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EXPERIMENTAL STUDY OF MIXTURE PROPORTIONS AND FRESH PROPERTIES OF CONCRETE WITH FLY ASH AND SILICA FUME AS A REPLACEMENT FOR CEMENT FOR 3D PRINTING

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Abstract

3D eco-concrete printing using fly ash and silica fume is an innovative and sustainable construction method that harnesses the power of digital concrete technology. By integrating these replacement cementitious materials, this approach aims to optimize fresh properties and enhance the environmental friendliness of concrete structures. Fly ash, a by-product of coal combustion, and silica fume, a waste product from silicon metal and ferrosilicon alloy production, are used as sustainable alternatives to traditional cement. During the study the main mechanical properties, in fresh state, were assessed. The 3D printable concrete control compositions have a percentage between 55% and 75% sand, 45% and 25% cement, 1.5% superplasticizer and 0.5% VMA and for the 3D printable eco-concrete the percentage of cement was replace with 20% fly ash and 10% silica fume. The results had shown a decrease in bulk density for the eco-concrete 3D printed specimens between 3.3% and 2.5%, and as a requirement for 3D printed concrete, the compositions need to have little to no flow after it is extruded, for the slump flow test and jump table test mix CSTAMIX6 and CSTBMIX4 showed the best results in terms of buildability of the printed layer which can support its own weight and the weight of the superior layers. This utilization not only reduces the carbon footprint associated with concrete production but also improves material optimization by enhancing strength, durability, and workability. The combination of 3D printing technology and eco-friendly materials allows for precise and intricate designs while ensuring sustainability in the construction industry. This emerging field presents a promising avenue for creating environmentally conscious buildings, minimizing waste, and promoting a greener future for construction.

Key words: environment friendliness, fly ash, fresh properties, material optimization, silica fume

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1. Introduction

The most common modern cement mixture, known as Portland cement has been used for near 200 years, but it is not very friendly to the environment as cement production led in 2020 to 8% of emissions of carbon dioxide, worldwide (https://psci.princeton.edu/tips/2020/11/3/cement-and-concrete-the-environmental-impact). For this reason, low-CO_2 binders are needed by using industries by-products such as fly ash, silica fume etc., instead of disposing them in a warehouse or open field (Fig. 1). 3D concrete printing is an emerging and innovative technology for fabricating concrete components employing the additive manufacturing technique (Tay et al., 2017) that shows great potential for increasing productivity and safety in construction (Buchli et al., 2018; Buswell et al., 2007).

This new method of construction can be used to build complex and unique geometrical shapes without formwork. Benefits of this process can include a reducing of material wastage, customized structures, freeform structures, and the ability to optimize the material distribution according to the

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need of strength requirements (Bos et al., 2016; Kothman and Faber, 2016).

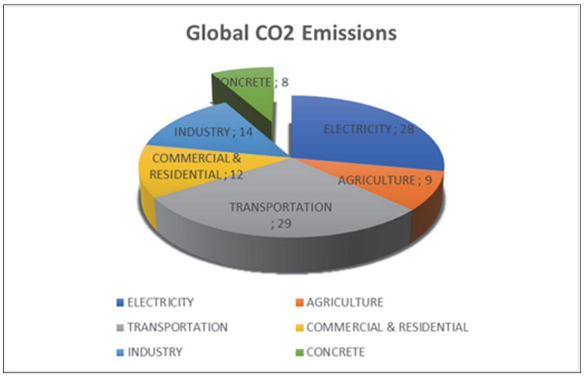

Fig. 1. Global CO_2 emissions by source category

In this context, 3D printable concrete (3DPC) is a "tailor-made" material that can be delivered by the pumping system and extruded through the nozzle of a 3D printer, and then, after deposition, maintain its shape stable under the gravitational load of subsequent printed concrete layers without the support by formwork (Zhang et al., 2021). 3DPC, as a digital technology, brings numerous benefits to construction, like highly flexible architectural design (Agustí-Juan et al., 2017; Lim et al., 2012), formwork-free fabrication (Lim et al., 2020), faster construction (Mechtcherine et al., 2018), better working conditions (Mechtcherine et al., 2019), material savings (Ngo et al., 2018) etc. All that is very promising also with respect to the cost-efficiency of construction.

The integration of 3D printing with methods for concrete construction will have an environmental impact at different levels of the building process. First, 3D concrete printing should limit or even eliminate the production and management of formworks, which can generate a large amount of waste, particularly in the case of formworks for complex structures with assembly components that are used only once (Perrot and Rangeard, 2019). In this sense, the reduction in environmental impacts related to 3D printing mainly comes from the design phase, which allows the topological optimization of structures and thus reduces the volumes of materials involved in a given mechanical function (Agustí-Juan and Habert, 2017).

Efforts have been made to mitigate the environmental effects of the cement industry by exploring substitutes for ordinary Portland cement (PC). One promising avenue of research focuses on sustainable building materials, specifically geopolymers made from fly ash (FA). These materials have garnered significant interest due to their ability to generate approximately 80% less CO_2 than PC (WBCSD, 2002). Additionally, geopolymers generally exhibit superior mechanical properties compared to PC-based materials, particularly in terms of early strength development (Bakharev et al., 1999; Duxson and Van Deventer, 2009).

Nowadays, construction concrete is based on multi-component cements, with a more or less diversified composition, containing one or more substitutes for the cement binder (Golewski and Szostak, 2022; Xie et al., 2020). Such materials are referred to as Supplementary Cementitious Materials (SCMs) (Bicer, 2020). The use SCMs allows to improve the efficiency of OPC production related to the possibility of using large amounts of mineral additives, including byproducts of industries or waste (Golewski, 2023), and meets the guidelines of sustainable development (Wang et al., 2023). It is important to mention that the increasing use of SCMs in OPC contributes to significant reduction of CO_2 greenhouse emissions and energy consumption during the production process. Furthermore, the synergistic effect of the interaction of several mineral additives has a more favorable effect on the properties of multi-component cements compared to cements containing only one mineral additive (Han et al., 2022). Therefore, on an increasingly larger scale, laboratory tests are carried out and then implementation works on the use of multi-component cements (Golewski, 2023) in 3D printing concrete technology are realized.

Multiple research papers and laboratory test carried out by universities and by the private sector came to the conclusions that a concrete mixture can be 3D printed if it's satisfied the following quality conditions or critical fresh concrete properties (Hou et al., 2021; Li et al., 2020):

- workability is the capacity of the mixture to be pumped through the printer pump;
- extrudability is the capacity of the mixture to be extruded through the nozzle of the printer;
- shape retention is the capacity of the mixture to keep its shape after it is extruded;
- open time refers to the time frame that the mixture can be pump without clogging the printer pump;
- buildability is the capacity of the mixture layers to support itself without major deformation or collapse.

The first two quality conditions can be related to the type of printer that it is use and the other quality conditions are related to the type of concrete mixture and composition of materials used.

The main objective of the experimental research in this paper is the analysis of the mix proportion compatible with the 3D technology and the physical-mechanical properties of some types of concrete that want to be ecological, using fly ash and silica fume as a substitute for cement.

2. Experimental program

2.1. Mixtures composition

For the preparation of the eco-concrete mixtures suitable for 3D printing, the following materials were used:

- cement type CEM II/A-S 52,5
- sand sort 0 - 1 mm
- water
- fly ash (FA) from CET Holboca Iasi

- silica fume
- Sika ViscoCrete -20 HE GOLD superplasticizer / water reducing additive in a dosage of 1.5% from cement's amount
- Pantarheo SE10 viscosity modifying admixture in a dosage of 0.5% from cement's amount

In the experimental program, a series of cement concrete mixtures were prepared, based on the proposed framework, which would be a starting point for systematic investigation on the printable concrete by researchers and would provide a basis for future specifications and guidelines (Kazemian et al., 2019). Printing concrete, has no relevant guidelines or proposed procedures for mixture evaluation, or any set of well-defined acceptance criteria. Establishing universal acceptance criteria for printing concrete would be possible only after many relevant studies have been carried out and a reasonable amount of data is available on the performance of different printing mixtures used in actual construction projects (Kazemian et al., 2017). The proposed framework is presented in Fig. 2.

2. 2. Experimental set up

During the research, it was proposed for testing and verification 25 mixtures with the ratio W/C between 0.3 and 0.5, the proportion of sand between 75% and 55% and the proportion of cement between 25% and 45% of the total mass of the mixture, with a density of approximately 2300 kg/m^3. Several types of recycled materials were studied in different dosages, 30% of the cement mass was replace with 20% fly ash and 10 % silica fume, to determine the physical and mechanical characteristics of the concrete obtained with each of the materials used for each dosage separately, in order to compare the results, thus aiming to create an ecological concrete, with costs of lower production than ordinary concrete, with a less negative impact on the environment, but with strengths comparable to those of ordinary concrete (Bărbuță et al., 2015; Bărbuță et al. 2016; Ioniuc et al., 2016).

The mixtures were prepared in a concrete mixer with a mixer capacity of 0.5 m^3. The aggregates were introduced into the concrete mixer, according to the standard (SR EN 12390-2, 2019), in the present case only the sand, then the cement. The dry mixture was mixed, then the water, the superplasticizer and viscosity modifier additives were introduced and mixed again for almost 5 min.

From 25 mixtures tested it was chosen 5 mixtures that satisfied the requirements needed for obtaining eco-concrete mixtures suitable for 3D printing.

- **CSTAMIX6-** Concrete with 75% sand, 25% cement with 20% fly ash and 10% silica fume as a substitute for cement.
- **CSTBMIX4**- Concrete with 70% sand, 30% cement with 20% fly ash and 10% silica fume as a substitute for cement.
- **CSTCMIX4-** Concrete with 65% sand, 35% cement with 20% fly ash and 10% silica fume as a substitute for cement.
- **CSTDMIX4-** Concrete with 60% sand, 40% cement with 20% fly ash and 10% silica fume as a substitute for cement.
- **CSTEMIX3-** Concrete with 55% sand, 45% cement with 20% fly ash and 10% silica fume as a substitute for cement.

The compositions of the mixtures are shown in Table 1.

3. Testing results and discussion

3.1. Eco-concrete 3D printing specimens

In many literature studies, the concrete mixture compatible with a 3D printer has some critical fresh concrete properties such as workability, extrudability, shape retention, open time and buildability (Panda and Ming, 2018). The most important one is buildability, which refers to the mixture's capacity to resist deformations during layer-on-layer printing (Malaeb et al. 2019).

The buildability test was carried out by stacking layers of concrete on top of each other until a major deformation or collapse occurred. The number of buildable layers was determined for all five mixtures. For sample CSTAMIX6 and CSTBMIX4 with four layers of printed concrete no major deformation was visible while sample CSTCMIX4 and CSTDMIX4 just after three layers deformation occurred and sample CSTDMIX3 from the first layer started to have small deformation. To increase the number of printed layers the quantity of superplasticizer will be decreased. The results of the printing process of the mixtures suitable for 3D printing are shown in Fig. 3.

In the printing process, based on the mix design, it is sought an appropriate balance, the concrete must have high compressive strength and shear strength to support its own weight and the water-cement ratio must be reduced as much as possible.

However, a certain water content must be maintained to ensure proper workability of the concrete. The consistency of the concrete must be fluid, but capable of supporting other layers after pouring. Finally, when poured, the mixture should dry as quickly as possible, but should be able to retain its moisture for a sufficient period of time to ensure a proper bond with the next layer.

Table 1. Chemical composition of raw materials (wt.%)

Material	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3
FA	58.83	32.62	3.44	7.52	1.075	1.538
OPC	19	7	6	60	5	3

SF	67	10	2.25	17	1.5	2.25

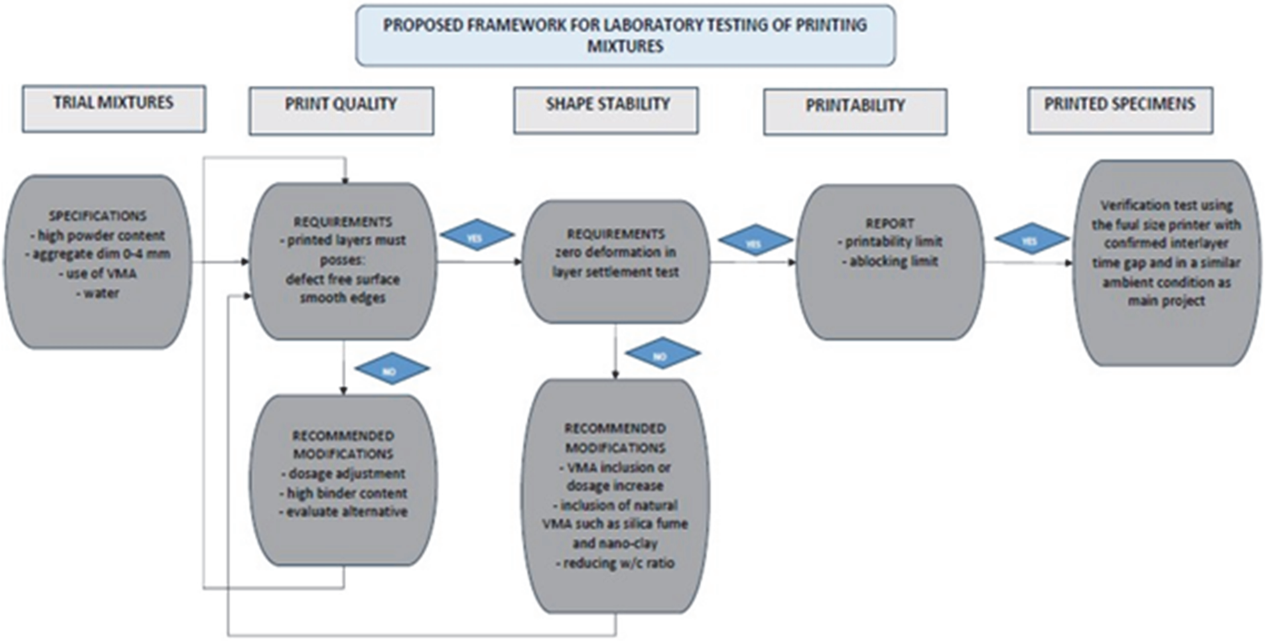

Fig. 2. Proposed framework for laboratory testing of suitable mixture for 3D printing

Table 2. Optimal eco-concrete mixtures suitable for 3D printing

Materials	**CSTAMIX6**	**CSTBMIX4**	**CSTCMIX4**	**CSTDMIX4**	**CSTEMIX3**
	W/C 0.5	W/C 0.43	W/C 0.38	W/C 0.33	W/C 0.30
Cement	638.4	625.8	614.6	602.7	591.5
Sand	1114	1093	1072	1052	1032
Fly ash 20%	182.4	178.8	175.6	172.2	169
Silica fume 10%	91.2	89.4	87.8	86.1	84.5
Water	274	313	351	387	423
Sika ViscoCrete 20he gold superplasticizer 1.5%	13.68	13.41	13.17	12.915	12.675
Pantarheo SE10 VMA 0.5%	4.56	4.47	4.39	4.305	4.225

Fig. 3. Printing process- extruded filament (left), shape stability (middle) multi-layer's structure (right)

3.2. Fresh state mechanical properties of eco-concrete 3D printed specimens

Experimental values of density determined at 28 days are presented in Fig. 4. The determination of apparent density consists in determining the mass of a fresh concrete sample and relating it to the volume of the respective sample in a compacted state. It is noted that the density of 3D printing eco-concrete is smaller than that of control concrete for all types of eco-concrete. For fresh concrete the density decreased with percentages between 3.3% and 2.50%. The hardened concrete density decreased with percentages between 4.56% and 3.16%. The densities of all mixtures, beside CSTAMIX6 of hardened eco-concrete with fly ash and silica fume are between 2000 kg/m^3 and 2500 kg/m^3, all the eco-concretes are normal weight concretes. The slump flow test is shown in Fig. 5.

The 3D technology required that the mix composition need to have lower slump values but still be pumpable, due to the requirements of shape retention and buildability of the printed layers. For the control mix and the eco-concrete, mix CSTAMIX6, CSTBMIX4 and CSTBMIX4 met the necessary requirements.

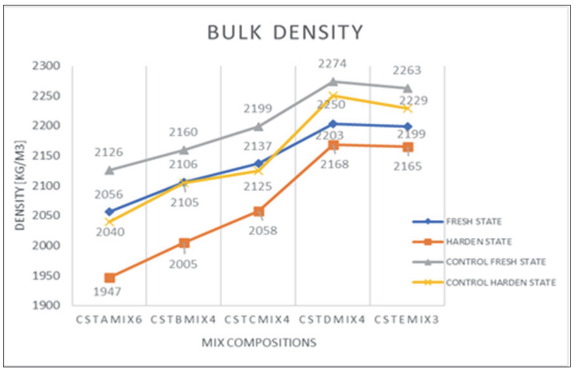

Fig. 4. Bulk density for experimental eco-concrete 3D printing samples

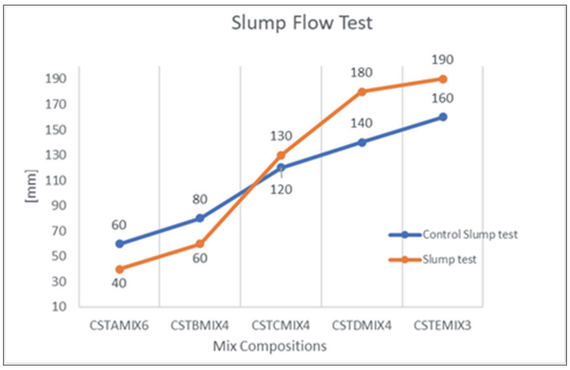

Fig. 5. Slump flow test experimental eco-concrete 3D printing samples

Fig. 6. The slump method for experimental eco-concrete 3D printing samples

The slump method is used to determine the consistency of fresh concrete and is applicable to concretes where the maximum aggregate size is 40 mm. The method consists in measuring the settlement of fresh concrete under its own weight. The fresh concrete is compacted in a truncated cone pattern. When the cone is raised vertically, the slump distance of the concrete allows the consistency of the concrete to be measured. The Jump table test is shown in Fig. 7.

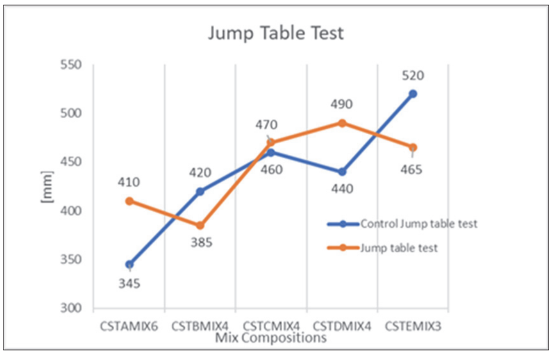

Fig. 7. Jump table for experimental eco-concrete 3D printing samples

For the jump table text, due to the reduced water intake in relation with the cement percentage, mix CSTAMIX6 and CSTBMIX4 show small spreading values but with inconsistency of the mixture, showing a need in the increasing of superplasticizer and viscosity modifier additives (Fig. 8).

Fig. 8. Jump table method for experimental eco-concrete 3D printing samples

This method is applicable to concretes with a maximum aggregate size of 63 mm, being useful for changes in concrete consistency corresponding to spreading values between 340 mm and 600 mm. The method does not apply to self-compacting concrete or expanded concrete. The principle of the method is to determine the consistency of fresh concrete by measuring the spread of concrete on a flat table that is subjected to shaking.

The approach taken is one of trial and error for this purpose as there is currently no guidance or suggested procedure for the design and testing of 3D eco-concrete mixtures.

4. Conclusions

In this research, it was investigated the use of different volumes of sand, cement, fly ash, silica fume and water content, to establish the right mixtures for the concrete to be 3D printed. The following conclusions were made which are based on the experimental results and observations:

- In the research, it was proposed for testing and verification 25 mixtures with the ratio W/C between 0.3 and 0.5, the proportion of sand between 75% and 55% and the proportion of cement between 25% and 45% of the total mass of the mixture, from which five mixtures were selected to be modified.
- The proposed use of 20% FA and 10% SF as a substitution for OPC brings clear improvements of the main fresh mechanical properties of 3D printed eco-concrete.
- All five mixtures are compatible with the 3D technology and can be printed as they fulfilled the quality requirements for workability, extrudability, shape retention, open time and buildability.
- The density of the eco-concrete samples has decreased for sample CSTAMIX6 by 3,3%, sample CSTBMIX4 by 2,5%, %, sample CSTCMIX4 by 2,8%, %, sample CSTDMIX4 by 3,1%, and sample CSTDMIX3 by 2,8%.
- The sump flow test showed that sample CSTAMIX6 and sample CSTBMIX4 show small spreading values but with inconsistency of the mixture, showing a need in the increasing of superplasticizer and viscosity modifier additives.
- The jump table test showed inconsistent values for the control and eco-concrete samples which implies that this method is not recommended to be use for 3D printed mixtures.
- Laboratory tests will continue with verification of mechanical properties in hardened state, the mixture will be modified further by increasing the percentage of FA and SF up to 80% SF as a substitution for OPC.

The potential of automation, the elimination of formwork in the building process, reduced or eliminated waste and the possibility for using green concrete mixtures compatible with the 3D printing technology highlights the importance of this technology for the construction industry.

3D printing significantly reduces on-site construction waste. By manufacturing components off-site and only printing what is necessary, the amount of construction waste generated during the building process is greatly reduced. This not only reduces landfill waste but also minimizes the need for waste disposal and cleanup.

Overall, the advantages shown by the 3D printing technology contribute to a more sustainable and environmentally friendly approach to the construction industry

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