Architected Cementitious Cellular Materials: Peculiarities and opportunities

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Microlab

Conventionally, the properties of cementitious materials are tailored by a simple but efficient method: mixture proportion design. For a given cementitious mixture, the chemical and physical properties of cementitious materials have already been determined. Consequently, the mechanical performance of the hardened cementitious material is determined. This is attributed to the nanoscale, microscale and mesoscale structures formed during the hydration process which are also dictated by the mixture proportion. Apart from this traditional methodology, a novel approach to tailor materials mechanical performance by combining architected cellular structure with certain constituent materials is introduced in this work. Inspired by cellular polymer materials and cellular ceramic materials which show enhanced properties comparing to conventional polymers and ceramics, creating cementitious cellular materials is also assumed to be a promising research direction. Specifically, geometrical features of promising cellular structures and their corresponding mechanical performance is reviewed. Potential processing methods for obtaining such cellular structures using cementitious materials are discussed. In addition, probable requirements on cementitious mixture for the cellular structure are analyzed.

Key words: Cementitious materials, architected cellular materials, 3D printing

1 Introduction

Cementitious materials are the most widely used construction materials in the world due to their excellent properties and relatively low cost. Since their invention, there has been a simple and efficient method to tailor the properties of cementitious materials: mixture design. It is well known that material properties are dictated by their microstructures. For cementitious materials, after a curing process in which hydration takes place, the spatial distribution of the mixture ingredients is generated. In this sense, the micro or meso structure of cementitious materials is determined and, therefore, mechanical properties of the cementitious materials are dictated. In the past several decades, intensive efforts have focused on configuring or modifying the micro or meso structure of cementitious materials to improve their performance. For instance, optimizing the packing density (Amario, Rangel, Pepe, & Toledo Filho, 2017; L. G. Li, Lin, Chen, Kwan, & Jiang, 2017; Sun, Wang, Gao, & Liu, 2018), modifying the pore structure (Keulen, Yu, Zhang, & Grünewald, 2018; X. F. Wang et al., 2018; Zhang, Islam, & Peethamparan, 2012), modifying the air void structure (Özcan & Emin Koç, 2018; M. Qiao et al., 2017) and introducing new phases as reinforcement (Lu, Li, & Leung, 2018; Yu, Lin, Zhang, & Li, 2015).

Besides this traditional approach, owing to the rapid development of digital fabrication technology, intentionally distributing constituent cementitious materials with certain architected cellular geometry may enable combining the “material behavior” of cementitious mixtures with “structural behavior” of architected cellular structures. This novel approach has been adopted in many types of materials, such as polymers, metals and ceramics, to created metamaterials (a metamaterial is any material engineered to have a property that is not found in naturally occurring materials) with superior properties (Buckmann et al., 2012; Greaves, Greer, Lakes, & Rouxel, 2011; Ren, Das, Tran, Ngo, & Xie, 2018; Valdevit, Jacobsen, Greer, Carter, & Pollock, 2011; Zheng et al., 2018). However, this new approach has not been applied on cementitious materials yet.

The term “cellular structure” normally refers to specially designed, tailored or constructed structures, regardless of the scale, ranging from nanoscale up to macroscale. In cementitious materials, as the structures below mesoscale are already determined by the mixture proportions, the definition of “structure” and “material” would be ambiguous if not clear definition is given. In case the system is only comprising a limited number of cells or elements, the system seems to be simply a “structure” instead of “material”. Then it seems reasonable to argue that the response of this structure should not be considered as material behavior. However, considering the approach of “architecting”, even it is a structure which consisted of, in an extreme case, only one unit cell, the global response is still a combination of the structural response of the unit cell and the constituent material. In this sense, it is valid to tailor the mechanical response of the global system by architecting the structure and the constituent, respectively. This complies well with the research topic of architected cementitious materials. For a global system consisting of a large number of cellular units behaves as a monolithic material, same as conventional solid materials. It is not difficult to recognize that the global response of the system should be seen as “material” behavior.

Furthermore, when discussing mechanical properties of cementitious materials, the size or the scale is of great importance. The cementitious matrix is already a composite which consist of microscopic grains. In architected cementitious cellular materials, the dimensions of each cellular unit should be at least greater than a couple of these grains. Considering the particle size of Portland cement (normally ranges within 10 ~ 100 μm), the minimum dimension of a cellular unit should be correspondingly at least 40 ~ 400 μm if cement paste is used, such that the characteristics of cementitious material can be properly presented. Actually, in most reported cases (Moini, Olek, Youngblood, Magee, & Zavattieri, 2018; Seyed Mohammad Sajadi et al., 2019), the dimension of the unit cells of cellular cementitious materials is not smaller than several millimeters. Herein, the cellular structure only refers to structures consisting of units within a millimeter up to a couple of centimeters; In this sense, potential cementitious cellular materials can be easily processed by existing techniques such as traditional mold casting or, by more advanced methods such as 3D printing techniques. Therefore, throughout this work, the term "*cellular structure*" refers to the system constructed by the cellular units at the scale not smaller than millimeters, independent from cementitious mixture propositions. On the other hand, the term "*constituent material*" refers to the substance used to form the unit cells of the cellular structure.

In this literature review, possibilities of several types of cellular structures to be adopted by cementitious materials to enhance physical or mechanical properties are introduced, as well as the processing methods related to the construction of the cellular structures. Features and requirements on architecting the constituent materials for these cellular structures are also discussed.

2 Architecting cellular structures

*2.1 **Increasing porosity***

In contrast to continuum materials, the most commonly referred cellular materials are highly porous and consist of periodic or randomized cells. In the field of cementitious materials, conventional foam concrete is the most frequently studied cellular material. Containing high content of air voids, foam concrete has significantly lower thermal conductivity (approximately 0.05 ~ 0.7 W/m·K (Amran, Farzadnia, & Abang Ali, 2015; Chica & Alzate, 2019)) than conventional concrete (approximately 1.6 W/m·K at 2200 kg/m^3 (Amran et al., 2015)), making it a good choice to be used as thermal insulation construction material. However, foam stability is one of the main concerns that has to be addressed. As the foam stability is very sensitive to the ingredients in cementitious mixtures, the foam structure is highly dependent on the mixture design. Therefore, it is almost impossible to independently optimize the cellular structure and the cementitious mixture in foam concrete.

Contrary to conventional foam concrete, the ability of maintaining and architecting cellular structure makes it possible to combine an optimized *structure* and an optimized *cementitious mixture*. Among numerous types of cellular structures. One of the most extensively studied structures is the lattice structure. The general concept of a lattice structure is defined as "*a cellular, reticulated truss or lattice structure made up of a large number of uniform lattice elements*" (Fleck, Deshpande, & Ashby, 2010). Accompanied by high porosity (normally mentioned as low relative density in the context of lattice structures), lattice-type structures have been found to have outstanding mechanical properties. For perfect lattices without out defects, stiffness and strength are closely related to their relative density and nodal connectivity. For instance, as shown in Figure 1, the stiffness of the two-dimensional lattice is highly dependent on the lattice type: those with higher nodal connectivity also have higher stiffness (Elsayed & Pasini, 2010; Fleck et al., 2010). A similar trend holds for the shear modulus of lattice materials (Elsayed & Pasini, 2010). Furthermore, when joints of the lattice elements are considered, the stiffness of the lattice system would be even higher due to the nodal stiffening effect (Dong & Zhao, 2018; Luxner, Stampfl, & Pettermann, 2005). The ability of achieving high relative stiffness may give cementitious lattices high deformation resistance which helps maintaining structural integrity if used in construction practice.

Strength might be a critical concern in cementitious lattices. In terms of tensile response, the tensile strength of lattice materials scales with its relative density as (Fleck et al., 2010):

$$\frac{\sigma_L}{\sigma_{TS}} = C\,\bar{\rho}^{c} \tag{1}$$

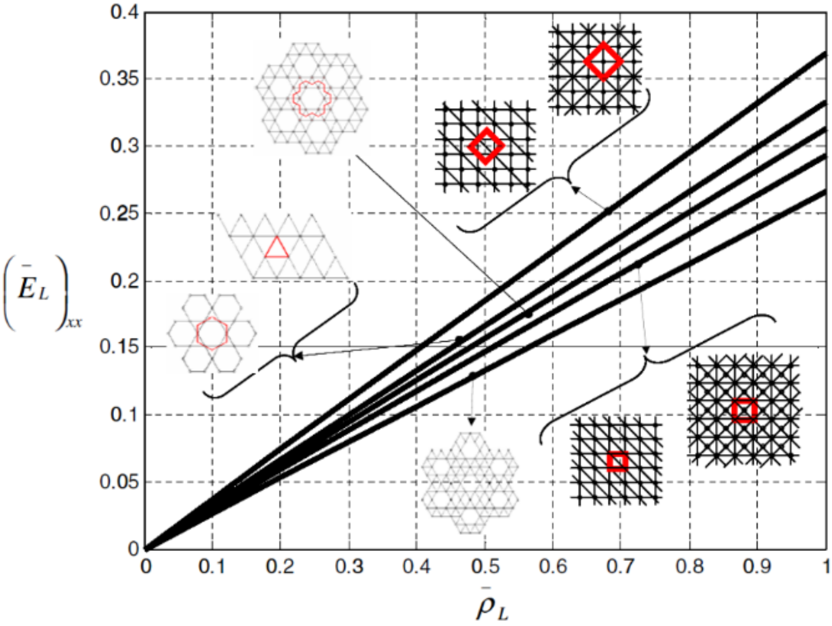
in which

σ_L ………….. is the tensile strength of the lattice material

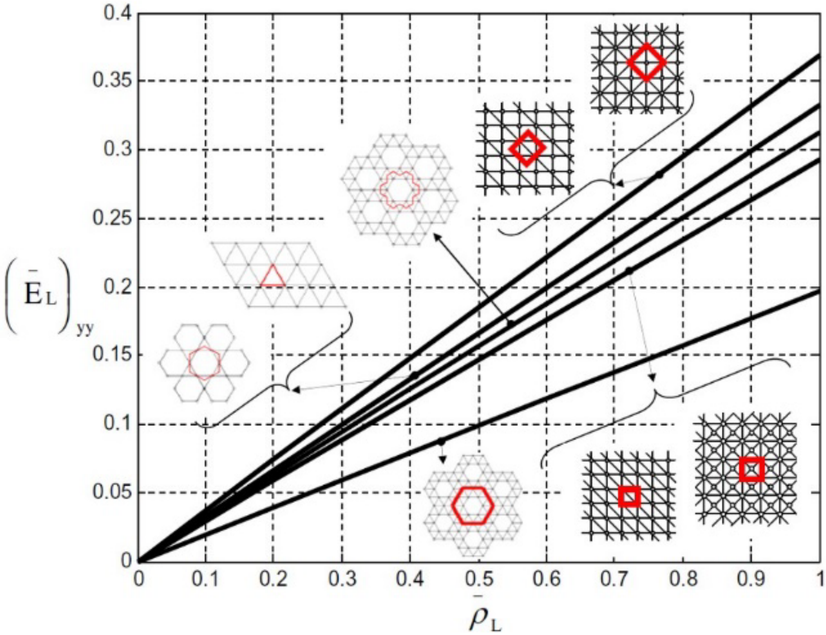
σ_{TS} …............... is the tensile strength of the constituent material

$\bar{\rho}$ ……............. is the relative density

C, c ………….. are structural constants which depend on the nodal connectivity



(a) E_{xx} direction



(b) E_{yy} direction

Figure 1. Relative Stiffness of 2D lattices, reprinted from (Elsayed & Pasini, 2010)

Even adopting a lattice with high nodal connectivity, for instance a fully triangulated lattice, the tensile strength scales with a magnitude of $0.3\,\bar{\rho}\sigma_{TS}$ (C and c equal to 3 and 1/3, respectively). Considering the well-known low tensile strength of cementitious materials, the tensile strength of cementitious lattices would be a primary issue to be addressed. In most cases, σ_{TS} of non-reinforced cementitious material rarely reaches 10 MPa, then σ_L of triangular cementitious lattice with 0.5 relative density would be only 1.5 MPa. In most cases, then, reinforced cementitious materials would need to be used as the constituent material. In (Aghdasi et al., 2019), ultra-high-performance cementitious materials (UHPC) reinforced by PE fibers were used as constituent material with flexural strength of 17 MPa such that the flexural strength of the cementitious lattice reached 10 MPa.

In compression, the strength of the cementitious lattice is highly dictated by local tensile failure of the lattice elements. As reported in (Wu, 2021), according to numerical simulation results, tensile stress appears in the lattice elements when loaded in compression. Cracks initiate from locations with high tensile stress concentration (see Figure 2). Therefore, the compressive strength of cementitious lattices is relatively low (around 0.6 MPa). Similar results were also found in (L. Li et al., 2020; Nguyen-Van et al., 2020; Song et al., 2021). The highest reported value of non-reinforced cementitious lattices is achieved by an octet structure at an approximately $\bar{\rho} = 0.65$ with a compressive strength of 11 MPa (L. Li et al., 2020). Even constituted by reinforced cementitious materials UHPC (see Figure 3) which has compressive strength of 144 MPa, the corresponding cementitious octet lattice only has compressive strength of 22 MPa ($\bar{\rho} = 0.52$). which is still not sufficient for most load bearing purposes.

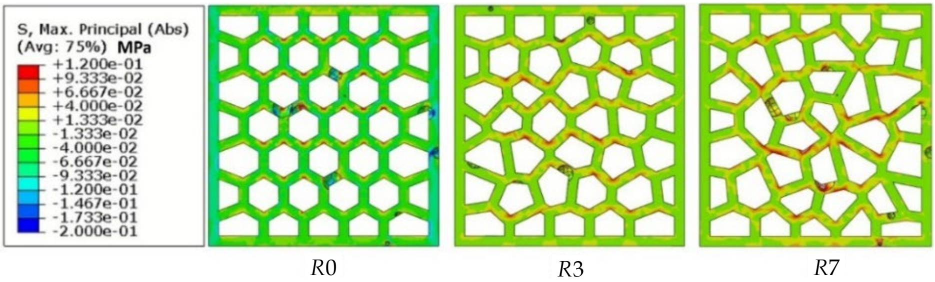


Figure 2. Simulated tensile stress distribution of cementitious lattice loaded in uniaxial compression, reprinted from (Wu, 2021)

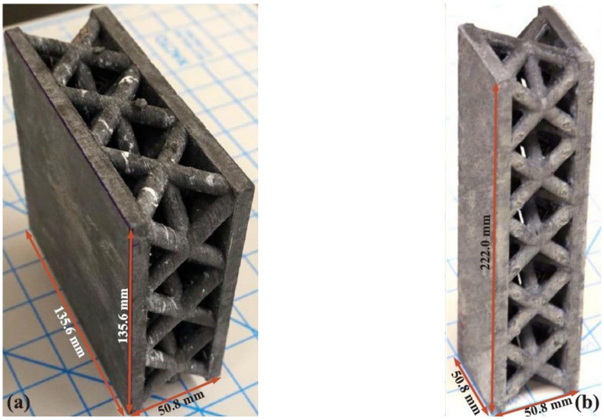


Figure 3. Cementitious octet lattice constituted by UHPC, reprinted from (Aghdasi et al., 2019)

In addition, lattice systems with defects, imperfections or non-uniformities are abundant in nature (Gibson, 2005; Keaveny, Morgan, Niebur, & Yeh, 2001) and artificial materials (Herrmann, Hansen, & Roux, 1989; Tanmoy Mukhopadhyay & Adhikari, 2017; Takano et al., 2017). These defects in the lattice system crucially influence mechanical performance of lattice materials, especially the strength of brittle or quasi-brittle materials. Even if the lattice structure is properly tailored, the possibility of introducing defects may be the main drawback which limits the mechanical performances of the designed lattices.

2.2 ***Modifying damage resistance***

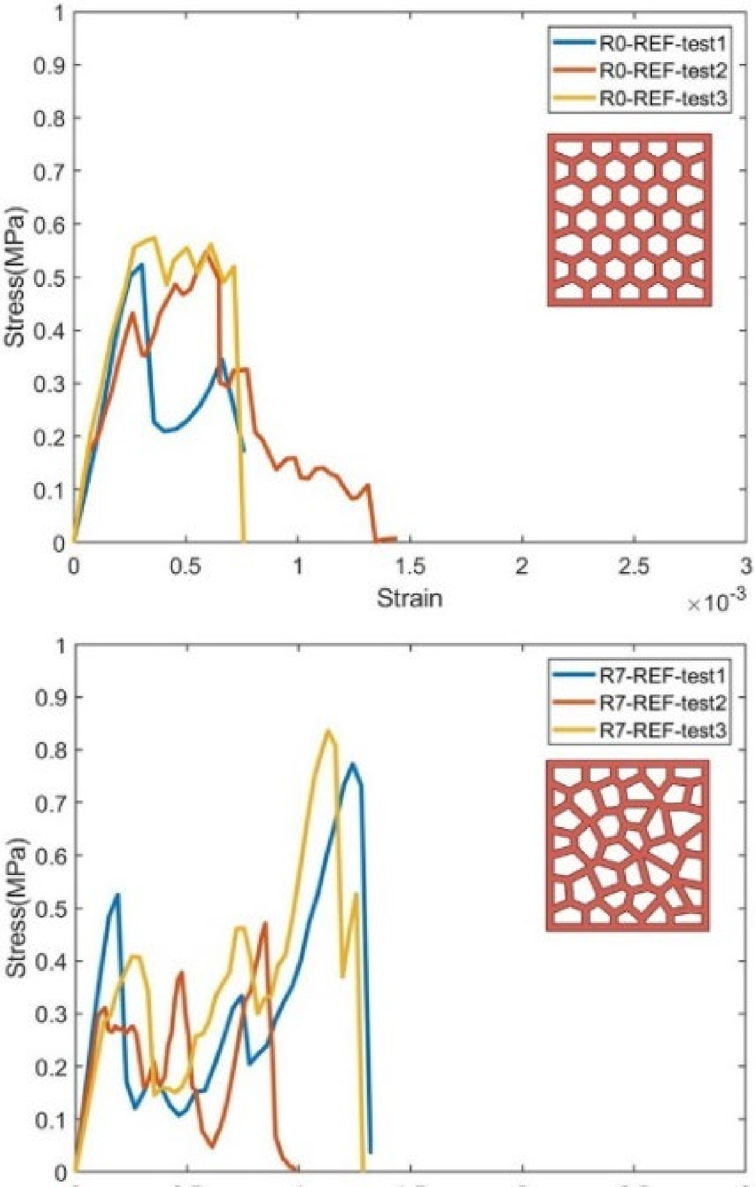
One main drawback of plain cementitious material is their brittleness, which is determined by the physical and chemical nature of cement hydration process and cement hydrates. By properly architecting the cellular structures, it may be possible for cementitious cellular materials to have enhanced damage resistance. In theory, the damage resistance is dependent on the fracture toughness of cellular materials. In pure tension and shear, it is clear that cellular structures with higher fracture toughness which can be achieved by architecting cellular structure is more damage resistant. For instance, a randomized honeycomb exhibits higher KII comparing to regular hexagonal honeycomb of the same relative density (Romijn & Fleck, 2007).

Comparatively, in compression, the mechanism of damage resistance is much more complicated. On one hand, the compressive fracture process involves both tension and shear fracture. Even for continuum materials it is difficult to isolate the contribution of each fracture mode on the overall fracture resistance. On the other hand, for cellular materials, compressive fracture process often refers to a crushing process during which the interaction between the cellular units has to be considered. Then the strategy of enhancing compressive damage resistance may not be limited to increasing the fracture toughness values. This of course raises high interest for scientific research. Several studies have already shown promising results. As reported by Wu (Wu, 2021), cementitious cellular composites with a highly randomized Voronoi structure show higher fracture resistance than regular honeycombs made using the same cementitious constituent material. The stress-strain response (see Figure 4a) exhibit multiple peaks which indicates an element-wise damage mechanism. This result implies that it is more difficult for cracks to propagate in randomized cellular structures. A similar results was also found by Pham (Pham, Liu, Todd, & Lertthanasarn, 2019), who designed a hybrid lattice structure inspired by the alloy crystals shows enhanced compressive strength and fracture resistance. Although cementitious materials were not used in (Pham et al., 2019), the mechanical behavior is in principle similar. As shown in Figure 4b, a regular lattice structure shows brittle compressive damage, which is sharply in contrast to the compressive behavior of lattice with meta-grains (lattice structure with the same orientation). In a recent study of Nguyen-Van (Nguyen-Van et al., 2021), cementitious cellular material with a fractal-like hierarchical structure was studied. According to their numerical simulations, the damage resistance of the cementitious cellular materials is dependent on the order of fractal: higher fractal order gives higher damage resistance. In these reported cases, cracking is either hindered, deflected or arrested by the architected cellular structure, which results in increased damage resistance.

2.3 *Enhancing deformability*

As a construction material, high stiffness of cementitious materials allows them to resist deformation which helps to maintain structural integrity. Nevertheless, unlike the cementitious materials used for common structural elements, in some specific engineering applications such as impact resistant structures (Madke & Chowdhury, 2020) and vibration mitigation structures (Chen, Wang, & Ma, 2020; Duc, Seung-Eock, Cong, Anh, & Khoa, 2017), materials with high deformability are required. Conventional cementitious materials are easily damaged undergoing a minor strain. By architecting the cellular structure, high deformability might be achieved by cementitious materials.

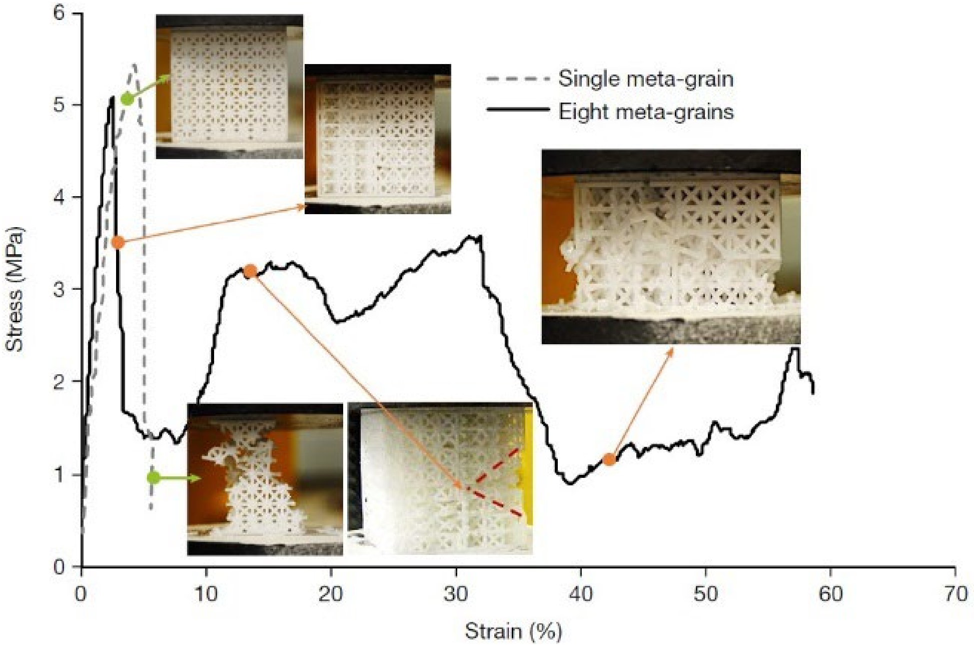
One promising structure is structures potentially exhibit auxetic behavior. The term “auxetic”, coined by Evans in 1991 (Ken. E. Evans, 1991), refers to a material possessing a



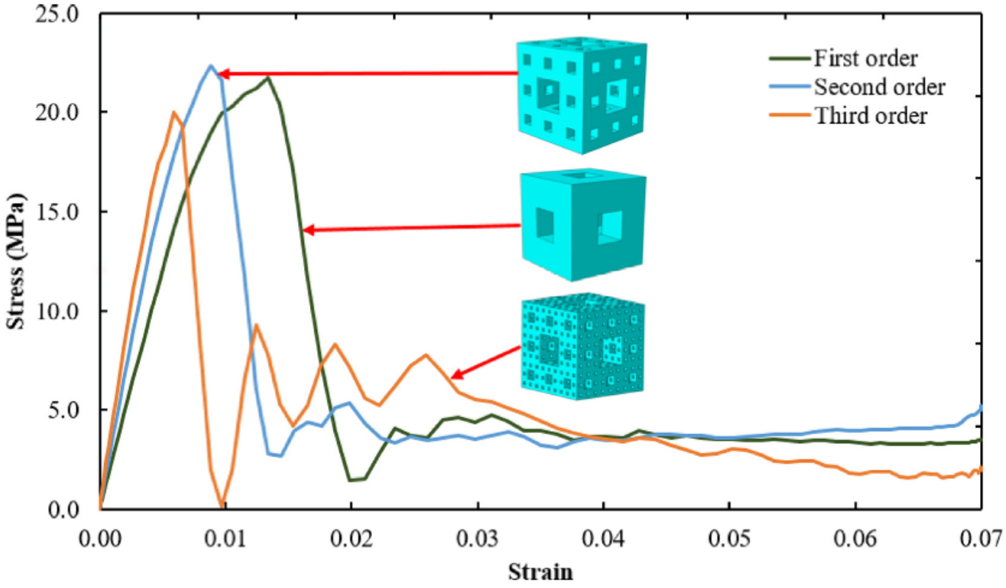
(a) Cementitious Voronoi structure

Figure 4. Architected cellular materials with improved damage resistance

Reprinted from (Nguyen-Van et al., 2021; Pham et al., 2019; Wu, 2021)



(b) Alloy crystal inspired structure



(c) Fractal-like structure

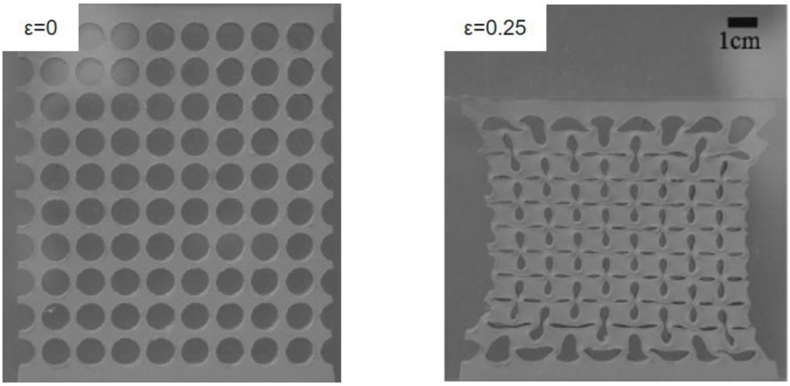
Figure 4. continued

negative Poisson's ratio. This means that the material exhibits lateral contraction or expansion when compressed or stretched vertically, respectively. This unusual behavior gives auxetic materials extraordinary mechanical properties: enhanced indentation resistance (Coenen & Alderson, 2011; Hu, Zhou, & Deng, 2019; Photiou, Prastiti, Sarris, & Constantinides, 2016), high specific energy absorption (Cheng, Scarpa, Panzera, Farrow, & Peng, 2019; Qi et al., 2017; Y. Wang, Zhao, Zhou, Gao, & Wang, 2018), and high shear resistance (Cheng et al., 2019; Ju & Summers, 2011). In cellular materials, auxetic behavior is usually achieved by special deformation mechanisms. Bertoldi (Bertoldi, Reis, Willshaw, & Mullin, 2010) created two-dimensional porous cellular structures consisting circular holes. Under vertical uniaxial-compressive load, elastic instability was introduced to the cellular structure. In terms of stress-strain response, the stress increases as the external load continues until a critical point (Bertoldi, Boyce, Deschanel, Prange, & Mullin, 2008). Pattern transformation was observed afterwards accompanied by negative Poisson ratio effect, see Figure 5a. This unique deformation process ensures high deformability of the cellular material with an auxetic structure. In a more recent research (Shen, Zhou, Huang, & Xie, 2014), similar auxetic behavior was observed (Figure 5b). As clearly shown on the corresponding stress-strain curves, a long plateau exists indicating high deformability of auxetic materials.

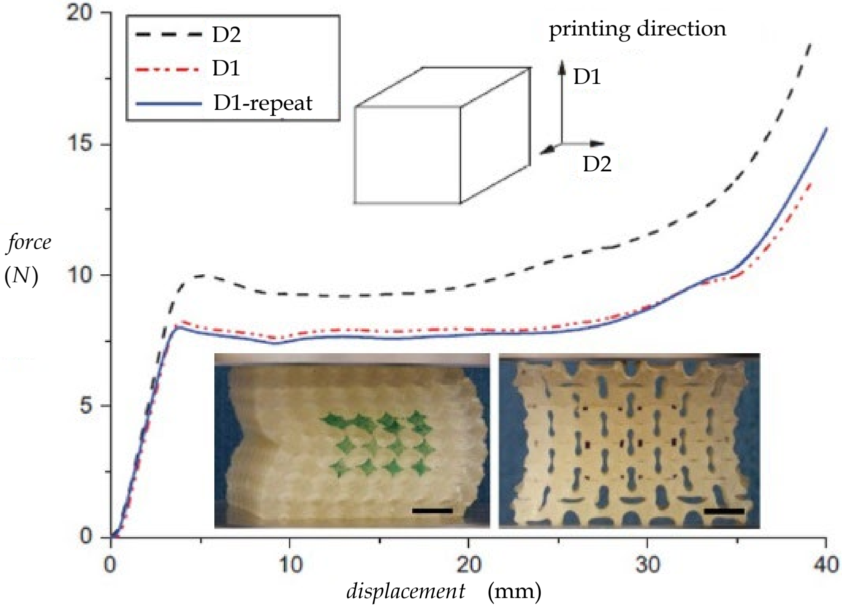
In theory, the auxetic behavior of cellular materials is mainly attributed to geometrical features of the constituent material arrangement. This can be achieved by numerous designs, such as re-entrant honeycombs (Kenneth E. Evans & Alderson, 2000; Lee et al., 2019; T. Mukhopadhyay & Adhikari, 2016; J. X. Qiao & Chen, 2015; Yang, Harrysson, West, & Cormier, 2015), rigid body rotation structures (Joseph N. Grima & Evans, 2000, 2006; J. N. Grima, Zammit, Gatt, Alderson, & Evans, 2007; Lim, 2017) and chiral structures (M.-H. Fu, Zheng, & Li, 2017; M. Fu, Liu, & Hu, 2018; Jiang & Li, 2017; Novak, Starčevič, Vesenjak, & Ren, 2019; Spadoni & Ruzzene, 2012). In practice, the mechanical response varies significantly when different constituent materials are used even for the same auxetic structure. Typically, for auxetic behavior to be achieved, the constituent material should be highly elastic and deformable. If cementitious materials are to be used as constituent materials, their brittleness and low deformability need to be addressed to achieve auxetic behavior. Therefore, the constituent cementitious materials might need to be toughened or reinforced.

2.4 *Turning brittleness to toughness*

More generally, the concept "cellular" not only refers to those structures constituted by dilute units with high porosity but also include the structures consisting of solid cells, tablets or blocks stacked in certain configurations.



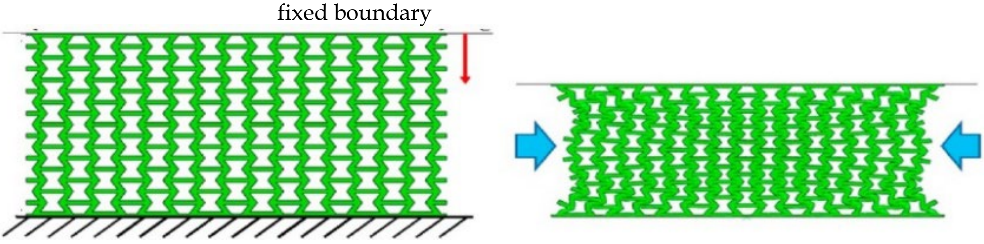
(a) pattern transformation observed in (Bertoldi et al., 2010)



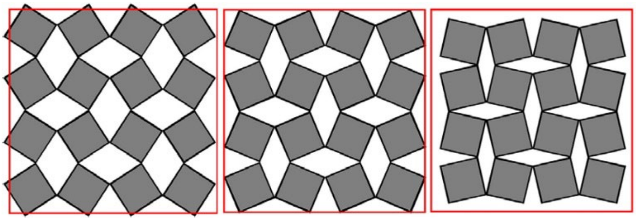
(b) Similar pattern transformation observed and corresponding stress-strain response reported in (Shen et al., 2014)

Figure 5. Deformation and stress-strain response of auxetic structure

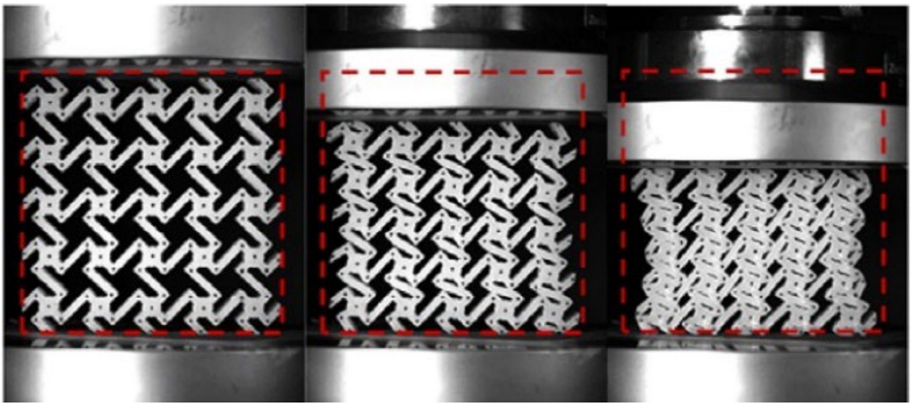
Bouligand is a typical structure with stacked-unit configuration (shown in Figure 7). This structure was found in the shell a of a specific beetle, *C. gloriosa* (Sharma, Crne, Park, & Srinivasarao, 2014), which has a special layered helicoidal structure. Bouligand has studied cholesteric liquid crystals which has a similar helical structure. Therefore, Pace (Junior, 1972) described this structure as Bouligand structure. One typical feature of this type of structure is the ability to enhance fracture resistance of laminated structures (Natarajan &



(a) re-entrant honeycomb



(b) rotating rigid units



(c) chiral structure 0% 22% 44%

Figure 6. Three types of auxetic structure

reprinted from (Jiang & Li, 2017; Lee et al., 2019; Lim, 2017)

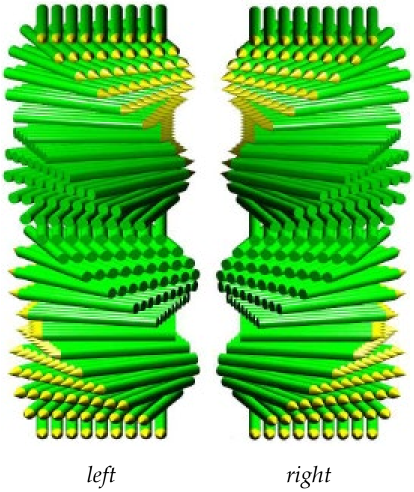


Figure 7a. The Bouliganad structure

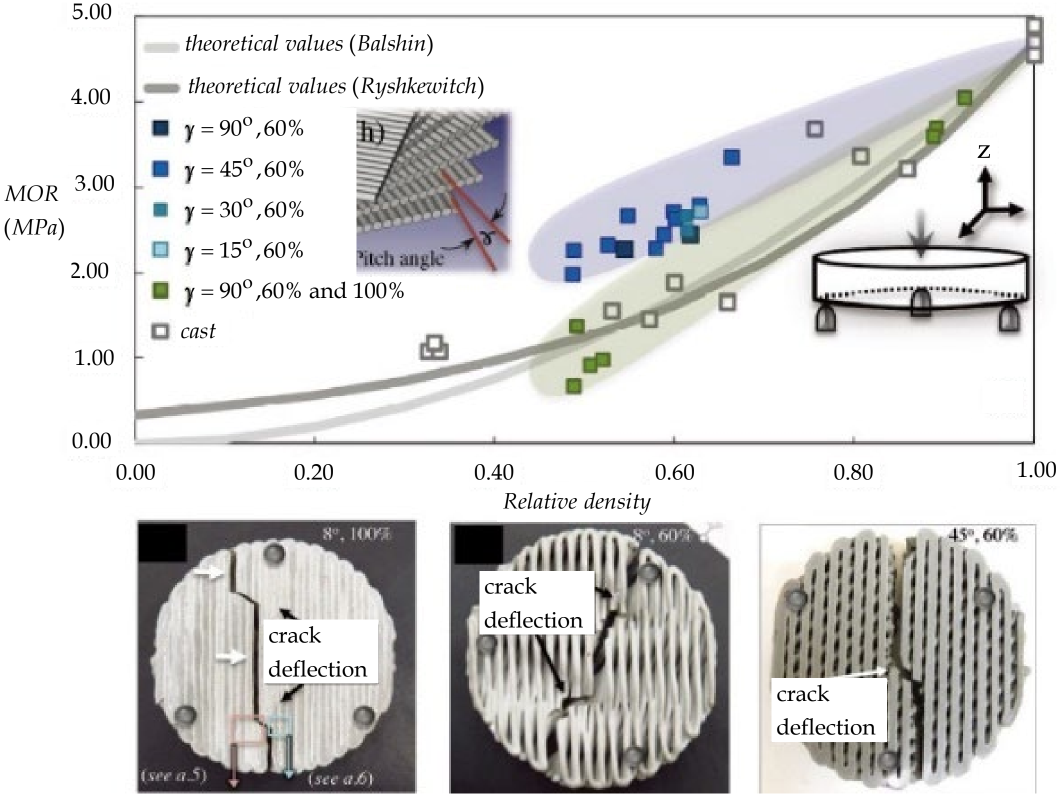


Figure 7b. Cementitious materials with Bouligand structure

Reprinted from (Moini et al., 2018; Sharma et al., 2014)

Gilman, 2018; Zimmermann et al., 2013). When adopted by Moini (Moini et al., 2018) to 3D printed cementitious materials, the Bouligand structured specimens have shown deflected crack pattern and improved fracture resistance without sacrificing strength which are usually two controversial aspects for brittle materials.

As mentioned, taking advantage of the ability to deflect cracking, fracture resistance of cementitious materials can be improved without decreasing strength. Similarly, another type of bio-inspired structure may also contribute to deflect cracks. Nacre, known as the "mother-of-pearl" (Barthelat, Tang, Zavattieri, Li, & Espinosa, 2007) is naturally generated with layered cells. Although constituted by brittle ceramic material, nacre shows higher strength and fracture toughness compared to its constituent. The extraordinary mechanical properties of nacre are achieved by its special hierarchical structures (shown in Figure 8). The nacre structure consists of small tablets adjacently stacked in the horizontal direction and densely packed in the vertical direction with multiple layers. Because of the interlocking between the tablets (Katti, Katti, Pradhan, & Bhosle, 2011) as well as the sliding between the layers (Barthelat et al., 2007), strength and toughness of the nacre structure is remarkable considering the brittleness of the ceramic tablet itself. For cementitious materials, Soltan (Soltan & Li, 2018; Soltan, Ranade, & Li, 2014) combined strain hardening cementitious composites and polymeric meshes to create nacre structured composites using 3D concrete printing (Figure 9). Distributed and deflected cracks were found on the flexural loaded nacre structured composites and the compressive strength, flexural strength as well as fracture toughness are found to be significantly enhanced. Similar approach and result have been reported by others recently (Ye, Cui, Yu, Yu, & Xiao, 2021; Ye, Yu, Yu, Zhang, & Li, 2021). Nevertheless, looking at the structural feature of

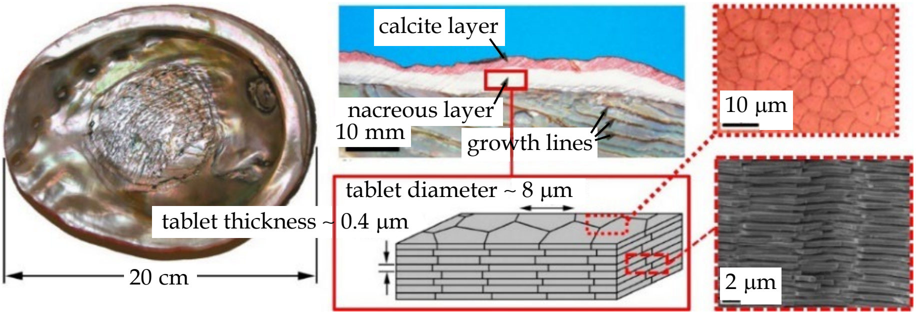


Figure 8. Hierarchical structure of Nacre, figures reprinted from (Barthelat et al., 2007)

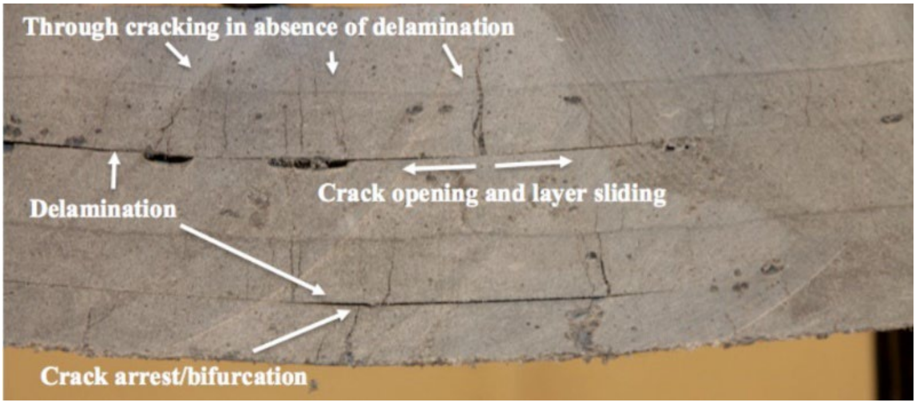


Figure 9. Deflected cracks by nacre-like structure reported in (Soltan et al., 2014)

the at Bouligand and nacre structure, the enhancement in mechanical properties is mainly in the direction parallel to the filaments or tablets. The improvement in interlayer mechanical properties between the stacked layers seems limited. Further studies in improving the interlayer mechanical properties are necessary.

3 Processing methods

Complex structure is a particular characteristic of cementitious cellular materials compared to conventional cementitious material. In order to prepare such complex cellular structures, special processing methods have to be applied. Owing to the rapid development of additive manufacturing (AM) technology in the past decade, 3D printing methods are becoming widely used for fabricating cellular materials with complex geometries (Bauer, Hengsbach, Tesari, Schwaiger, & Kraft, 2014; Buckmann et al., 2012; Taheri Andani, Karamooz-Ravari, Mirzaeifar, & Ni, 2018; Xia & Sanjayan, 2016). In most 3D printing techniques such as fused deposition modeling (FDM) (Kaur, Yun, Han, Thomas, & Kim, 2017; Kucewicz, Baranowski, Małachowski, Popławski, & Płatek, 2018), selective laser sintering (SLS) (Takano et al., 2017) and stereolithography (SLA) (He et al., 2018; Hollander et al., 2018; Xing, Zou, Li, & Fu, 2017), the materials are precisely deposited layer by layer to build up objects under the control of an automated system to ensure high printing quality even with complex geometries. For cementitious materials, different types of processing methods can be adopted to fabricate cellular structures.

3.1 *Indirect 3D printing*

The so-called "indirect 3D printing" actually refers to a method that combines 3D printing technique with traditional casting. The basic principle is similar to the "investment casting" (Pattnaik, Karunakar, & Jha, 2012) usually used in metal industry.

Normally, a mold with a designed cellular structure is prepared first, using materials that are easy to be printed and demolded. Polymers are an excellent choice to be the mold material. Their simpleness in printing ensures the possibility of making complex geometries. Meanwhile, equipment for printing polymers is much more accessible than any other materials. Even a commercial desktop 3D printer can be used to prepare polymeric 3D printed objects with complex geometries. This simplicity also makes it more economical to be used for field constructions. Another aspect is the ease of demolding. Even when complex three-dimensional geometries are used, the printed polymeric mold can be dissolved in corresponding solvent. For instance, Sajadi (S. M. Sajadi et al., 2021) printed a cellular mold by PVA (Polyvinyl alcohol) which is dissolvable in water. Aghdasi (Aghdasi et al., 2019) printed octet lattice molds (see Figure 10) with ABS (Acrylonitrile butadiene styrene) which is dissolvable in acetone. As the cementitious mixture needs to be casted into the mold, it naturally requires the mixture to possess high flowability otherwise air void defects easily occur (Wu, 2021).

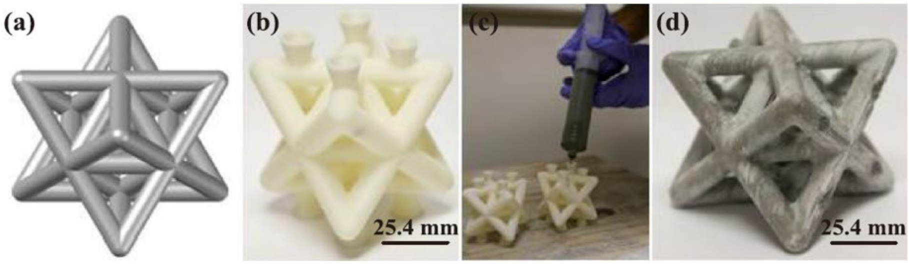
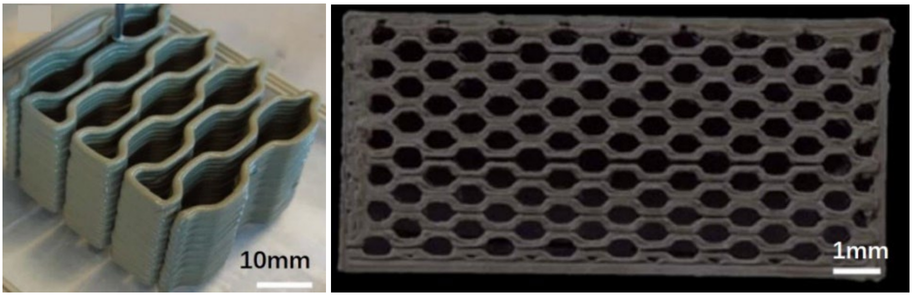


Figure 10. Schematics of typical procedures for indirect printing
a) designed structure, b) 3D printed polymer mold, c) casting cementitious mixture into the 3D printed molds, d) specimen after dissolving the mold (Aghdasi et al., 2019)

3.2 *Extrusion-based 3D printing*

Cellular cementitious composites with a layered structure are of particular interest for digitally fabricated cementitious materials as the layer-wise structure is almost an intrinsic characteristic induced by the processing method. This stacked-unit structure is widely seen in nature.

In terms of the processing methods of cementitious cellular materials, the ability for fabricating complex structures at a high resolution is crucial. For the extrusion-based printing technique, the extrudability and buildability of the cementitious mixture have to be properly addressed. In order to increase printing quality, a smaller nozzle size is preferable; however, this increases the difficulty of extruding mixed cementitious slurry. In addition, the extruded material needs to set fast such that the geometry can be properly maintained. At the moment, some authors reported case shows good printing quality at high resolution (here the resolution indicates the minimum size of a printed filament which can be extruded) by properly modifying the cementitious mixtures (Moini et al., 2018; Seyed Mohammad Sajadi et al., 2019). For large size concrete printing, larger nozzle size is need. Then, reaching a high relative printing resolution is required in order to get good printing quality. Under this condition, the size effect of the cementitious cellular materials has to be carefully considered. Normally, the global mechanical response of cellular structure is independent from the structure size if the property of the constituent material is not considered. However, due to the inherent size-dependent characteristic of cementitious materials, the mechanical properties of cementitious cellular materials should also exhibit size-dependent behavior.



(a) *(b)*

Figure 11. Printed cementitious cellular materials with high resolution, figures are reprinted from (Moini et al., 2018; Seyed Mohammad Sajadi et al., 2019)

3.3 ***Particle bed 3D printing***

Particle bed printing technique is also a layer-wise building up technique. Compared to the extrusion-based 3D printing which directly deposits ready mixed material, the particle bed printing technique deposits liquid binders and dry material separately. Basically, it consists of two repeated and consecutive steps: first, a layer of dry material (for example cement particles and aggregates) is deposited on a building platform; afterwards, a liquid phase is selectively sprayed on the deposited particles according to the designed printing path. The sprayed liquid phase is used to bind the particles. These two steps are repeated such that object can be built up layer-by-layer. After the printing steps are finished, excessive non-bonded particles can be removed. As shown in Figure 12 (Lowke et al., 2018), depending on the combination of binders and dry particles, several types of methods can be used to print cementitious materials.

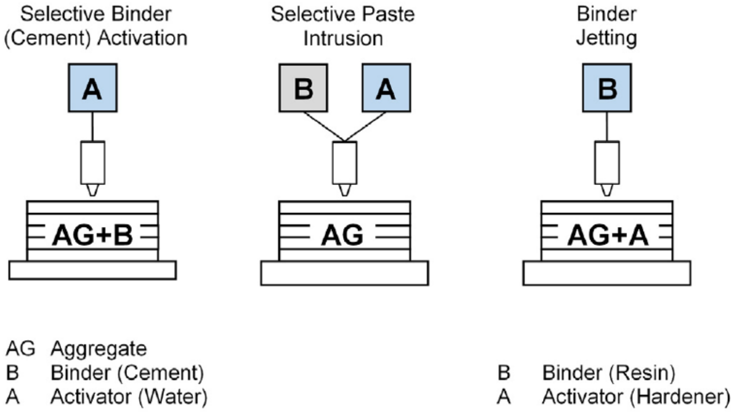
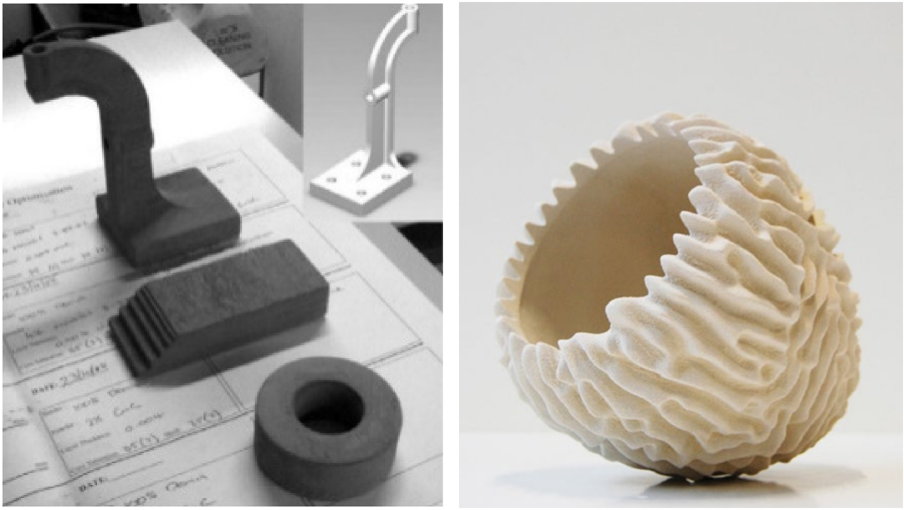


Figure 12. Schematics of powder, reprinted from (Lowke et al., 2018)

Compared to the indirect printing and extrusion process, the particle bed printing has higher freedom of form in fabrication. This is mainly attributed to two reasons. The first is that the dry materials and liquid phase can be independently designed. This almost eliminates the negative influence of certain ingredients on the flowability or buildability as seen in the indirect printing process and the extrusion-based printing process. Therefore, there is much more freedom to design a variety of cementitious mixtures without sacrificing the printing quality. The second is that during the printing process, the non-bonded particles serve as supporting material which ensures the particle bed printing method high ability to construct complex structure, for example overhangs, undercut or cavities, which are rather difficult for extrusion-based printing. Meanwhile, the size of the printed object by this technique covers a large range, from desktop objects size with a couple of millimeters (Xia & Sanjayan, 2016) up to construction size in meters (Lowke et al., 2018). However, because the liquid phase and dry particle are deposited in two steps, the equipment complexity of the particle bed printing technique is much higher than the other two techniques, which to some extent limits the application of this technique.

4 Architecting constituent materials

According to previous definitions, the *constituent material* is the cementitious mixture that is designed and processed to consist the single "unit" of the cellular structure. The concept of architected cementitious cellular materials not only refers to modifying the cellular structure as indicated in section 2 but combining the geometrical traits of the cellular structure with tailoring the constituent material to achieve better material performance. Compared to other materials, the design flexibility of constituent materials is one of the major advantages of cementitious cellular materials. By simply modifying the mixture design, the properties of cementitious material can cover a large range. For example, one main drawback of cementitious materials is their lack of ductility: they have low tensile strength and are prone to cracking (Van Mier, 1997, 2012). In order to overcome the brittleness, fibers have been used as reinforcement for cementitious materials (V. C. Li, 2003; Luković et al., 2014; Purnell & Beddows, 2005; Stähli, Custer, & Van Mier, 2008). A typical type of fiber reinforced cementitious material is engineering cementitious composites (ECC) (V. C. Li, 2003), also known as the strain hardening cementitious

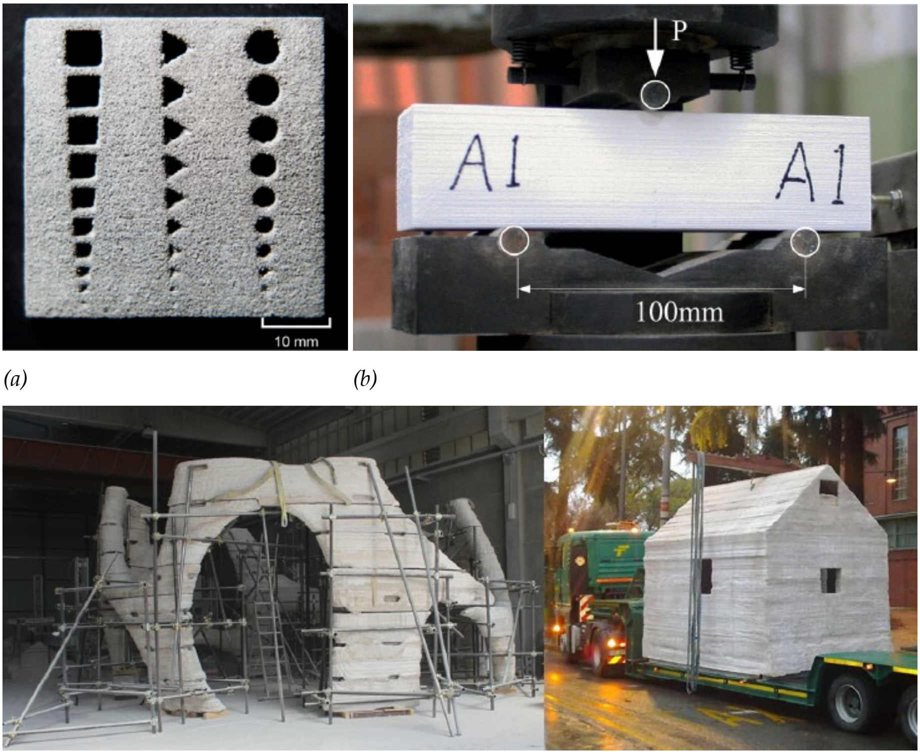


(a) *(b)*

Figure 13. Objects with complex features fabricated by cementitious particle bed printing, reprinted from (Gibbons, 2010; Lowke et al., 2018)

composites (SHCCs) (Mechtcherine, 2013; Pakravan, Latifi, & Jamshidi, 2017) in which PVA (Polyvinyl alcohol) fibers are often used. The main feature of this type of fiber reinforced cementitious composites is the so-called multiple cracking and pseudo strain hardening behavior in uniaxial tension: the SHCCs are able to crack multiple times before eventual failure, while the ultimate failure strength is higher than their first cracking strength. Owing to this, the SHCCs are significantly toughened compared to plain cementitious materials. Recent research (Soltan & Li, 2018) shows great potential by combining SHCCs with architected structure, where the architected cementitious composites exhibit obviously enhanced mechanical properties.

Before designing the mixture, the compatibility of the constituent materials with processing methods needs to be carefully addressed. Depending on the processing method, the compatibility indicates different requirements on properties of the constituent



(a) *(b)*

(c)

Figure 14. Particle bed printed objects with different sizes, reprinted from (Feng, Meng, Chen, & Ye, 2015; Lowke et al., 2018; Xia & Sanjayan, 2016)

material raised by the processing technique. Taking the indirect printing method as an example, as the cementitious mixtures are casted in molds with complex structures, good flowability of the cementitious materials is a basic requirement such that the presence of defects can be minimized. In this sense, the desired mechanical performance and potential influence of mixture ingredients on flowability need to be considered. Using the same example of the SHCCs, fibers significantly increase ductility while decreasing the flowability. Meanwhile, fiber length might also be a factor that needs to be considered in case unintended fiber orientation or clogging of cellular molds during casting may occur at small geometries.

Similarly, compatibility requirements raised by the processing method can be also found in extrusion-based 3D printing, for which good extrudability and buildability of the cementitious mixture are the required properties (Le, Austin, Lim, Buswell, Gibb, et al., 2012; Le, Austin, Lim, Buswell, Law, et al., 2012). For the paste intrusion particle bed printing method, paste penetration into the packed particles has been found to be critical on the strength of printed material (Lowke et al., 2020). By properly tailoring the particle size and the rheological properties of the cement paste, cementitious mixture with good compatibility can be designed. However, the aforementioned reinforcing method only focus on the "in-layer" material property. Due to the layer-wise production characteristic of the extrusion based printing, a lack of interlayer bonding is one of the main drawbacks of the extruded cementitious materials (Sanjayan, Nematollahi, Xia, & Marchment, 2018). Therefore, many studies have focused on reinforcing the interlayer of printed filament also improves the performance of printed material. These methods include depositing adhesive polymer as a bonding layer (Hosseini, Zakertabrizi, Korayem, & Xu, 2019), introducing interlocking geometry (Zareiyan & Khoshnevis, 2017), penetrating paste coated reinforcement bars (Marchment & Sanjayan, 2021), meshes (Marchment & Sanjayan, 2020) and penetrating nails (L. Wang, Ma, Liu, Buswell, & Li, 2021) between two printed layers. Obvious improvement in the interlayer bonding was found in these studies.

Extruded cementitious exhibit anisotropy in terms of mechanical behavior (Ding, Xiao, Zou, & Zhou, 2020; Panda, Chandra Paul, & Jen Tan, 2017), even in-layer and interlayer reinforcement is applied. It can be expected that the mechanical properties of extruded cementitious cellular materials may be significantly influenced by, not only the global structural response of the cellular structure but also the local anisotropy of the extruded elements. This influence can be a drawback of using extrusion-based method to prepare cementitious cellular material. However, similar to the results obtained in (Moini et al., 2018; Soltan & Li, 2018; Ye, Yu, et al., 2021), the anisotropy of extruded cementitious materials can be also used to improve mechanical properties, for instance fracture resistance. At the moment, the study on the mechanical properties of extruded cementitious cellular materials is still scarce, more studies in this area is expected in the future.

In general, the simplicity of tailoring cementitious mixtures facilitates achieving the desired mechanical properties independently without hindering the cellular structure of architected cementitious cellular materials. Nevertheless, potential requirements induced by the processing techniques have to be taken into consideration in the mixture designs.

5 Final remarks

In this work, several important aspects regarding cellular structures, processing techniques and constituent materials of developing architected cementitious cellular materials have been reviewed. Preliminary studies have shown great potential of achieving excellent mechanical properties by combing cellular structure with cementitious constituent materials. However, compared to the number of studies focusing on other types of materials such as metals and ceramics, there is still very limited knowledge on architecting cementitious cellular materials. This includes, but not limited to, potential cellular structure to be adopted by cementitious materials, fundamental mechanics, requirements in processing methods and constituent materials, mechanical properties of architected cementitious cellular materials as well as potential applications in engineering practice. These aspects hold great interest for future studies.

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