Nano World Journal

https://doi.org/10.17756/nwj.2023-s2-045

Research Article

Open Access

Application of the Taguchi Method for the Optimization of Cementitious Mixtures' Printability Using Appropriate Chemical Admixtures

I. Tsilikis*, **D. Kioupis, A. Skaropoulou, K. Aspiotis and S. Tsivilis**

National Technical University of Athens - School of Chemical Engineering, 9 Heroon Polytechniou str., Athens, Greece

***Correspondence to:**
I. Tsilikis
National Technical University of Athens,
School of Chemical Engineering,
9 Heroon Polytechniou str., Athens, Greece.
E-mail: ioannistsilikis@outlook.com

Received: July 25, 2023
Accepted: September 26, 2023
Published: September 27, 2023

Citation: Tsilikis I, Kioupis D, Skaropoulou A, Aspiotis K, Tsivilis S. 2023. Application of the Taguchi Method for the Optimization of Cementitious Mixtures' Printability Using Appropriate Chemical Admixtures. *Nano World J* 9(S2): S262-S267.

Published by United Scientific Group

Abstract

This study examines the development of an initial framework for laboratory testing and evaluation of fresh 3D printable concrete mixtures' performance. Firstly, twelve printable mixtures were prepared consisting of cement, silica fume and three different chemical admixtures: accelerator, retarder, and superplasticizer. The rheological properties such as workability and flowability, were evaluated through flow table testing and a lab-scale additive manufacturing process was held to check their extrudability and buildability properties. To determine mix initial setting time a Vicat device was used (EN 196-3) while compressive strength measurements (EN 196-1) were also conducted. Secondly, the optimum combination of the aforementioned chemical admixtures was investigated through the application of the Taguchi method which efficiently reduces the number of required experiments. The preliminary results indicated that the addition of any type of chemical admixture improved the paste's printing properties and performance. Moreover, an indirect correlation between the printable mixtures properties was derived: buildability and workability as well as extrudability and the determination of the initial setting time of each composition. The ideal flowability of the mixtures ranges from 0.20 to 0.25, regardless of the admixture type. Above this range, the printing material is less buildable while below this range the material is not extrudable. The application of the Taguchi method allowed the investigation of the combinatorial effect of superplasticizer, accelerator and retarder on the strain generated on the extruded material during the printing process. Superplasticizer has the major contribution on the strain revealing that flowability is critical for the successful printing process.

Keywords

3D concrete printing, Taguchi method, Chemical admixtures, Additive manufacturing, Construction materials, Laboratory testing methods

Introduction

3D concrete printing technology (3DCP), a cement-based construction manufacturing process, has seen tremendous development in recent years, as it is characterized by many advantages [1-3]. Concrete printing automation can improve construction efficiency by eliminating the need for formwork, construction durability by providing precise control of concrete compositions, and construction safety by removing people from hazardous work environments. Through rapid construction implementation using 3D printed concrete techniques, housing can be created in communities affected by natural disasters, taller wind turbine towers can be built to harness higher energy winds, and concrete can be repaired in areas that are impossible using conventional construction equipment. To date, research work on this technology has limited its focus to the development of printing systems and the materials used to produce high-quality structures [4-7]. More specifically, from several printing systems only two have been adapted in concrete construction, extrusion-based layering, and powder-bed, allowing the in-site and offsite construction respectively [4-11].

Various secondary binders (Silica fume, fly ash, and gypsum) and additives (superplasticizers, accelerators, and retarders) have been examined to adjust setting time and rheology [12-17]. However, printable concrete, defined as the most recent specific type of concrete, lacks relevant guidelines or proposed procedures for the evaluation of mixtures and new materials or any set of clearly defined criteria regarding their quality and performance. Although a limited number of studies have provided an initial understanding of some of the desirable properties of printable concrete, during the last few years, experimental data are scarce [12]. Thus, the characterization of the fresh state behavior of a printable mix requires deeper investigation. Therefore, a comprehensive list of performance requirements and test methods for a printable mix should be developed.

The purpose of this work is to investigate the development of a structured laboratory testing framework based on the performance evaluation of printed cement pastes. More particularly, the relationship between fresh state properties (rheological, mechanical, etc.) and the ability of cementitious materials to be printed are investigated. Through this process, the permissible ranges of values of key performance indicators of quality printing are defined. The proposed laboratory tests include classical methods for assessing the fresh state of the material (flow table test, determination of initial setting time using a Vicat device) and a simple, non-standard method for extrudability testing. Then, a multifactorial design of experiments (DoE) was applied to investigate the 3DCP synthesis. In particular, this type of DoE was used to explore the combined effect of different additives (accelerator, retarder, and superplasticizer) on the printability of the cement pastes and therefore to get closer to a successful 3DCP synthesis.

Materials and Method

Raw materials - samples preparation

For the experimental procedure, a type CEM I 42.5N (C) ordinary Portland cement supplied by Lafarge Holcim Cement Company S.A., Greece; Sika silica fume HRD (SF), accelerator HyCon S 3200 F BASF (ACC), retarder HyCon R 7200 F BASF (RET), superplasticizer Sika Viscocrete ultra 600 (SP) and reverse osmosis water (W), were used. Cementitious mixtures are nanoscale materials.

A total of 16 different compositions were produced with different proportions, based on literature [5, 12, 18], of additives and admixtures (Table 1). The materials were mixed according to EN 196-1. Three different techniques were tested to check the rheological properties of each fresh paste: extrudability test, workability of the material and initial setting time. Furthermore, mechanical strength in uniaxial compression of the hardened paste was tested.

Rheological properties

The rheological properties of each fresh paste were evaluated using three different approaches: (1) extrudability test, (2) workability of the material, and 3) initial setting time. For the extrusion evaluation, a silicone gun (Figure 1a) was used as an extrusion device, to which a metal nozzle of the conical cross-section with an internal end diameter of 15.18 mm was manufactured and attached. Upon completion of the mixing process, the extrusion apparatus was supplied and the extrusion of the first layer was then carried out (time 0 min). Three layers were deposited at times 0, 10, and 20 min while the mixing continued throughout the experiment (Figure 1b and 1c). For each experiment 3 samples of three layers were prepared. The dimensions of each layer at all three intervals (0, 10, and 20 min) were measured using the ImageJ, an image analysis program (Figure 2).

Table 1: Codename, composition (% w/w) and water-to-binder ratio (W/B)^ of compositions.

Codename	C	SF	ACC	RET	SP	W/B
REF	100	-	-	-	-	0.27
SF5%	95	5	-	-	-	0.28
SF10%	90	10	-	-	-	0.29
SF20%	80	20	-	-	-	0.32
ACC0.5%	90	10	0.5	-	-	0.29
ACC1.0%	90	10	1.0	-	-	0.29
ACC1.5%	90	10	1.5	-	-	0.29
ACC2.0%	90	10	2.0	-	-	0.29
RET0.5%	90	10	-	0.5	-	0.29
RET1.0%	90	10	-	1.0	-	0.29
RET1.5%	90	10	-	1.5	-	0.29
RET2.0%	90	10	-	2.0	-	0.29
SP0.05%	90	10	-	-	0.05	0.28
SP0.10%	90	10	-	-	0.10	0.28
SP0.15%	90	10	-	-	0.15	0.28
SP0.20%	90	10	-	-	0.20	0.28

Note: ^ The accelerator, retarder and superplasticizer have been added as a percentage (%) of the binder.

Finally, a system was developed to evaluate the extrusion of each composition based on the number of successfully extruded layers, the consistency of the specimens and the ease of extrusion of the material from the nozzle.

Paste's workability evaluation is carried out by the Flow Table method and based on ASTM C230-14. The measurements were conducted from time 0 min until the initial setting of the paste, every 10 min and the spreading were measured using ImageJ image analysis software. The determination of the initial setting time was carried out using a VICAT apparatus (EN 196-3).

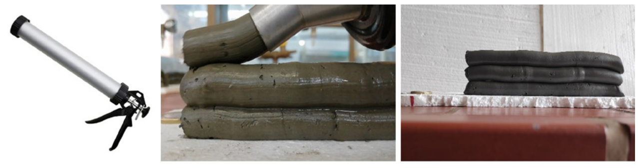
(a) (b) (c)

Figure 1: (a) Extrusion device to which the nozzle is attached, **(b)** Material extrusion through the nozzle, and **(c)** 1st, 2nd and 3rd layer at 0', 10' and 20', respectively.

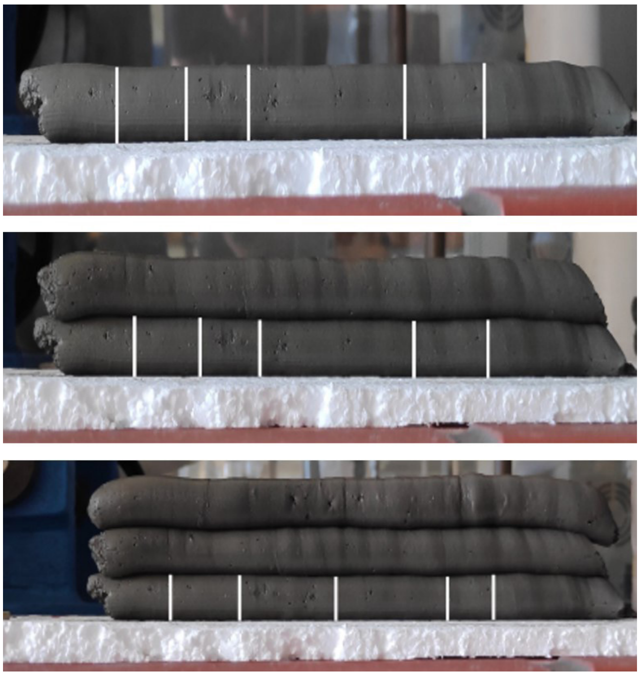

Figure 2: Dimensions measuring using ImageJ image analysis.

Mechanical properties

Mechanical strength in uniaxial compression of 3 specimens of the hardened paste was measured according to ASTM C109-16 after 2, 7, 14, and 28 days of ageing. The criteria that must be satisfied for a material to be classified as printable are, in order, as follows: (1) Be extrudable, (2) Be buildable, and (3) Meet the structure's strength criteria (Figure 3).

Taguchi method

In the second part of the experiment, the adoption of a multifactorial DoE, also known as the Taguchi method, was chosen for the optimization of 3DPC synthesis due to the complexity of the extrusion process. The main benefit of this DoE that has already been used in the optimization of construction materials' production [19], is that it can completely define the inspected process or product with the least number of experiments, saving both time and resources. Minitab™ software was used in this study to design the experiments and statistically analyze the produced data.

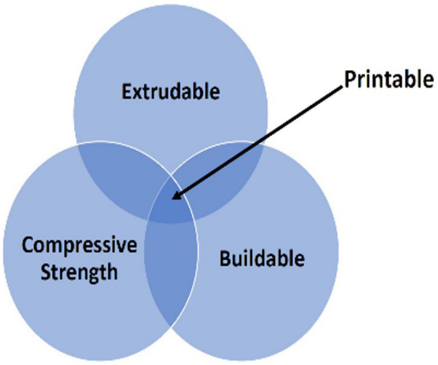

Figure 3: Criteria for printability.

Table 2: Factors for Taguchi method.

Factor	Level 1	Level 2	Level 3
Accelerator	0.50	1.00	1.50
Retarder	1.00	1.50	2.00
Superplasticizer	0.02	0.06	0.10

The DoE objective was to minimize the strain generated on the printed layers during extrusion. According to the preliminary experiments, accelerator, retarder, and superplasticizer addition majorly influences the fresh properties of the prepared pastes and thus were selected as the examined factors of the Taguchi model. Table 2 displays the examined factors along with their value range. The water to binder ratio was kept constant at a value of 0.28.

DoE was set by applying the L9 orthogonal array that significantly reduced the number of requisite experiments from 27 (full factorial DoE) to 9. Table 3 summarizes the designed experiments along with the measured strain after their completion. Finally, Analysis of Variance (ANOVA) was applied to interpret the DoE results.

Results and Discussion

Effect of silica fume addition

Figure 4 presents the effect of silica fume addition in extrusion and initial setting time. No significant improvement was noticed compared to reference sample (REF), since although there is an apparent jump in the SF20% composition this can be attributed to the W/B ratio increase. Silica fumes contribute to the buildability improvement, enhancing the av-

Table 3: Codename, composition (% w/w) and measured strain of the samples generated by multifactorial DoE.

Codename	Accelerator	Retarder	Superplasticizer	Measured Strain			
				Self-weight	Under 2nd layer	Under 3rd layer	Mean
Exp. 1	0.50	1.00	0.02	-0.19	-0.21	-0.22	-0.21
Exp. 2	0.50	1.50	0.06	-0.18	-0.21	-0.23	-0.21
Exp. 3	0.50	2.00	0.10	-0.14	-0.17	-0.17	-0.16
Exp. 4	1.00	1.00	0.06	-0.17	-0.20	-0.21	-0.19
Exp. 5	1.00	1.50	0.10	-0.15	-0.17	-0.18	-0.17
Exp. 6	1.00	2.00	0.02	-0.23	-0.28	-0.25	-0.25
Exp. 7	1.50	1.00	0.10	-0.12	-0.19	-0.18	-0.16
Exp. 8	1.50	1.50	0.02	-0.17	-0.22	-0.23	-0.20
Exp. 9	1.50	2.00	0.06	-0.18	-0.21	-0.21	-0.20

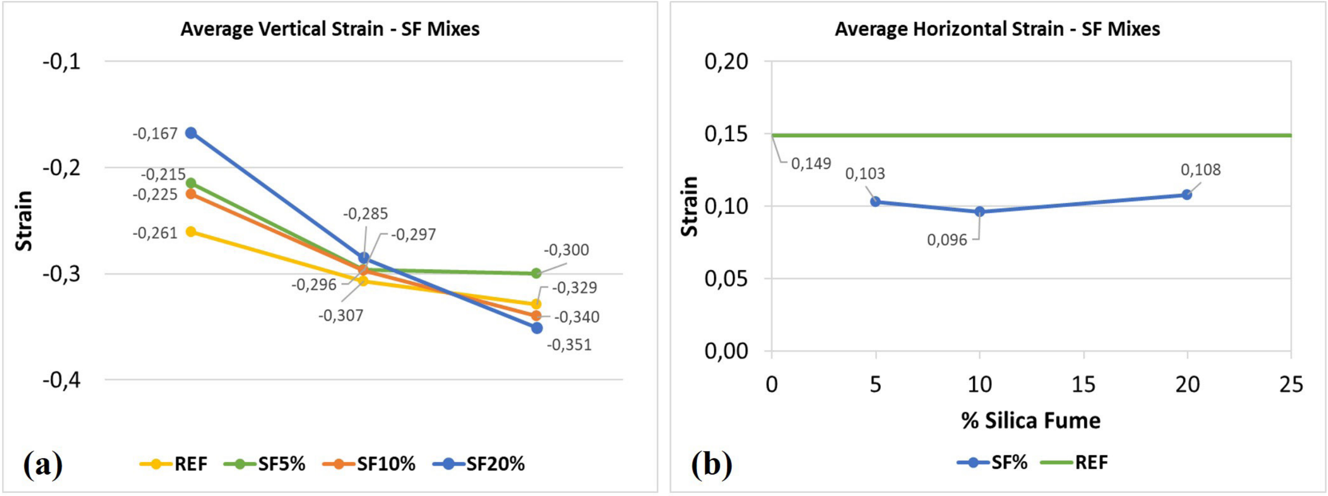

Figure 4: (a) Average vertical deformation of the layers due to the same weight, under one and two layers in the silica gel and REF mixtures and **(b)** Average horizontal deformation of the layers in the silicate and REF mixtures.

erage vertical, especially in case of self-weight, and horizontal strain (Figure 4b). Based on the above results and literature, the composition with 10% SF will be considered the basis for the additives incorporation.

Incorporation of chemical admixtures

Figure 5a shows the results of the extrusion evaluation system. The addition of admixtures significantly improves the extrudability of the blend. The addition of an accelerator to the SF10% blend improves the extrusion of the blend by 50% - 250%. The addition of a retarder to the SF10% mixture improves the extrusion of the mixture by 150% - 350%. And finally, the addition of superplasticizer to the SF10% mixture improves the extrusion of the mixture by 300% - 500%. Figure 5b shows the results of the measurements of the initial setting time of each composition. The addition of admixtures also prolongs the onset of the setting of the mixture. With the addition of an accelerator to the SF10% blend, the setting time start is in the same range (-2% to 4%). With the addition of a retarder to the SF10% blend, the setting time start is extended by 15% to 36%. Finally, with the addition of superplasticizer to the SF10% mixture, the setting time is prolonged by 20% to 81%.

The average horizontal strain of the layers characterizes the buildability of the material. Figure 6 summarizes the percentages of admixtures selected as best per mix, by weight, under one layer and two layers. In general, it is observed that the addition of the silica fume improved the horizontal strain compared to the reference sample. In particular, adding admixtures in the mixture with the silica fume improved the vertical strain even more. The optimum mixture is the one with 1.5 % retarder, which is the composition with the lowest ductility.

During the Workability test, the deformation of the paste over time is observed. The material should retain its workability for as long as possible. Ideally the graph should be straight parallel in x'x axis, and the initial deformation should be around 0.20 - 0.25. More specifically, above this range the constructability is degraded while below it the extrusion is disfavored (resulting from the appearance of the extrusions compared to the spreading table graphs) as a material that theoretically shows zero deformation indicating its unprintability. Conversely, a material that exhibits very high deformation does not retain its shape after printing. Figure 7 summarizes the percentages of admixtures selected as optimum per mixture and the silica fume mixture used as a reference sample. It was observed that the silica fume - admixture combination tends to flatten the curve, which is considered positive. In general, the introduction of admixtures into the mixture of silica fume improves the properties of the printable material. The initial distortion is within the desired limits and the curves

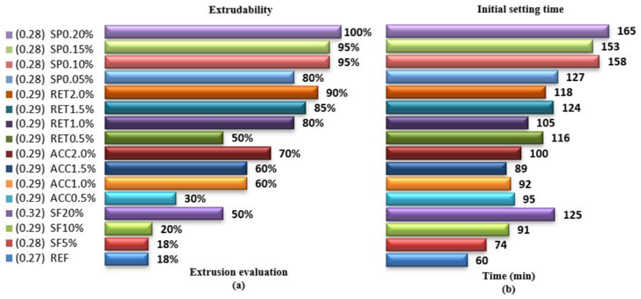

Figure 5: Results of evaluation of **(a)** extrudability and **(b)** initial setting time of compositions.

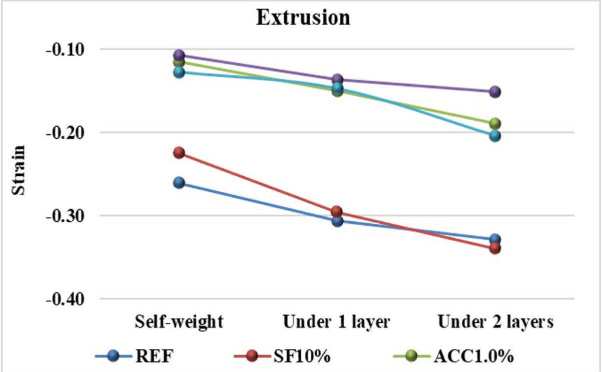

Figure 6: Average vertical strain of the layers for the optimal compositions.

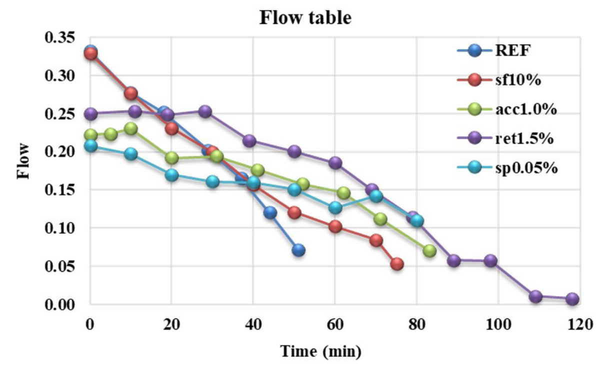

Figure 7: Flow table results for the optimum compositions.

tend to be more strongly horizontal. In particular, the curve of RET1.5% in the interval 0 - 30 min is parallel to the "Time" axis, which makes it ideal in terms of the parallelism criterion, while satisfying the deformability criterion (0.20 - 0.25). That is, the application of such a mixture ensures a continuous extrusion of material without discontinuities and "breaks". The curve of SP0.05% has the optimum initial deformation and tends to horizontalization. The curve of ACC1.0% in the interval 0 - 10 min is considered parallel to the "Time" axis having the desired initial deformation.

Table 4 lists the results of the mechanical strengths at 2, 7, 14, and 28 days of the optimum compositions, based on which: the addition of accelerator at 28 days increases the mechanical strengths by 5 - 8%, the addition of retarder, reduces the strength of the obtained specimens by 4 - 8% and addition of superplasticizer, increases the strength of the obtained specimens by 2 - 15%.

Table 4: Uniaxial compressive strength (MPa) of the optimum compositions.

Time (days)	REF	SF10%	ACC1.0%	RET1.5%	SP0.05%
2	69.9	66.6	64.8	58.4	71.2
7	77.2	87.2	76.8	69.9	86.4
14	94.9	86.8	87.7	76.9	88.2
28	98.2	90.1	96.9	85.3	103.5

In the next step, an effort to explore the combined effect of the examined additives' content on the buildability of 3DCP synthesis is performed through the Taguchi method. Indeed, the performance of the 3DCP synthesis was based on the measured strain of the first layer during the printing process.

The ANOVA results are depicted in figure 8 revealing the effect of the additives' content on the deformation strain of the first extruded layer. The percent contribution of each additive in the buildability of the 3DCP synthesis is also presented as calculated by the significance level. The additive with the greatest impact on the deformation strain is the superplasticizer (84.5%) while the other two additives, accelerator (8.1%) and superplasticizer (7.4%) make a minor contribution to the deformation strain. In particular, the results showed that the strain is kept at a minimum-optimum level when accelerator, retarder and superplasticizer content take the respective values: 1.5, 1.0, and 0.1% wt. The optimized strain value for a 95% confidence interval was calculated as 0.1505 ± 0.0403. A 3DCP synthesis was prepared by applying the optimal additives' contents, to confirm the model prediction. The mean strain measured by the extruded layers was found to be 0.1632 falling within the predicting range and thus confirming the validity of DoE.

From the aforementioned, it became clear that when a 3DCP synthesis combines the examined additives adequate amounts of superplasticizer should be present to produce a flowable paste that can smoothly be extruded from the printing nozzle. In addition, the presence of accelerator seems to benefit the buildability since it helps to keep the appropriate consistency in the cement paste. The fact that retarder has only a marginal effect on the strain of the extruded material indicates that its presence can be beneficial in extending the open time of printing process. Further experimentation is underway to also reveal the combine effect of the additives in other properties (open time, consistency, mechanical properties, etc.) of the printable material.

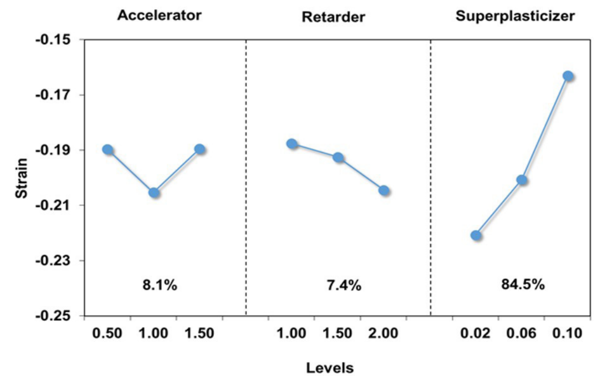

Figure 8: Effect of the additives' content on the deformation strain of the first extruded layer.

Conclusions

In this study, a laboratory testing framework based on the performance of fresh printable mixtures was proposed and an iterative laboratory testing procedure was described to evaluate a printable mixture. A comparison of the results indicates that there is a high probability of drawing an indirect conclusion about the printability properties of a mixture by testing the workability and determining the setting time principle of the composition in question. More specifically, the flow table test helps to draw an indirect conclusion about the printing of the material. The ideal initial deformation in the spreading test, for a printable material, is in the range of about 0.20 - 0.25. Above this range the machinability degrades and below it, the extrusion degrades. Determining the coagulation time principle helps to draw an indirect conclusion about the material's extrudability from the device's nozzle.

A better printable mixture is found to be that containing 10% silica fume and 1.5% retarder, but it shows reduced strengths compared to the initial mixture containing only 10% silica fume, which should be taken into account. As an alternative optimum solution, the mixture containing 10% silica fume and 0.05% superplasticizer can be considered, which shows the most excellent results in terms of extrudability and early setting time, having quite a good workability and machinability compared to the other admixtures, while showing the highest final strengths among all the compositions.

The DoE based on the Taguchi method successfully defines the contribution of each additive in the strain generated during extrusion. The superplasticizer content has the greatest impact while the other two (accelerator and retarder contents) only marginally affect the strain. Superplasticizer and accelerator in adequate amounts help the extrudability and buildability of the printing material since they regulate the material's flowability and consistency, respectively. Deformation strain is almost independent in respect to the retarder's content indicating the capability of its free use to adjust the open time in the 3DCP synthesis.

A future step of this research that will accurately correlate the synthesis of the printable materials with their structure and properties will be the examination of their microstructure through scanning electron microscopy.

Acknowledgements

None.

Conflict of Interest

None.

Funding

The authors received no funding support for this research work.

References

1. Buswell RA, De Silva WL, Jones SZ, Dirrenberger J. 2018. 3D printing using concrete extrusion: a roadmap for research. *Cem Concr Res* 112: 37-49. https://doi.org/10.1016/j.cemconres.2018.05.006
2. Behm JE, Waite BR, Hsieh ST, Helmus MR. 2018. Benefits and limitations of three-dimensional printing technology for ecological research. *BMC Ecol* 18: 1-13. https://doi.org/10.1186/s12898-018-0190-z
3. Rahito, Wahab DA, Azman AH. 2019. Additive manufacturing for repair and restoration in remanufacturing: an overview from object design and systems perspectives. *Processes* 7(11): 802. https://doi.org/10.3390/pr7110802
4. Al-Qutaifi S, Nazari A, Bagheri A. 2018. Mechanical properties of layered geopolymer structures applicable in concrete 3D-printing. *Constr Build Mater* 176: 690-699. https://doi.org/10.1016/j.conbuildmat.2018.04.195
5. Gosselin C, Duballet R, Roux P, Gaudillière N, Dirrenberger J, et al. 2016. Large-scale 3D printing of ultra-high performance concrete–a new processing route for architects and builders. *Mater Des* 100: 102-109. https://doi.org/10.1016/j.matdes.2016.03.097
6. Bos F, Wolfs R, Ahmed Z, Salet T. 2016. Additive manufacturing of concrete in construction: potentials and challenges of 3D concrete printing. *Virtual Phys Prototyp* 11(3): 209-225. https://doi.org/10.1080/17452759.2016.1209867
7. Paul SC, van Zijl GP, Tan MJ, Gibson I. 2018. A review of 3D concrete printing systems and materials properties: current status and future research prospects. *Rapid Prototyp J* 24(4): 784-798. https://doi.org/10.1108/RPJ-09-2016-0154
8. Marchment T, Sanjayan JG, Nematollahi B, Xia M. 2019. Interlayer Strength of 3D Printed Concrete: Influencing Factors and Method of Enhancing. In Sanjayan JG, Nazari A, Nematollahi B (eds) 3D Concrete Printing Technology. Butterworth-Heinemann, pp 241-264.
9. Nematollahi B, Xia M, Sanjayan J. 2017. Current progress of 3D concrete printing technologies. In 34th International Symposium on Automation and Robotics in Construction Taipei, Taiwan.
10. Panda B, Paul SC, Mohamed NAN, Tay YWD, Tan MJ. 2018. Measurement of tensile bond strength of 3D printed geopolymer mortar. *Measurement* 113: 108-116. https://doi.org/10.1016/j.measurement.2017.08.051
11. Sanjayan JG, Nematollahi B, Xia M, Marchment T. 2018. Effect of surface moisture on inter-layer strength of 3D printed concrete. *Constr Build Mater* 172: 468-475. https://doi.org/10.1016/j.conbuildmat.2018.03.232
12. Kazemian A, Yuan X, Cochran E, Khoshnevis B. 2017. Cementitious materials for construction-scale 3D printing: laboratory testing of fresh printing mixture. *Constr Build Mater* 145: 639-647. https://doi.org/10.1016/j.conbuildmat.2017.04.015
13. Ma G, Wang L, Ju Y. 2018. State-of-the-art of 3D printing technology of cementitious material-an emerging technique for construction. *Sci China Technol Sci* 61: 475-495. https://doi.org/10.1007/s11431-016-9077-7
14. Vaitkevičius V, Šerelis E, Kerševičius V. 2018. Effect of ultra-sonic activation on early hydration process in 3D concrete printing technology. *Constr Build Mater* 169: 354-363. https://doi.org/10.1016/j.conbuildmat.2018.03.007
15. Malaeb Z, AlSakka F, Hamzeh F. 2019. 3D concrete printing: machine design, mix proportioning, and mix comparison between different machine setups. In Sanjayan JG, Nazari A, Nematollahi B (eds) 3D Concrete Printing Technology. Butterworth-Heinemann, pp 115-136.
16. Han J, Wang K, Shi J, Wang Y. 2015. Mechanism of triethanolamine on Portland cement hydration process and microstructure characteristics. *Constr Build Mater* 93: 457-462. https://doi.org/10.1016/j.conbuildmat.2015.06.018
17. Le TT, Austin SA, Lim S, Buswell RA, Gibb AG, et al. 2012. Mix design and fresh properties for high-performance printing concrete. *Mater Struct* 45: 1221-1232. https://doi.org/10.1617/s11527-012-9828-z
18. Le TT, Austin SA, Lim S, Buswell RA, Law R, et al. 2012. Hardened properties of high-performance printing concrete. *Cem Concr Res* 42(3): 558-566. https://doi.org/10.1016/j.cemconres.2011.12.003
19. Kioupis D, Skaropoulou A, Tsivilis S, Kakali G. 2018. The application of experimental design models in order to optimize the synthesis of geopolymers. In 2nd International Congress on Materials and Structural Stability, Rabat, Morocco.