



Innovative concretes provide the ultimate solution for rising construction costs and environmental footprint

By Rouzbeh Shahsavari and Sung Hoon Hwang

Concrete—integral to building today’s infrastructure—is facing high costs that are linked with material processing and overall properties. However, recent technological innovations and nanotechnology advances are enabling new concrete approaches that can relieve the rising cost of global construction activities and related environmental issues.

The global construction industry is expected grow to \$15.5 trillion by 2030. This rapid growth is boosted by numerous ongoing major construction projects, such as Al Maktoum airport in the United Arab Emirates and Grand Paris Express in France, which each have costs exceeding \$30 billion.¹ The “One belt, one road” initiative proposed by China, which plans to connect more than 60 countries via new infrastructures, is expected to receive a total investment of \$1 trillion.² Further, the American Society of Civil Engineers in 2014 reported a renewal program worth \$3.6 trillion for infrastructure in the United States.³

Worldwide cement consumption has drastically increased since 1926 (Figure 1). Overall, owing largely to the aforesaid high-cost construction activities, initial construction costs as well as maintenance and repair costs for infrastructures are rapidly rising. This is now adding unprecedented pressure on the construction industry to take active yet appropriate measures and make significant changes. Another key concern is energy and environmental footprints—current concrete manufacturing is responsible for 8%–9% of global anthropogenic CO₂ emissions and 2%–3% of global primary energy use.

Because concrete is the most common building block of today’s infrastructure, cost issues are highly linked with processing and overall properties of concretes. If concrete structures can be placed in large scale without manual labor and in a timely fashion, significant portions of current construction costs, such as labor, can be negated. Further, stronger and more durable concretes under different types

Capsule summary

RISING COSTS

Worldwide cement consumption has drastically increased in the past century, and construction costs, as well as maintenance and repair costs, are rapidly rising. In addition, energy and environmental costs are high as well—current concrete manufacturing is responsible for 8%–9% of global anthropogenic CO₂ emissions and 2%–3% of global primary energy use.

EMERGING SOLUTIONS

Engineering stronger and more durable concretes that ultimately possesses a longer service life will significantly reduce economic, energy, and environmental costs of cement production. In addition, incorporating industrial byproducts in concrete designs can also help mitigate the CO₂ footprint of cement production.

SHIFTING PATHS FORWARD

Innovative approaches that can relieve the rising cost of global construction activities are now receiving a significant boost from technological innovations and recent nanotechnology advances. The construction industry is undergoing a major paradigm shift, and concrete—the most widely used synthetic material in the world—is at the center of the ongoing shift.

of mechanical stresses that ultimately possesses a longer service life will significantly reduce the economic cost as well as CO₂ and energy footprint stemming from cement production, in accordance with a “do more with less” approach. Also, incorporating industrial byproducts, such as fly ash, ferrous slag, and silica fume in a concrete design can also help mitigate CO₂ footprint, which arises largely from cement production.

All of the aforementioned approaches, which can relieve the rising cost of global construction activities and related environmental issues, are now receiving a significant boost from technological innovations and recent nanotechnology advances. Self-healing concretes, which may have sounded like surreal science fiction only a few years ago, have recently been tested in construction sites with promising success. CeraTech USA (Baltimore, Md.) has commercialized CO₂-free concretes with zero cement by using industrial byproducts. Now it is undeniable that the construction industry is undergoing a major paradigm shift, and concrete, the most widely used synthetic material in the world, is at the center of the ongoing shift.

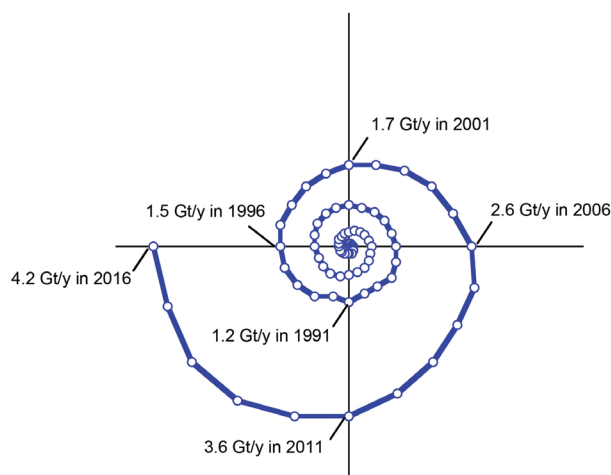
Additive manufacturing offers unique opportunities

Additive manufacturing, or 3-D printing, is a key state-of-the-art technology that will revolutionize the construction industry during the next industrial revolution. Additive manufacturing will negate the need for basic construction tools, including molds and physical framework, thereby drastically reducing material costs. This in turn enhances the degree of design freedom to ensure greater creativity during construction. Further, computer-controlled automation can significantly reduce labor costs, as the number of qualified construction workers has been decreasing over the past years. Automation also guarantees a decreased amount of material waste.⁴

The first attempt to apply additive manufacturing on construction sites was made by Shimizu Corporation in the 1980s, implying that the demand for automation in construction is not something new but existed more than 30 years ago.⁵ In 1997, Pegna implemented laboratory-scale additive manufacturing using sand and Portland cement, illustrating for the first time the prospect of applying automated layer-by-layer fabrication in the construction industry.⁶ The technique alternatively deposited uniform layers of sand and a patterned layer of Portland cement, using steam to cure the cement. The as-created structure with controlled cavities exhibited promising mechanical properties compared to conventional concretes.

Since then, there has been a myriad of successful efforts to

World Cement Production from 1926 to 2016 (Gt/y)



Credit: Dr. Paulo Monteiro, University of California, Berkeley

Figure 1. World cement consumption since 1926.

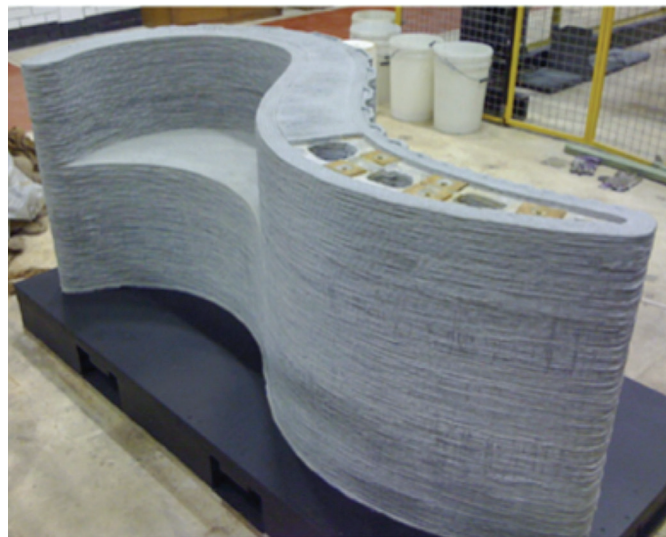


Figure 2. Wall-like artifact created using a concrete printing process.⁵

create small-scale, individual building elements via additive manufacturing. Nevertheless, we are yet to observe ubiquitous on-site application of 3-D printing to construct large-scale houses or high-rise buildings. Contour crafting is one of the major 3-D printing techniques that has been developed for large-scale construction.⁷ A nozzle attached to an automated

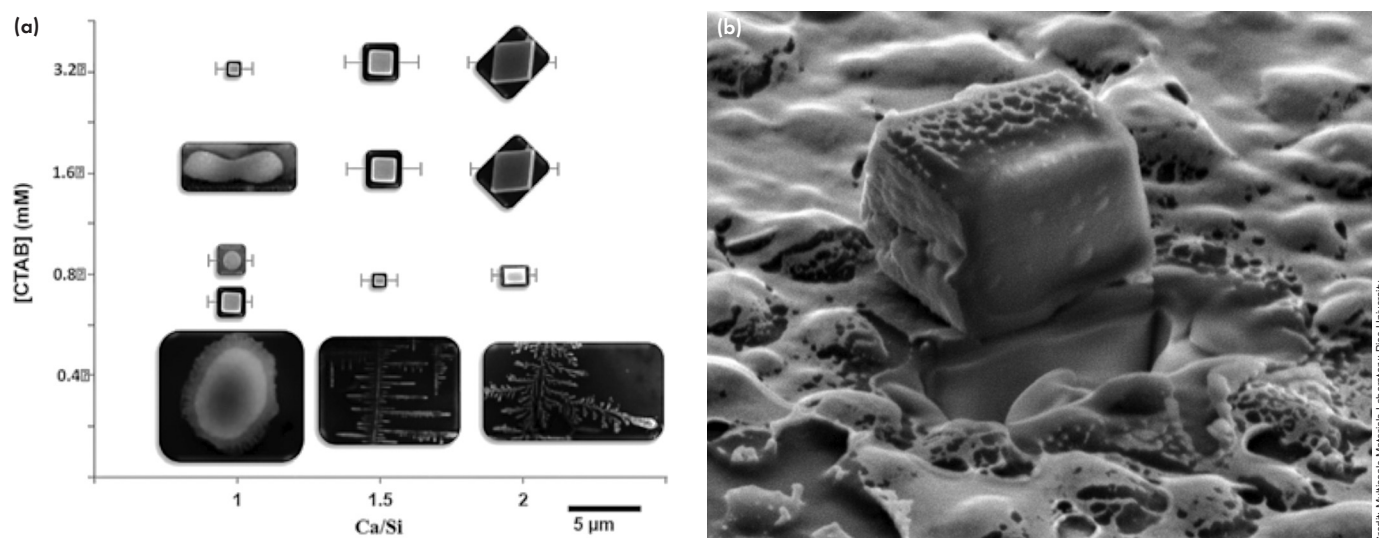


Figure 3. (a) Morphogenesis of cement hydrate. (b) Rectangular C-S-H produced via an ultrasonication-assisted method.

crane extrudes cement paste in a layer-by-layer fashion, with top and side trowels on the crane enabling surface finishing with unprecedented quality. Owing to its advantages, such as ease of transport and preparation of building materials on-site, contour crafting is expected to provide shelters at the site of natural disasters within the next couple of years.⁸

Concrete printing is also an automated technique that carries out extrusion-induced layer-by-layer fabrication using concrete. The technique has been developed and applied at Loughborough University in the United Kingdom to create various architectural pieces with large complexities, such as a wall-like artifact that features 128 total layers and 12 voids for reinforcement (Figure 2).⁹ A major advantage of concrete printing lies in the significantly enhanced degree of design freedom.

Owing to consistent efforts to apply automated additive manufacturing to large-scale construction, 3-D printing in construction is now entering a new era of practical applications. For example, researchers now have employed extrusion-based 3-D printing to create a 512 ft² concrete-based barrack for the U.S. Army.¹⁰ As in contour crafting described earlier, the work involved in situ preparation of concrete materials, thereby confirming the prospect of using the technique to create a protective hut during military operations. Also, Construction Robotics (Victor, N.Y.) recently developed a brick-laying robot, the Semi-Automated Mason, which can lay 3,000 bricks per day—a rate six-times higher than a human worker.¹¹ This robot is expected to replace manual labor within the next couple of years.

Enhancing concrete by improving material properties

Additive manufacturing is concerned with reducing time and cost and endowing construction designs with greater creativity. In other words, it is about improving the “processing” stage of concretes, but it is not linked to improving their intrinsic material properties. To achieve the latter, advances in nanofabrication techniques and nanomaterial characterizations come into play. One definitive strategy to mitigate the environmental footprint and economic cost associated with use of con-

cretes is simply enhancing their mechanical properties, which will in turn extend service life. The question is if modern science, typically known as nanoengineering, can play a critical role in improving the mechanical properties of concretes.

Nanofabrication techniques are typically divided into “top-down” or “bottom-up” approaches depending on reaction pathways. For improving concrete mechanics, bottom-up fabrication, where nanoscale building blocks are assembled to synthesize a larger product, is more relevant. The fundamental, strength-giving building blocks of concretes are calcium-silicate-hydrate (C-S-H), which is present as nanoscale particle aggregations.¹² C-S-H is semi-crystalline, lacking long-range order, and occupies 60% of total mass of the cement paste.¹³ It also is non-stoichiometric with varying proportions of calcium, silicon, and hydrate ions. No universal structure exists for C-S-H up to this point, but several different models based on crystalline analogs have been proposed.^{12,14}

The general notion is that the C-S-H structure resembles that of a layered mineral called tobermorite, which comprises parallel silicate chains with calcium ions between them. Considering that C-S-H is the major strength provider for concretes, there are ongoing efforts to synthesize C-S-H in the laboratory, tune its properties at a microscopic scale, and assemble it via bottom-up engineering to ultimately develop mechanically-enhanced concretes.

For example, Ca/Si molar ratio is an important chemical property that defines C-S-H. Pelisser et al.¹⁵ synthesized C-S-H with different Ca/Si molar ratios and performed nanoindentation, a technique often employed to probe mechanical behavior of nanomaterials. The authors concluded that a lower Ca/Si molar ratio enhances nanoscale and microscale mechanical properties.

In addition to chemical properties, structural morphologies of C-S-H can be fine-tuned to improve mechanics at multiple length scales. Our group recently attained comprehensive morphological control of C-S-H, whose shapes can be systematically varied from irregular to dendritic and, finally, to well-

defined cubic (Figure 3).¹⁶ Naturally-formed calcite particles form cubic seeds, providing facile routes for nucleation and semi-epitaxial growth of C-S-H. Subsequent mechanical testing proved that the well-defined cubic morphology benefits mechanical properties at multiple length scales from a single particle to assembled states. This suggests that controlling the morphology of concrete nanoparticles can ultimately induce enhanced mechanical properties of concretes.

Because the major shortcoming in mechanical properties of concretes is high brittleness, research has also applied biomimetic approaches based on the structural ensemble of organic and inorganic components to C-S-H. Inspired by natural materials such as nacre, where a small fraction of organic components remarkably enhances mechanical toughness,¹⁷ a significant number of efforts have been directed towards combining inorganic C-S-H with soft, organic polymers.¹⁸ In fact, adding polymer-based plasticizers during concrete mixing is a common practice to enhance workability.

Orozco et al.¹⁹ identified the nature of interaction between C-S-H and polycarboxylate-based superplasticizers with silyl functionalities and found that the two components interact via combination of ligand-type interactions and covalent bonding. Nevertheless, such C-S-H-polymer composites naturally created by adding polymer-based plasticizers to wet concrete mixtures do not contain an ordered microstructure, where C-S-H and polymer are structurally coordinated with each other in an orderly manner.

The first successful attempt to intercalate polymer within the basal spacing of synthetic C-S-H was made by Matsuyama, who successfully incorporated polyvinyl alcohol during the precipitation process.²⁰

Power of modelling

So far, only experimental techniques for fine-tuning properties of C-S-H have been discussed. In fact, theoretical simulations have played immense roles in decoding the relationship between structure, composition, and mechanical properties of C-S-H over the past decade. Our group performed atomistic simulations on tobermorite and jennite, two mineral analogs of C-S-H, to ascertain their anisotropic mechanical properties.²¹ We found that unlike the common intuition that a layered direction serves as the weakest link in layered crystals, such as tobermorite and jennite, inclined regions forming a hinge mechanism are the softest parts.

Later we employed a combination of three distinct types of modelling techniques—atomistic simulations, Monte Carlo, and molecular dynamics (MD) techniques—to propose for the first time a realistic structural model for C-S-H.¹² The authors started by applying a Ca/Si ratio (1.7) and density (2.6 g/cm³) of C-S-H acquired from experiments as compositional constraints for modelling. This solved potential accuracy issues associated with using perfectly crystalline tobermorite or jennite as a model system for C-S-H, since their Ca/Si ratios deviate from experimental values. Qomi et al.²² created an impressive database of atomic configurations for C-S-H with specific defect attributes, each of which results in distinct mechanical

properties. This in turn allows selection of structural configurations with optimum mechanical properties.

MD is a computational methodology particularly useful for simulating an ensemble of thousands of atoms, a scale that cannot be modelled using first principles quantum simulations. Therefore, MD has been particularly useful for simulating the macroscale response of C-S-H to a variety of mechanical stresses.^{23–25} Tao et al.²⁶ recently employed tobermorite as a model system for C-S-H and studied its global deformation under tension, compression, and shear loading as well as local deformation under nanoindentation. The authors found that mechanisms for global deformation are governed by displacive and diffusive mechanisms, while local deformation under nanoindentation exhibits size-dependent mechanical behavior.

Further, Zhang et al.²⁷ performed more than 600 MD simulations using C-S-H-FF potential²⁸ and found that the inclusion of portlandite particles or nanoscale voids induces higher toughness for C-S-H, thereby proposing useful experimental strategies to enhance concrete toughness. The same groups of authors performed several simulations on dislocations within cement crystals^{29–31} and found that screw dislocations within the layered structure of C-S-H can serve as a bottleneck during shear loading, thereby impeding interlaminar gliding (Figure 4).³² This relieves stress and increases toughness in contrast to the conventional, logical perception that a defect is detrimental to mechanical properties of a material.³³

Cracking down on concrete with self-healing strategies

All above strategies are important recipes to improve the mechanics of concrete and “to do more with less”—thereby reducing concrete’s environmental and energy footprints. Another way is to reduce maintenance costs. This can be achieved by developing self-healing concretes. Since microcracks are the major culprit for increased water and chloride penetration, they exert detrimental effects on concrete integrity. Further, small-scale cracks, if left untreated, can rapidly propagate and coalesce together, ultimately leading to catastrophic failure of the entire structure.

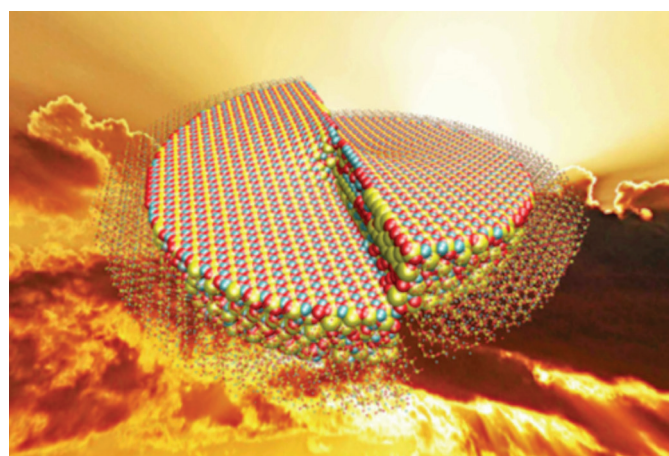


Figure 4. Illustration of a screw dislocation created in a computational model of tobermorite.³²

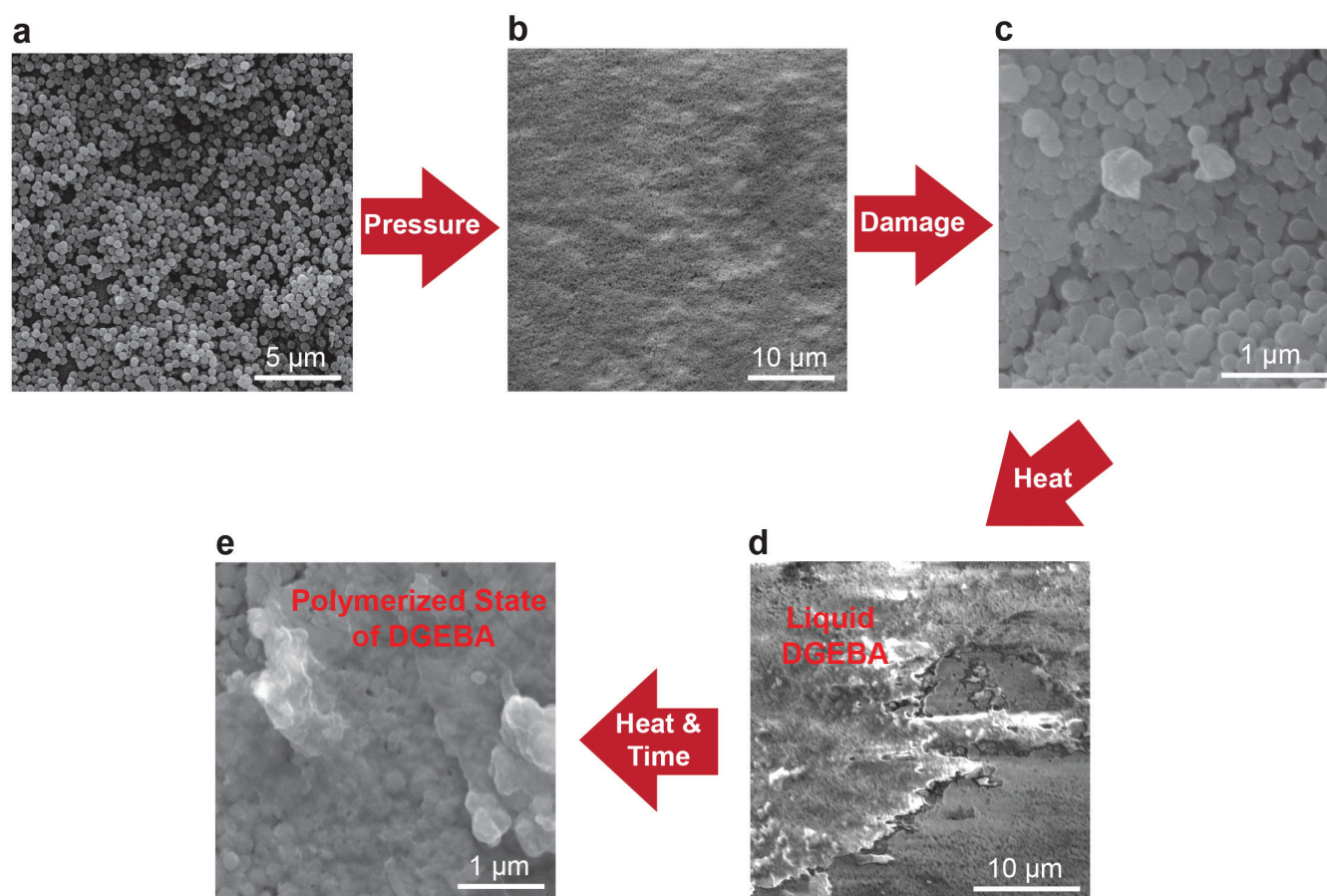


Figure 5. Sequential mechanism of self-healing. (a) Mixture of sealant-loaded calcium-silicate porous nanoparticles. (b) Top-most surface of the sealant-loaded tablet, created using pressure-induced assembly. (c) Nanocrack created under flexural mechanical loading. (d) Release of liquid epoxy resin DGEBA onto top-most surface at the initial stage of heating. (e) Polymerized DGEBA on the top-most surface.⁴³

Due to inherent high brittleness, concretes are highly susceptible to formation of microcracks, which favor ingress of water and other harmful ions, including chloride and sulfates. Penetration of the aforesaid chemicals can be a major cause of corrosion of reinforcing bars within concrete structures. In light of potential dangers brought upon by microcracks, researchers have proposed a variety of self-healing strategies that enable concretes to self-detect and independently undergo healing processes.

The concept of self-healing concretes can be divided into two types of mechanisms at large. One mechanism is autogenous healing, which relies on natural healing of concrete in the presence of water.³⁴ Examples include hydration of unreacted cement particles and carbonation of calcium hydroxide to produce calcium carbonate minerals. Also, partial replacement of cement with fly ash

can trigger similar healing processes, as unreacted fly ash particles can react with water at late ages.³⁵ The other type of mechanism is autonomous healing, where external agents are incorporated into concretes and actively participate in the healing process.

Since autogenous healing is often constrained to cracks with limited widths (<100 μm) and is only effective when sufficient moisture is present, a greater proportion of recent research efforts has been devoted to autonomous healing.³⁴ The idea of autonomous self-healing for concretes was initiated more than 20 years ago, when Dry et al.³⁶ employed hollow glass tubes filled with healing agents as extrinsic agents. The design ensured that formation of a crack fractured the glass tubes, mimicking self-healing of human skin where a complex network of blood vessels exists underneath the skin.

Since then, one of the most classical and most-attempted approaches has been a capsule-based approach, where nanocapsules or microcapsules containing organic self-healing agents are incorporated into concrete. Capsules in pathways of crack propagation rupture, triggering release of the capsules' inner contents and initiating self-healing processes. Outer shells of the capsules are typically composed of polymer-based materials, including silica, poly(urea-formaldehyde), poly(melamine-formaldehyde), and poly(urethane). Core regions are occupied by self-healing agents, including sodium silicate solution, epoxy resin, and methylmethacrylate.³⁷

Nevertheless, this capsule-based strategy suffers from critical shortcomings, such as intrinsic properties of matrix materials compromised upon the addition of large volumes of capsules and low survival rate due to vigorous mixing

and highly alkaline conditions.

Some researchers have avoided those challenges by developing self-healing coatings for concrete surfaces or steel rebar. Chen et al.³⁸ developed a self-healing epoxy coating for concrete rebar by encapsulating tung oil in poly(urea-formaldehyde) microcapsules, because tung oil polymerizes simply upon exposure to air. The coating not only provides enhanced resistance against corrosion, but also induces better bond strength between the rebar and concrete. Song et al.³⁹ also developed a similar protective, self-healing coating for the surface of concrete by encapsulating photoresponsive monomers and initiators in urea-formaldehyde capsules. The capsules successfully incorporate into an epoxy coating matrix and polymerize after induction with natural UV light from the sun, opening the door towards new opportunities for self-healing capsules in concretes.

In addition to capsule-based strategies, bacteria-based self-healing concretes are another proposed approach. Jonkers and colleagues⁴⁰ mixed alkaline-resistant bacterial spores along with calcium lactate into cement pastes and showed that the bacteria can catalyze formation of calcium carbonate minerals, which can plug cracks. Calcium carbonate is first formed from the metabolic conversion of calcium lactate catalyzed by bacteria. This conversion produces carbon dioxide as a byproduct, which further reacts with portlandite to generate more calcium carbonate minerals.

Despite novelty of a concept that exploits the chemical environment of concrete rich in portlandite, this bacteria-based approach faces similar problems as polymer-based capsules. A large proportion of bacterial spores are destroyed by rigorous mixing and highly alkalinity. Consequently, numerous follow-up investigations have focused on immobilizing bacterial spores in a protective medium prior to addition to cement paste. For example, Wiktor et al.⁴¹ immobilized bacterial spores in porous expanded clay and showed that the self-healing system can heal up to 0.46-mm-wide cracks in concrete, while normal concrete can only heal 0.18-mm-wide cracks. Since then, bacterial spores

have been encapsulated in a variety of core-shell type capsules, including silica gel, polyurethane, and microcapsules, to enhance their viability inside concretes.⁴²

A myriad of other unique self-healing approaches has been proposed and tested. Very recently, our group devised a novel strategy to synthesize strong, tough, and self-healing cement⁴³ integrating soft and hard components within scaffolded universal porous building blocks, mimicking naturally strong and tough materials.⁴⁴ The composite system is comprised of calcium-silicate porous nanoparticles with unprecedented monodispersity over particle size, particle shape, and pore size, which facilitate effective loading and unloading with organic sealants—resulting in a 258% and 307% increase in indentation hardness and elastic modulus of the composite, respectively. Further, heating the damaged composite triggers controlled release of nanoconfined sealant into the surrounding area, enabling recovery in strength and toughness (Figure 5).

Prompted by the advent of novel strategies, self-healing concretes have recently begun taking major steps towards applications on actual construction sites. In 2015, one site integrated three distinct self-healing strategies—adding shape memory polymers, embedding a blood vessel-like network of healing agents, and incorporating extrinsic carriers containing organic healing agents or bacteria—into a single system and tested its on-site viability.⁴⁵ Initial successful on-site trials have led to a more recent project that received an investment of £4.7 million.⁴⁶

New recipe: Adding recycled industrial waste

Another approach to reduce production of Portland cement is addition of recycled industrial wastes, including fly ash, iron and steel slag, rice husk, and silica fume. Since they are byproducts of the world's biggest manufacturing, food, and energy industries, these industrial waste products have large availability and resultant low costs.

U.S. production of ferrous slag, a major byproduct of the iron and steel manufacturing industry, reached 20–50 million tons in 2016.⁴⁷ During the same

time, worldwide production reached 460–600 million tons. Ferrous slag is rich in calcium oxide and silica and can be used as an aggregate or partial replacement of cement.⁴⁸ Further, Osborne et al.⁴⁹ found that replacement of 70% (by mass) conventional Portland cement with blast-furnace slag enhances chemical resistance of the resultant concrete against sulfates, chlorides, and water. Slag also reduces heat release as well as rate of temperature increase, thereby decreasing the chance of detrimental thermal cracking.

In addition to use as an alternative binder, its exploitation as a new type of aggregate also offers additional benefits compared to conventional counterparts, such as limestone aggregates. Slag-based aggregates are particularly effective in enhancing fracture toughness of concretes and reducing occurrence of alkali-silica reactions, which degrade mechanical integrity of concretes.⁴⁹ Slag can also serve as useful aggregates for stone mastic asphalt mixtures, thereby improving high-temperature properties and enhancing resistance to low-temperature cracking.⁵⁰

Fly ash is another coal combustion byproduct with proven capabilities as a binder material. As with slag, its annual production exceeds 100 million tons in India and China, offering opportunities for use as a low-cost, environmentally friendly binder.⁵¹ It is already highly established in construction markets as a supplementary cementitious material, which partially replaces Portland cement and offers a plethora of benefits, such as improved durability, enhanced late-age strength, and reduced bleeding.⁵² Because the current replacement level is ~20%–30% by mass in a typical concrete design, there are active ongoing research efforts to achieve 100% replacement to completely solve environmental concerns associated with cement production.⁵³ CeraTech USA received an Environmental Production Declaration for its trademarked product ekkomaxx in 2014 for achieving virtually zero carbon footprint with 95% fly ash and 5% liquid binder.⁵⁴

The future of concrete

Owing to global construction activities with high capital costs and rapidly

rising construction markets, including China and India, rising construction costs and CO₂ footprint have evolved into societal concerns. Scientists now realize that the major key to solving those problems lies in enhancing properties of concretes, the most commonly used infrastructure material. In this context, decade-long efforts are finally reaping fruits, with 3-D printing successfully constructing large-scale houses and self-healing concretes being tested on actual construction sites.

Nevertheless, there remain limitations that need to be overcome to ensure those successes eventually evolve into ubiquitous future applications. For example, there exists a large knowledge gap between properties of C-S-H produced on a laboratory scale and macroscale behavior of hydrated cement. C-S-H is indeed the major product of cement hydration, ultimately responsible for mechanical strength of concretes. However, the overall properties of concretes do not rely solely on the behavior of C-S-H, but arise from a complex interplay between multiple hydration products, including but not limited to portlandite, C-S-H, and calcite. Therefore, there should be constant efforts to apply state-of-art nanomaterial characterization techniques to decode the link between microscopic behavior of C-S-H and real phenomena observed at a macroscale of cement hydration.

This also signals the importance of inventing new simulation techniques to correctly describe the interplay between various hydration products at a larger scale than is possible with the likes of atomistic simulations or MD. In this context, current global trends in big data, computational materials science, and artificial intelligence (e.g., machine learning), when combined with advanced experiments, can perhaps provide the most promising strategy to streamline the processing-structure-property design landscape. Broadly, mimicking the promises of emerging hybrid nanomaterials,⁵⁵⁻⁵⁷ other strategies such as creating hybrid cementitious materials,⁵⁸⁻⁶¹ also will be important to create innovative multifunctional construction materials (e.g., with high

thermal, electrical, and mechanical properties), offsetting cost and environmental footprint of secondary materials, thereby making innovative concretes an ultimate solution to rising construction costs and environmental concerns.

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