and its temporal setting profile (rate of emergence of mechanical strength). As such, the rheological characteristics of the material have to be intricately tuned. For instance, the rheology of fresh cement paste depends on the speciation of hydration products, inter-particle interactions, and the extent of dispersion and concentration of particles. Superplasticizers (organic admixtures) are generally used to enhance the workability of the cement paste by dispersing the particles, modifying interfacial energies, and hence facilitating more rapid development of strength (Shah and Ahmad, 1994; Uchikawa et al., 1997; Ouellet-Plamondon and Habert, 2016). Most superplasticizers are polymeric dispersants, which modify inter-particle interfacial properties and reduce the yield stress of solid paste, thereby yielding cement mixtures that are more readily flowable (Flatt et al., 2012). Superplasticizers have facilitated the development of self-compacting concrete, which requires the inclusion of fewer aggregates as compared to conventional concrete and thereby becomes load bearing on its own without the need for reinforcing additives (De Larrard, 1999). Polycarboxylate ether (PCE) is one of the major components of commercial superplasticizers (Lange et al., 2014; Li et al., 2014) and comprises cross-linked carboxyl groups at its backbone with polyethylene oxide (PEO) side chains (Winnefeld et al., 2007). Clay minerals such as kaolinite, illite, and montmorillonite interact with PCE owing to the intercalation of the PEO side chain within the aluminosilicate clay layers resulting in strong adsorption of the plasticizer onto the clay mineral (Liu et al., 2004; Suter and Coveney, 2009; Li et al., 2017). As a result, the inclusion of even small amounts of clay minerals in concrete can greatly modify its flowability and mechanical strength (Li et al., 2017). As such, in order to incorporate clays within conventional geopolymer formulations and concrete without compromising rheology and mechanical strength of the 3D printed material, appropriate surface modification schemes have to be designed to facilitate compatibility and to potentially engender pozzolanic reactions.

A HISTORICAL PERSPECTIVE OF SOIL AS A BUILDING MATERIAL

Almost 95% of the Earth's crust is composed of quartz and rock-forming silicates (Lagaly et al., 2000; Ito and Wagai, 2017). Abundant minerals in the Earth's crust include plagioclase feldspar (39%), alkali feldspar (12%), quartz (12%), pyroxenes (11%), non-silicates (8%), amphiboles (5%), micas (5%), clays (5%), and other silicates (3%) (Hart, 1969). These minerals thus form the essential palette that must be utilized for digital fabrication with naturally sourced materials. The construction of dwellings from indigenous soil and fibers has a long history and this practice continues to be widespread across the world (Scalenghe et al., 2015; Zhang et al., 2017). Naturally-occurring clay minerals have a variety of mineralogical and chemical characteristics appropriate for construction (Reeves et al., 2006). At low water content and high bulk density conditions, clay minerals have the tendency to compact to form rock-like materials, whereas at high water content, they oftentimes form moldable or pourable gels or pastes making them suitable for extrusion processes. Kaolinites, talc, and pyrophyllite have been used for centuries for manufacturing bricks, ceramics, tiles, and glazed products. Smectitic clays (such as montmorillonite) combined with natural binders, such as dried straw, are used in traditional adobe bricks (Zhang, 2013; Smith et al., 2016; Shubbar et al., 2019). Although clay minerals are some of the fundamental components of nearly all soils, the clay content of soils usually is less than 50%. The remainder of the soil is composed of a wide spectrum of inorganic minerals, amorphous inorganic solids, and organic residues. Therefore, one of the first challenges in employing soils as a component of construction material is determining the composition of the soil to ensure that the soil contains the necessary content of building blocks. Low clay content or predominantly coarse particles will restrict the reactivity of the soil and have a negative impact on rheology (Mueller et al., 2016). This can be overcome in part by decreasing the particle size of the soil through mechanical grinding or making the soil a minor component of the composite. High clay contents are more desirable, but not all clays easily lend themselves to construction applications. Smectitic clays (such as montmorillonite) expand dramatically when wet, a property ideal for drilling muds but detrimental to building foundations. However, the expansive properties of clays can be overcome by exfoliating the clays (separating layers of structural units) and placing binders between the layers. The inherent heterogeneity of soils can add to the challenge of finding the appropriate type and quantity of soil to be used. Soils always vary with depth and can vary dramatically over short lateral distances.

Brick-making from clay soils is well-established for the construction of dwellings across the world. The process can be as simple as mixing grass with wet clay soil and drying the bricks under the sun. Various additives can be used to increase strength or hydrophobicity, and the bricks often are heated in a kiln for curing. Perhaps the most obvious use of soils is as a framework for road construction or other horizontal construction. Lime or cement stabilization of the soils is well documented and involves adding lime or cement as required, adding water, compacting, and allowing the passage of time for hardening. Soil-cement technology has been attempted for vertical construction, but not to the extent as its horizontal counterpart, and the science is still maturing. Other approaches have been attempted, but a common problem is the lack of resistance to water (Reeves et al., 2006). More sophisticated approaches have been attempted using more chemically complex additives including "water glass" (a highly concentrated aqueous solution of sodium or potassium silicate), gypsum, and epoxy additives to imbue additional cross-linking modes and resistance to water permeation.

As an example of the practical feasibility of harvesting local soils and plant fibers to build load-bearing structures, we have recently reported a method for preparing a densified composite material suitable for use in roadworks from muskeg fibers and wood chips sourced from Northern Alberta that are cross-linked using soluble silicates and functionalized cellulose (Waetzig et al., 2017). The marshy soil in this sub-arctic region limits road transport to peak winters when the top soil is frozen solid; transportation on solid pack ice is associated with tremendous cost and risk. Alternative approaches require excavation of the muskeg layer and backfilling with engineered geofills, which is oftentimes cost prohibitive. This novel *in situ* geopolymerization method allows for reinforcement of native muskeg with wood fibers derived from native vegetation with the addition of inorganic silicate precursors. A continuous siloxane network is formed that links together the muskeg, wood fibers, and added silicates yielding a load-bearing and low-subsidence composite. Where fibrous peat has compressive strengths on the order of 3.5—11 kPa, our geopolymerized muskeg samples attain compressive strengths as high as 33 MPa upon cross-linking with sodium silicate and reinforcement with an interpenetrating network of mulch fibers (Waetzig et al., 2017). Notably, we have further illustrated that the siloxane framework is readily dissolved by base treatment, allowing for restoration of the soil to its native condition as represented in **Figure 2A** (Waetzig et al., 2017).

An alternative approach we have explored involves the incorporation of naturally derived clays within cement to reduce thermal conductivity without deleteriously impacting compressive strength. Such an approach provides a sharp reduction of thermal conductivity, thereby mitigating heat losses through oilwell cement during thermal extraction processes. Specifically, we have examined the incorporation of hydroxyethylcellulose-functionalized halloysite in class G Portland cement (Cho et al., 2018; Udayakantha et al., 2019). Approximately 2—5 wt.% of halloysite nano-clays with nanotubular voids are added to the cement achieving 7 days compressive strengths as high as 15.71 MPa (with even a slight enhancement of compressive strength with respect to control specimens without halloysite) whilst engendering upto ca. 83% reduction of thermal conductivity. The amount of clay added in the cement in the study is along the lines of soil added to concrete in previous reports (Lei and Plank, 2012; Li et al., 2017). The reduction of thermal conductivity derives from the inclusion of nanoscopic voids and the phonon scattering at interfaces within the composite (Cho et al., 2018; Udayakantha et al., 2019).

ADDITIVE MANUFACTURING USING NATURALLY SOURCED MATERIALS: BUILDING FROM THE BOTTOM UP

Additive manufacturing along with generative design processes that search potential geometric configurations (Rocca, 2012) in search of optimal functionality enable the construction of self-supporting, load-bearing geometries for full-sized building blocks. Layer-by-layer deposition techniques used in additive manufacturing allow for greater flexibility and versatility of architectural forms while resulting in improved materials economy and consequently reduced energy consumption. In general, establishing patterns for 3D printing (e.g., layer thickness and layering pattern) necessitates the selection of four distinct parameters to obtain optimal mechanical properties of the printed material (Buswell et al., 2018; Flatt and Wangler, 2018; Kalantar et al., 2018; Reiter et al., 2018; Wangler et al., 2019): (1) Modification of soils to facilitate dispersion; (2) preparation of an extrudable "ink" formulation; (3) delivery of the formulation to the nozzle or application unit; and (4) design of print head and implementation of the printing process. In extrusion printing, in addition to the materials properties of the formulation, process parameters such the height (z), nozzle speed, and flow rate critically affect the quality of 3D-printing. The thickness of the 3D-printed layer is controlled by the printer nozzle and affects the duration of the printing. In contrast, the height of each layer, the interfacial properties between consecutive layers, and the layering patterns affect the macroscopic mechanical properties of the printed material (Bhardwaj et al., 2019). The promise of 3D printing is the generation of structures that can be fabricated to be simultaneously light and rigid with forms having capacity to endure internal and external forces (Mansoori et al., 2018). The development of printheads and delivery systems tuned to the needs of complex reconfigured soils represents a substantial challenge for the discipline. Analogously, matching the rheology of soil formulations to specific printheads and delivery systems and establishing control over curing kinetics further represent substantial challenges that remain to be satisfactorily resolved. Premature curing results in clogged nozzles (an altogether familiar bane of additive manufacturing), whereas delayed curing implies that load-bearing function cannot readily be achieved and establishes severe constraints over "buildability" and the dimensions and designs that can be accessed using additive manufacturing (Buswell et al., 2018; Flatt and Wangler, 2018; Reiter et al., 2018).

A CASE STUDY FOR ADDITIVE MANUFACTURING: PRINTING LOCALLY SOURCED SOIL (BURLEWASH SERIES CLAY)

In order to create a 3D printable and load-bearing soil-composite structure, we have used montmorillonite-rich Burlewash series clay sampled in College Station, Texas and evaluated through standard soil characterization methods. The soil is acidic in nature with a pH of 4.94, which is typical of the B horizon in Burlewash series. The collected soil has an electrical conductivity of 108.3 μS/cm. The soil sample was dried under air and ground using a BB 200 jaw crusher (Retsch, Verder Scientific, Inc.) to remove external moisture present in the soil. The ground soil was sieved through a 200 × 200 mesh sieve (75 μm pore size) for consistency in particle size of each sample along with higher reactivity of soil and for the ease of 3D printing thereafter. The particle size distribution of the processed burlewash soil was measured using a Horiba laser scattering particle size distribution analyzer (LA-960) (**Figure 2B**); the d_{50} of the soil was found to be 10.8 microns which means 50% of the soil has a particle size of 10.8 microns. The fine-grained nature of this soil imbues higher reactivity. A consistent solid concentration of 2.8 g/L was maintained while performing particle size analysis on three replicates

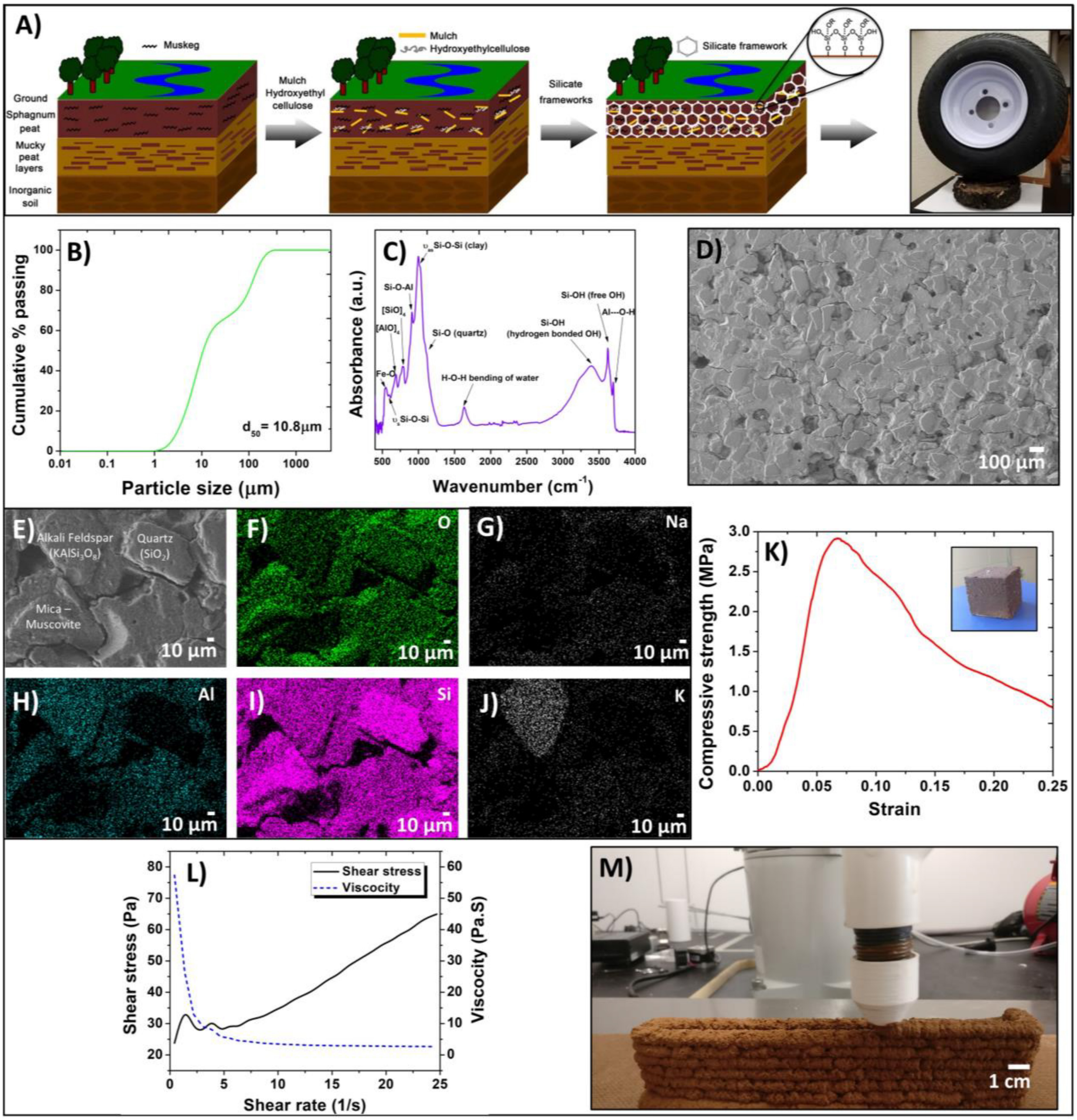

FIGURE 2 | (A) Illustration of geopolymerization approach developed to solidify muskeg to a load-bearing silicate composite, as represented by the force of an automobile tire on road reinforced by the composite, reprinted with permission from Waetzig et al. (2017) Copyright 2017 Springer Nature. **(B)** Particle size analysis of locally sourced Burlewash clay. **(C)** Attenuated total reflectance – Fourier transform infrared spectrum (ATR-FTIR) measured for Burlewash clay. **(D,E)** Scanning electron micrograph of the Burlewash clay composite. Spatially resolved elemental mapping using energy dispersive X-ray analysis at **(F)** oxygen; **(G)** sodium; **(H)** aluminum; **(I)** silicon; and **(J)** potassium edges. **(K)** Compressive strength testing of the burlewash clay composite; a digital photograph of the cube used for testing is shown in the inset. **(L)** Rheological curves measured for the printable burlewash clay formulation. **(M)** Digital photograph depicting the extrusion step of the 3D-printing process for burlewash series clay composite paste.

for each sample. Fourier transform infrared (FTIR) spectra were acquired using a Bruker VERTEX-70 FTIR instrument equipped with a PIKE MIRacle single-reflection horizontal attenuated total reflectance (ATR) accessory. **Figure 2C** depicts the ATR-FTIR spectrum for the ground Burlewash series clay. Distinctive IR bands associated with stretching modes of aluminosilicate frameworks are observed and assigned in the spectrum (Calabi-Floody et al., 2011).

In order to develop an extrudable formulation, a 1:1 (w/w) mixture of sodium silicate and Burlewash series clay (ground soil) was prepared. Alkaline water and a cellulosic admixture were added to the matrix and the components were initially mixed using a shear blade mixer (KitchenAid, professional 600 series) at 181 rpm for 1 min. Intimate mixing of the mixture was facilitated by extensive shear mixing at successively higher speeds, reaching 454 rpm for 1 min. The admixture has been added to facilitate dispersibility of the sodium silicate with the clay particles, thereby enabling the formation of a siloxane framework across the composite. Rheological characteristics of the formulation have been measured using a DHR-2 rheometer (TA Instruments). The static yield stress was determined by applying consecutively increasing shear rates in the range of 0–25 s^{-1}. The yield point represents the critical shear stress which is required to overcome the initial stress applied to the material. In other words, yield stress is the minimum shear stress necessary to initiate flow at a certain shear rate. **Figure 2L** shows the yield stress of the mixture which was found to be 34 Pa with a kinematic viscosity of 26.4 Pa·s at a shear rate of 1.4 s^{-1}. A shear rate-independent viscosity of 3 Pa·s is observed at a shear rate of 10 s^{-1}. The low viscosity of the mixture showed that it is a readily printable material amenable to 3D printing. An extrudable ink formulation has been developed from this mixture. An ABB IRB 1200 robot was used for 3D printing of this modified-clay—geopolymer—admixture composite. A visual programming plugin Grasshopper that runs on Rhino 6 along with Robotstudio was used for programming the robot. Compressed air at a pressure of 4.5 bar was used with varying auger speeds and a robot speed of 3 cm/s.

Figures 2D,E show scanning electron micrographs (JEOL JSM-7500F field-emission scanning electron microscope) displaying the microstructure of the printed composites, illustrating the continuous connectivity of the mineral particulates linked together through a siloxane framework. The composites have further been characterized using energy dispersive X-ray (EDX) spectroscopy. **Figures 2F—J** represent spatially localized maps corresponding to elemental distributions of O, Na, Al, Si, and K. Overlay of these signals enables identification of mica, alkali-feldspar, and quartz particulates, which are bound together within the composite through siloxane linkages formed by addition of sodium silicate.

In parallel to printing of the composite, a standard 2′′ × 2′′ block specimen (**Figure 2K**) has been molded for compressive testing as per ASTM methods. Compressive strengths approaching 3 MPa are measured using an Instron 5984 testing device. **Supplementary Video S1** demonstrates the ability of the hardened composite to withstand mechanical stresses. **Supplementary Video S2** demonstrates the extrusion of the modified Burlewash clay to constitute a load-bearing nanocomposite architecture (**Figure 2M**).

As perspective, soil and sodium silicate are used to fashion an extrudable 3D-printed composite without need for concrete. The facile availability of clay-rich soils in Earth's crust makes the approach demonstrated in **Figure 2** widely generalizable to different geographies. Burlewash soil harvested here is fairly typical of soils in terms of clay content and the presence of minerals such as feldspar, quartz, and halloysite. Silica and soda ash are the raw materials needed for producing sodium silicate (Lagaly et al., 2000) which enables formation of the siloxane framework of pivotal importance to holding together the composite. The United States alone produced 12M tons of soda ash in 2018 (Bernhardt and Reilly, 2019). In the United States, 120M tons of sand containing 95% or more silica was produced in 2018, whereas worldwide production reached 300M tons (Bernhardt and Reilly, 2019). Silica sand and soda ash production is forecasted to increase in the coming years. The mass production and easy availability of these materials renders them viable prospects as the fundamental building blocks of printed architectures.

Ongoing research is focused on developing optimal formulations that offer desired temporal profiles of strength development, improved compressive strengths, as well as higher tensile strengths through the incorporation of reinforcement fibers. Detailed characterization of cross-linking reactions and the bonding framework is further underway utilizing synchrotron-based spectroscopy and imaging methods in parallel with vibrational spectroscopy (Horrocks et al., 2017). In parallel, generative design approaches are being used to determine macroscopic structure—function relationships between printed geometries and their response to mechanical loading.

A LOOK TO THE FUTURE

In this perspective article, we have attempted to capture the urgency of affecting a paradigm shift in the construction industry to mitigate the catastrophic environmental impact of conventional concrete production, remedy deteriorating infrastructure, provide accessible dignified habitats to populations in economically disadvantaged nations, and to bring about unprecedented enhancements in productivity. These objectives hinge on advancements in the design and effective utilization of novel structural materials. We propose a roadmap to the use of naturally sourced materials modified to be extrudable, compactable, and cross-linkable with rapid strength gain as is necessary for the construction of load-bearing structures. The design of such soil composites necessitates multiscale design spanning the range from devising multiple cross-linking modes to modification of clay particles, incorporation of fibrous reinforcements, and control of mesoscale porosity. Several examples are illustrated including the harvesting of muskeg soils for road construction in Alberta, Canada, clay-incorporated concrete for oil-well cementing, and the 3D printing of soil composites derived from the modification of burlewash soils. Advancements in the design of naturally sourced construction materials must furthermore be closely matched to the constraints and opportunities afforded by additive manufacturing technologies. Generative design, the search for optimal geometries, further provides a means of obtaining geometry-derived properties with economy of materials use.

Going beyond printing of architectural facades to load-bearing applications compliant with code will require approaching the strength profile of reinforced concrete either through approaches that allow for incorporation of coarse grain aggregates or through methods for embedding of rebar (alternatively, *in situ* printing around pre-assembled re-bar or utilization of dual nozzle constructs that print rebar and concrete). The forms immediately accessible are suitable for walls, roofs, and architectural forms and pave the way for the design of higher strength structures.

The design space of naturally sourced materials is vast and the set of desired functionalities for a viable 3D printable construction material is stringent. Effectively navigating the multivariate design space will require the effective use of statistical learning methods in conjunction with the design of high-throughput materials libraries, development of multiscale models that extrapolate atomistic bonding to macroscopic response, and precise elucidation of the interfacial bonding modes that result from different reaction chemistries. A detailed investigation of such design space nevertheless holds great promise for realizing a new paradigm of "dust to structures."

DATA AVAILABILITY STATEMENT

The datasets generated for this study are available on request to the corresponding author.

AUTHOR CONTRIBUTIONS

AB, MB, UZ, and AP performed laboratory research and contributed to experimental results as well as analysis of data. LE and MD contributed to the LCA discussion. MD, PS, NK, BB, ZR, and SB supervised the research, analyzed data, secured funding, and established research directions. AB, ZR, and SB drafted the article.

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SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/fmats.2020.00052/full#supplementary-material

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