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Feasibility of Using Low CO₂ Concrete Alternatives in Extrusion-Based 3D Concrete Printing

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Abstract. In conventional concrete, replacing high-volume (more than 45%) of ordinary Portland cement (OPC) by supplementary cementitious materials (SCMs) is not a novel CO₂ reduction method, whereas rarely in 3D printable concrete. This study attempts to explore the feasibility of using SCMs in 3D printable concrete. Initially, the existing binder mixes, required fresh properties and a research method of 3D printable concrete are investigated by reviewing the relevant papers. Additionally, the constraints and opportunities of using SCMs in 3D printable concrete are illustrated and summarized. Finally, it has been found that up to 45% of cement can be replaced by a blend of fly ash and silica fume. The essential fresh properties of 3D printable concrete include extrudability, workability, open time, buildability and structural build-up, which are influenced by the binder mix, particle size distribution, water to binder ratio, binder to aggregate ratio, admixture addition, the dosage of reinforced-fibers, etc. On the other hand, there are many limitations to develop SCMs-based 3D printable concrete, such as few relevant studies, a lack of the certificated standard, massive related-parameters and the shortage of common SCMs. For the first three problems, it can be solved with the development of 3D printable concrete. For the last one, calcined clay is one potential alternative for developing sustainable 3D printable concrete in the areas where are in short supply of fly ash and silica fume.

Keywords: 3D printable concrete · Low CO₂
Supplementary cementitious materials · Fresh properties

1 Introduction

In the recent periods, extrusion-based 3D concrete printing (3DCP) as a novel concrete construction method has been significantly developed by many research institutions and enterprises throughout the world. 3DCP can be defined as a fabrication method that employs an additive, layer-based manufacturing technique to make concrete components without formwork [1]. The potential advantages of 3DCP include increasing flexibility in architecture [2], reducing labor usage, as well as saving in-situ construction time and the building costs [3]. As a future construction trend, 3DCP may be a potential low CO₂ approach [2] since no formwork is needed. Moreover, decreased

amounts of ordinary Portland cement (OPC) might be consumed by using 3DCP. Lim et al. [4] identified that the 3D model could be optimized for strength before concrete printing and thus the final print only requires the minimum amount of concrete. Besides, it has been found that small amounts of supplementary cementitious materials (SCMs) can be blended with OPC to improve the fresh properties of printing concrete. In conventional concrete, replacing the high-volume of OPC by SCMs is a CO₂ reduction strategy, especially when SCMs are sourced from industrial by-products like slag and fly ash [5]. However, there is no exploration for using the high-volume of cementitious alternatives in printable concrete. The experiences of using SCMs in conventional concrete cannot be directly referenced in 3D printable concrete since the different manufacturing processes are employed. Thus, this study initially aims to investigate existing binder mixes and the required fresh properties of 3D printable concrete by reviewing the relevant publications over the last 20 years. Furthermore, the constraints and opportunities of using those low CO₂ concrete alternatives in extrusion-based 3DCP are illustrated and discussed.

2 Binder Mix and Fresh Property of 3D Printable Concrete

2.1 Literature Survey of 3D Printable Binder Mix

As shown in Table 1, fly ash, and silica fume, as well as limestone, have been mixed into the binder of printing concrete in different research groups. The total amount of those cement alternatives in binders is around 10–45% by weight. OPC still possesses the highest content of binder mix in the existing printing concrete proposals. The primary objective of blending SCMs and limestone in the binder mix may be to achieve the required rheological requirements for printable concrete. Kazemian et al. [6] stated that adding silica fume in printing concrete could improve its cohesion property at the fresh state as well as the mechanical performance and impermeability when hardened. Adding ultra-fine fly ash is beneficial to the workability of 3D printable concrete by reducing yield stress and viscosity [11] at the early stage. Limestone as a kind of inert filler is commonly used to improve the workability of self-compacting concrete [12].

Table 1. Cementitious binder content of 3D printable concrete.

| Source: | Cementitious binder content (by weight) |
|---------|--|
| [6] | OPC (90%) and silica fume (10%) |
| [7, 8] | OPC (70%), fly ash (20%) and silica fume (10%) |
| [9] | OPC (55%), fly ash (22%) and silica fume (23%) |
| [10] | OPC (60–67%), limestone filler (17–20%) and silica fume (17–20%) |

2.2 Fresh Property of 3D Printable Concrete

Compared with conventional concrete, 3D printable concrete has many required properties especially in the fresh state, such as no slump and fast setting, due to the absence of formwork [2]. Utilizing different types and amounts of SCMs in the binder

mix may significantly affect the fresh properties of 3D printable concrete. The required fresh properties and dominant parameters are demonstrated in Fig. 1.

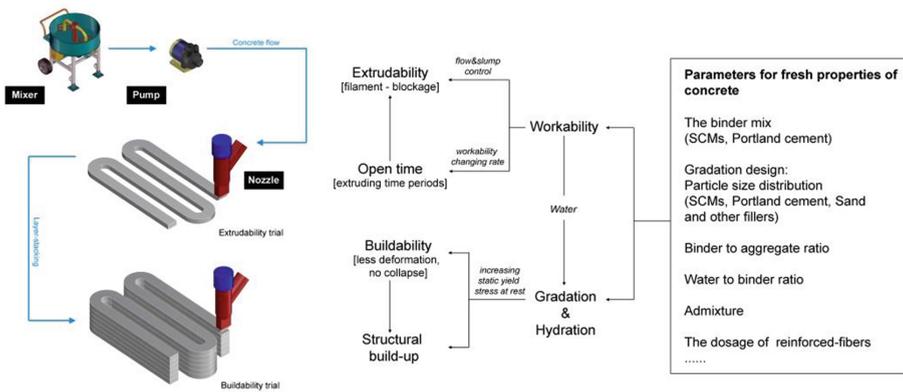


Fig. 1. Fresh properties of 3D printable concrete and dominant parameters

Extrudability. It is used to describe the property of a material that could be quickly and reliably delivered out from the transmission system [4, 7, 8]. Extrudability can be determined by using the visual inspection method. The comparable extrudability is evaluated by the continuity and conformity of the extruded filaments [6, 8]. According to Ma, Li, and Wang [8], the extrudability is primarily affected by the amount and distribution of the dry components in the blend. Besides, Le et al. [7] pointed out that the particle size distribution, binder to aggregate ratio, the dosage of superplasticizer and fibers influence the extrudability of printable concrete as well.

Workability. The conventional evaluation methods include slump, flow, and compact tests, which are inadequate for the printable concrete research. The workability of fresh printable concrete is feasible to be determined by conducting a rheological test [7]. In the study of Paul et al. [3], a Viskomat Rheometer is utilized to examine the workability of fluid concrete. By using a calibration coefficient [13], the viskomat values can be transferred to the parameters of plastic viscosity and yield stress which are expressed in the Bingham model for non-Newtonian flow [3]. The proper amount of superplasticizer is added to printable concrete to achieve the appropriate workability of fresh concrete with the lower water to binder ratio (0.2–0.3) [8, 9]. Additionally, the workability of fresh printable concrete may be influenced by using different types of cementitious alternatives as stated in Sect. 2.1.

Open Time. It should be defined as the time period for printing fluid concrete with proper workability [8]. It starts with extruding stable and consistent filaments and ends up with hardly printing the filament with standard quality. Open time is closely related to the changes of workability which can be determined by measuring shear strength of concrete with time by using a shear vane apparatus [7]. The decrease of workability with time is mainly due to the loss of water in fresh concrete. Both hydration and

evaporation processes contribute to the water consumption at this stage. For a specific environment condition (temperature, humidity, and wind), the length of open time is directly decided by the decreasing rate of workability which may depend on the hydration rate of printable concrete. Apart from the environmental factors, the parameters which can influence the hydration rate of concrete, like the water content, types of SCMs and admixture also affect the open time of 3D printable concrete. Besides, the impact of physical operations is non-negligible. According to Le et al. [7], the agitated fresh concrete shows longer open time than the non-agitated.

Buildability. It is considered as the ability of fresh concrete to resist the deformation and avoid collapse during the layer-based additive manufacturing process [4, 6, 8]. For adequate buildability, it is necessary that the first layer of printed concrete has sufficient yield stress to sustain the weight from itself and upper deposited layers [14]. Based on the study of Kazemian et al. [6], layer settlement and cylinder stability tests are utilized for determining the buildability of fluid concrete. The authors also illustrate that adding the proper dosage of rheology (or viscosity) modifier, silica fume or nano-clay will help to achieve the required shape stability of fresh printable concrete. On the other hand, Weng et al. [15] point out that the continuous gradation of particles in concrete will benefit to get the high yield stress of the printable mixture for better buildability. However, only the sand particle gradation was applied in their research. It is necessary to implement continuous gradation design for all constituents including cement, SCMs, and other fillers.

Besides, the time interval between two layers is also a critical parameter in this sector. If the time interval is not long enough, four filaments may form a void due to the deformation property of fresh concrete (Fig. 2). The higher porosity will affect the mechanical performance and durability of printed concrete. The longer time interval, the better shape stability of deposited layers can be got. Whereas, the longer time interval will also weaken the bond strength between two layers. Therefore, buildability and layer adhesiveness need to be considered together in further research.

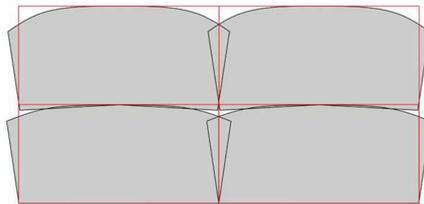


Fig. 2. A section view of 4 printed rectangle filaments (deformation)

Structural Build-up. It is a similar concept to buildability. The structural build-up is defined as the fact that the stiffness of fluid concrete increases with time due to hydration and physical operations [8, 16]. It is required to achieve a high structural build-up rate in 3D printable concrete. However, the higher structural build-up rate leads to lower bond strength between layers. In contrast to thixotropy, the concept of

structural build-up is applicable in both revisable and irreversible processes of fluid cement-based materials [16]. The penetration resistance method is utilized for measuring the structural build-up rates of printed concrete at different rest times by Ma, Li, and Wang [8]. In the study of Yuan et al. [16], replacing the partial amount of Portland cement by SCMs can affect the structural build-up of fresh concrete. To what extent the structural build-up behavior will be influenced should depend on the specific physical and chemical characteristics of SCMs.

A Trial & Error Process. Overall, based on the roles of different fresh properties of printable concrete, a testing method to explore the printability of mix designs is generated (Fig. 3). It is practicable to develop SCMs-based printable concrete by using this method.

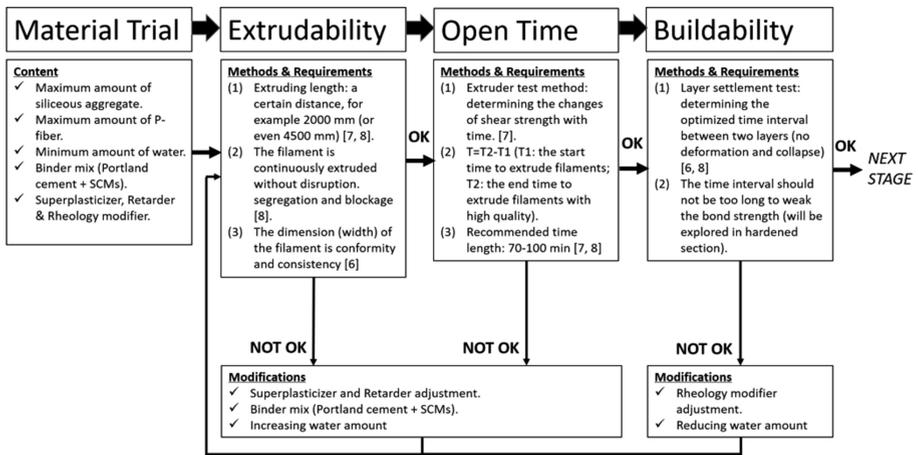


Fig. 3. A trial & error process for exploring the printability of SCMs-based concrete

3 Constraint and Opportunity to Develop SCMs-Based Printable Concrete

Utilizing the high-volume of SCMs or the mixture of limestone and SCMs to replace more than 45% of clinker is not a novel CO₂ reduction method in the conventional concrete industry. However, according to Table 1, this low CO₂ method is not widely adopted in 3DCP at present. There are four constraints to develop SCMs-based 3D printable concrete as follow. First, few studies have attempted to explore low CO₂ binder mixes of 3D printable concrete. The effects on fresh and hardened properties of different types and amounts of SCMs substitution in 3D printable concrete are unknown. Second, 3DCP is a novel technique. There is no certified standard for 3D printable concrete at present [17]. Current research efforts are built on the experiments from only a limited number of academic institutions. More specific and efficient test methods for determining the fresh and hardened properties of 3D printable concrete

need to be developed and evaluated further. Third, besides the binder mix, there are many other parameters that affect the fresh properties of 3D printable concrete, such as the particle size distribution, water to binder ratio, binder to aggregate ratio, admixtures addition, and dosage of fiber-reinforcement. Thus, to develop the low CO₂ printable cement by using large amounts of SCMs becomes more complicated and difficult. Fourth, replacing the clinker by the common SCMs (fly ash, slag, and silica fume) might not be a proper way for the long-term development of 3D printable concrete. The world production of silica fume is about 0.5–1.0 million tons per year which is quite limited compared to other SCMs [18]. According to Scrivener [19], the total amount of slag is only 5% of clinker, and the fly ash which is unavailable in many countries is around 30% of clinker worldwide. Therefore, it is necessary to seek the new and widely available source of SCMs.

The first, second and third problems may be solved with the development of 3D printable concrete. For the fourth constraint, in countries with an abundant resource of fly ash, it is worthwhile to develop low CO₂ printable concrete by utilizing high-volumes of fly ash. However, for countries lacking a supply of fly ash, it is necessary to use alternative SCMs which are abundant locally. In the conventional concrete industry, calcined clay has attracted more and more attention from researchers. Kaolinitic clays abundantly exist in the crust of the earth. After the dehydroxylation of the kaolinitic clay under a calcining process between 600 and 800 °C, metakaolin which shows comparable pozzolanic properties will be generated [20, 21]. Most properties of concrete can be enhanced by adding limestone and calcined clay [22]. The characteristics of the ternary blend have been illustrated by Antoni et al. [20], and Avet et al. [23]. The mortars which contain about 45% of metakaolin and limestone with a 2:1 proportion in the binder mix demonstrates better mechanical performance than the mortars with 100% of OPC at 7 and 28 days [20]. However, the price of the pure metakaolin in the study of Antoni et al. [20] is about three times of OPC. The high-grade kaolinitic clay and metakaolin usually are used by other industries, for example, ceramics, and paper [24]. Utilizing lower grade kaolinitic clays which are widely available and much cheaper to substitute clinker may be an ideal solution. According to the study of Avet et al. [23], replacing partial clinker (even 50% in the LC3 blend: 15% of limestone, 30% of calcined clay, and 5% of gypsum) by the lower grade calcined clay which contains at least 40% of calcined kaolinitic clays can achieve the same compressive strength after seven days. Thus, it is feasible to use lower grade calcined kaolinitic clays in the concrete industry. However, no one attempted to implement calcined clay cement or limestone calcined clay cement in 3DCP currently.

4 Conclusion

Overall, through reviewing the relevant literature published over the past 20 years, it is found that SCMs like fly ash, silica fume, and limestone have been applied for making printable concrete. Up to 45% of OPC can be substituted by the blend of fly ash and silica fume in the binder mix of 3D printable concrete. Additionally, this study reports the required fresh properties of 3D printable concrete, including extrudability, workability, open time, buildability and structural build-up. Those properties are significantly

affected by the binder mix, particle size distribution, water to binder ratio, binder to aggregate ratio, admixture addition, the dosage of reinforced-fibers, etc. Based on those fresh properties and parameters, a trial & error process method for testing the printability of SCMs-based concrete is generated.

However, many constraints still exist for using SCMs as low CO₂ cementitious alternatives in extrusion-based 3DCP. Only a few studies have attempted to explore the feasibility of SCMs as OPC replacement in 3D printable concrete, especially using high-volume of SCMs as substitutions. No certified standard of printable concrete is currently available. The fresh properties of 3D printable concrete depend on not only the binder mix but other material conditions. Increasing the amount of SCMs in 3D printable concrete should also consider other parameters, which would make the experimental process difficult and complicated. The geographical distribution of fly ash is uneven worldwide. Silica fume and slag are in limited supply and cannot satisfy a long-term global demand.

After a series of analysis, the opportunities of developing low CO₂ printable concrete by using SCMs are summarized as follow. In the place with abundant sources of fly ash, it is worth to explore the printability of high-volume of fly ash-based blends. Calcined kaolinitic clays as a widely available SCM has been investigated and applied in the conventional concrete industry. Using lower-grade calcined clay or the blend of limestone and calcined clay as low CO₂ alternatives is one potential direction for making 3D printable concrete in the future.

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