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Empirical models to predict rheological properties of fiber reinforced cementitious composites for 3D printing

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ABSTRACT

3D printable construction materials need to be conveyed through a delivery system whilst possess certain flow resistance to ensure materials can sustain the weight of subsequent layers. To meet these requirements, material rheological properties should be optimized. In this study, factorial design was adopted to evaluate the influences of five variables (water-to-binder ratio, sand-to-binder ratio, fly ash-to-cement ratio, silica fume-to-cement ratio, and dosage of fiber) on material rheological properties (flow resistance, torque viscosity and thixotropy). Empirical models were established to predict rheological properties and were verified by experiment. Results imply that the increment of the dosage of fiber boosts all the rheological parameters, which are declined with the increment of water-to-binder ratio. Torque viscosity raises while flow resistance and thixotropy are decreased with the rise of fly ash-to-cement ratio. Conversely, the influence of silica fume-to-cement ratio shows an opposite trend on rheological properties as compared to that of fly ash-to-cement ratio. Flow resistance and torque viscosity are improved whilst thixotropy is declined if sand-to-binder ratio increases. Different formulations were adopted in printing test for verification and demonstration purpose via a robotic arm printing system in the end.

Keywords: rheological properties; 3D cementitious materials printing; thixotropy; statistical design.

1. Introduction

3D printing technology for cementitious materials builds up objects from successive cross section layers [1]. In a 3D printing process, materials are firstly premixed and conveyed to a nozzle head via a pumping system. A printing system then controls nozzle movement to deposit materials [1–4] based on a CAD model. To ensure a successful printing process, both robust printing system and consistent printable cementitious materials are essential.

Several types of printing systems have been developed in recent years, such as the Contour Crafting system [5], the mortar printer [6] and the robotic concrete printer [7,8]. Besides printing systems, the cementitious materials also plays a crucial role in a successful 3D printing process [9–15], which needs the printable materials to meet pumpability and buildability requirements [16–21]. Material pumpability is directly associated with pumping pressure in the delivering phase of the 3D printing process, and buildability defines the height of the printed structure. In recent studies, researchers have revealed that rheological properties, especially static/dynamic yield stress and plastic viscosity, were critical material characteristics for printable materials to meet such standards. Material constituents serve as vital factors that influence material rheological properties.

Intensive research has been conducted to investigate the influence of material constituents on rheological properties [22–26]. However, limitations still remain. Firstly, previous works mainly reached qualitative conclusions. Few quantitative result was established to quantify the impact of material constituents on rheological properties. Besides, research needs to be carried out to explore the influence of material constituents on thixotropy, which is an essential time-dependent parameter for the printing process. To overcome these limitations, an efficient approach should be adopted for the experimental design to reduce experimental runs. Meanwhile, quantitative models should be built to provide guidance for the predictions of rheological properties [27]. The influence of material constituents on thixotropy also needs further exploration.

Design of Experiment (DoE) is a scientific experimental methodology for experimental design and data analysis based on fundamental mathematical statistics [28]. As a powerful approach to

exploring the correlation between factors and responses, DoE has been successfully used in various research fields [27,29–31]. One of useful methods that adopt DoE is called factorial design, which quantifies the influences of variables on responses through constructing statistical models.

The factorial design is more efficient than traditional one-factor at a time experiment. The method can simplify the experimental process and reduce experimental runs, but enough information still can be extracted for data analysis. Meanwhile, the method can allow the influence of a factor to be estimated at several levels of other factors.

The focus of this study is to construct statistical models to predict how different factors influence rheological properties by adopting the factorial design. The pre-determined factors in this work are sand-to-binder ratio, water-to-binder ratio, fly ash-to-cement ratio, silica fume-to-cement and the dosage of fiber. The responses are flow resistance, torque viscosity and thixotropy of materials. The analysis of variance (ANOVA) was adopted to conduct statistical analysis and create statistical models for predictions. Experimental results indicate that the statistical models can be used to predict material rheological properties with appropriate degree of accuracy. Although the constructed statistical models may not be universally applicable due to change in material chemical composition, different particle size distribution, etc, the validation test indicates that factorial design is efficient to find the desirable formulation within a given boundary.

2. Background and Experimental Design

2.1. Bingham model

Rheology describes the deformation and flow behavior of materials [32,33]. Bingham model is commonly used for cement pastes due to its simplicity and wide acceptance [34,35]. The correlation between shear stress τ (Pa) and shear rate $\dot{\gamma}$ (1/s) in the Bingham model is described as follows:

$$\tau = \tau_0 + k \dot{\gamma} \quad (1)$$

where τ_0 (Pa) and k (Pa·s) are yield stress and plastic viscosity, respectively. Yield stress τ_0 represents minimum shear stress required to initiate or maintain the flow of a material, while plastic viscosity k describes the change of shear stress with altering shear rate. These two rheological parameters are measured by a rotational rheometer test, in which Bingham model can also be expressed as the following formula for convenience of experiment design and data analysis:

$$T = G + hN \quad (2)$$

Eq. (2) describes the correlation between the measured torque T (N·m) and rotational speed N (rpm). The parameter G (N·m) is flow resistance, representing the minimum torque required to initiate or maintain the flow of a material. The parameter h (N·m·min) is torque viscosity. Similar to the k in Eq. (1), the parameter h describes the change of applied torque with altering rotational speed.

Bingham model indicates that if both parameters (yield stress τ_0 and plastic viscosity k ; or equivalently, flow resistance G and torque viscosity h) are kept at a higher level, the material is difficult to flow at any given speed. As indicated in the study of pumpable concrete, the rise of τ_0 and k yields the increment of pumping pressure [36], which is undesirable to the pumping process and delivering process. In the printing phase, the restraining deformation due to subsequently printed layers requires a high yield stress τ_0 [16,37]. Considering the delivering phase and the printing phase as a whole process, yield stress should be enhanced whilst plastic viscosity should be kept at a low level to achieve appropriate rheological control of 3D printable fiber reinforced cementitious composites for a successful printing process.

2.2 Time-dependent effect of rheology

The correlations between buildability, pumpability and rheological properties are expressed as follows [37]:

$$H = \frac{\alpha}{\rho g} \tau(t) \quad (3)$$

$$P = \left[\frac{8\tau(t)}{3R} + \frac{8k(t)}{\pi R^4} Q \right] L \quad (4)$$

where H (mm) and P (Pa) are printed height and pumping pressure, respectively; R (mm) and L (m) are the radius and length of a hose, respectively; Q (m³/s) is an average flow rate. ρ (g/cm³) and g (m/s²) are the density of materials and gravitational constant, respectively.

The rheological properties evolve with time due to the hydration process. Roussel [38] proposed a model to quantify time-dependent effect of yield stress that is shown as follows:

$$\tau_0(t) = \tau_0(0) + A_{thix}t \quad (5)$$

where t is resting time; A_{thix} is thixotropy parameter, which is assumed to be a constant for a given material; $\tau_0(0)$ is yield stress at resting time zero. Generally a high A_{thix} is required to drive up yield stress to make certain that materials possess enough buildability for a 3D printing process [37].

The time effect on dynamic viscosity is expressed as follows [36]:

$$\mu_0(t) = \mu_0(0) + (1000 - \mu_0(0))(t/t_v)^n \quad (6)$$

where t_v is the moment at which the slurry reaches 1000 Pa·s, $\mu_0(0)$ is viscosity at resting time zero. However, yield stress is greatly higher than plastic viscosity in general. Compared with the influence of plastic viscosity on the pumping pressure, the influence of yield stress on pumping pressure is the dominant factor.

2.3. 2^k design and 2^{k-p} fractional design

2^k design (factorial design) is a special design method in DoE [39]. In 2^k design, each factor has two levels, i.e. high level and low level. The total number of experiments is 2^k if there are k factors. The 2^k design can effectively reduce a total number of experiments via optimizing experimental runs so that the method saves time and efforts in the material design stage and more factors can be studied in experiments that those studied in conventional experimental design. Additionally, effect values can be calculated through Analysis of Variance (ANOVA). A total number of 2^k experiments can be further reduced via the 2^{k-p} fractional design, in which p means

the number of independent generators. The design method reduces the total number of experiments to $1/2^p$ of the original 2^k design [39].

3. Material properties, mixture proportions and printing investigation

3.1. Materials properties

Cement slurries were produced from Ordinary Portland cement (OPC, ASTM type I, Grade 42.5), silica fume (SF, undensified, Grade 940, Elkem Pte. Ltd.), silica sand (S), fly ash (FA, Class F), polyvinyl alcohol (PVA) fibers (Kuraray Pte. Ltd.) [40,41], water and superplasticizers (SP, 181N, Grace Pte. Ltd.). Particle size distributions of cement, silica fume, silica sand and fly ash are depicted in Fig. 1. The physical properties of powder ingredients were analyzed by Mastersizer 2000 and BET surface area analyzer and results are shown in Table 1. The chemical compositions of OPC and FA are summarized in Table 2. Characteristics of PVA fiber are presented in Table 3.

Fig. 1 Particle size distribution of powder ingredients

Table 1 Physical properties of powder materials used in this study

	Density(g/cm ³)	Specific surface area(m ² /g)
OPC	3.15	2.66
FA	1.3	0.285
SF	2.2-2.5	7.92
Silica Sand	2.66	0.305

Table 2 Chemical composition of fly ash and Ordinary Portland Cement (OPC)

Formula	Concentration / %	
	Fly ash	Cement
SiO ₂	58.59	24.27
Al ₂ O ₃	30.44	4.56
Fe ₂ O ₃	4.66	3.95
TiO ₂	2.02	0.55
K ₂ O	1.51	0.61
CaO	1.21	62.2
MgO	0.776	3.34
P ₂ O ₅	0.531	0.15
Na ₂ O	-	0.21
SO ₃	0.0914	-
ZrO ₂	0.04	-
MnO	0.0351	-
Cr ₂ O ₃	0.027	-
CuO	0.0254	-
ZnO	0.0229	-

Table 3 Characteristics of PVA fiber

Tensile Strength/ MPa	1560
Modulus/GPa	41
Diameter/ μ m	40
Length/mm	8
Elongation at break/%	6.5
Oil coating	1.2%

3.2. Mixture proportions

In this study, the rheological properties of fiber reinforced cementitious materials for 3D printing are measured via the fractional design. Firstly, a 2^{5-1} experiment is designed as shown in Table 4, in which the total number of experiments should be conducted is 16. Herein, A, B, C, D, and E represent sand-to-binder ratio (S/B), water-to-binder ratio (W/B), fly ash-to-cement ratio (FA/OPC), silica fume-to-cement ratio (SF/OPC) and the dosage of fiber, respectively. ABCDE is then selected as the generator. Binder means the total mass of OPC, FA and SF. The content of S/B, W/B, FA/OPC, SF/OPC is expressed by the mass while the dosages of fiber are expressed by volume. Finally, new experiment runs (2^{5-1} design) can be designed by four factors (A, B, C, and D) [42].

Coded values and actual values of fractional factorial design are shown in Table 5 and 6 respectively, in which BCD and ABE are selected as generators. D and E are expressed by BC and AB in the tables, respectively. Taking formulation No.1 for example: the high level and low level of S/B (factor A) are 0.28 and 0.3, respectively; therefore, the mean value of S/B is 0.29 with 0.02 as the range between high level and low level; when the S/B is at a low level, the factor mean and range of S/B are then substituted into Eq. (5) to attain the coded value. Similarly, the formula can be applied when the factor is at a high level.

Table 4 2^{K-1} design matrix

No.	Run	A	B	C	D	E/%
1	(-)	0.28	0.30	1	0	1
2	a	0.30	0.30	1	0	0
3	b	0.28	0.32	1	0	0
4	ab	0.30	0.32	1	0	1
5	c	0.28	0.30	2	0	0
6	ac	0.30	0.30	2	0	1
7	bc	0.28	0.32	2	0	1
8	abc	0.30	0.32	2	0	0
9	d	0.28	0.30	1	0.1	0
10	ad	0.30	0.30	1	0.1	1
11	bd	0.28	0.32	1	0.1	1
12	abd	0.30	0.32	1	0.1	0
13	cd	0.28	0.30	2	0.1	1
14	acd	0.30	0.30	2	0.1	0
15	bcd	0.28	0.32	2	0.1	0
16	abcd	0.30	0.32	2	0.1	1

*The SP used in all mixture is equal to 1.8 g/L.

Table 5 Coded table of fractional factorial design matrix

No.	A	B	C	D	E
1	-1*	-1	-1	1*	1
2	-1	-1	1	-1	1
3	-1	1	-1	-1	-1
4	-1	1	1	1	-1
5	1	-1	-1	1	-1
6	1	-1	1	-1	-1
7	1	1	-1	-1	1
8	1	1	1	1	1

-1 and 1 represents high and low level respectively

Table 6 Absolute values of fractional factorial design matrix

No.	A	B	C	D	E	SP/(g/L)
1	0.28	0.30	1	0.1	1	1.8
2	0.28	0.30	2	0	1	1.8
3	0.28	0.32	1	0	0	1.8
4	0.28	0.32	2	0.1	0	1.8
5	0.30	0.30	1	0.1	0	1.8
6	0.30	0.30	2	0	0	1.8
7	0.30	0.32	1	0	1	1.8
8	0.30	0.32	2	0.1	1	1.8

$$\text{Coded value} = (\text{Actual value} - \text{Factor mean}) / (\text{Range of factorial value} / 2) \quad (7)$$

3.3. Mixing, testing procedures and printing investigation

Rheological properties are affected by various factors, such as mixer types, mixing procedures, mixing speed [37], temperature [22] and the timing of the SP addition [16,37]. Thus, all the aforementioned factors were fixed in this study. Six liters mortars were prepared in the Hobart mixer X200L with a 20 L capacity. The environmental temperature was fixed at 26 °C. Fig. 2 shows mixing procedures. The rheological test was then conducted by a Viskomat XL with a fish-bone probe.

Fig. 2 Mixing procedure

Flow resistance, torque viscosity and thixotropy can be attained via the rheological test. Fig. 3 shows the typical test program [22,37], in which test speed is increased from 0 rpm to 60 rpm in

5 minutes and then reversed from 60 rpm to 0 rpm in another 5 minutes. Fig. 4 shows a characteristic hysteresis loop obtained in this test. According to Bingham model [37], fitting line's slope of the descending curve in Fig. 4 represents the torque viscosity of pastes whilst the intersection between a fitting line and the Y axis stands for flow resistance. The area between ascending and descending curves is the thixotropy of tested materials.

Fig. 3 Rheological testing programs

Fig. 4 Typical rheological test result

The printing tests were then conducted by a robotic printing system [7] as shown in Fig. 5. Premixed materials were firstly poured into a black funnel (Fig. 5 (b)) sand was then pumped to a nozzle head, which was fixed on the robotic arm that controlled the nozzle to print an object based on a CAD model (Fig. 5 (c)). The CAD model is 75 mm in triangle edge length and 9 layers.

225

226

Fig. 5 The robotic printing system: (a) the robotic arm; (b) the pictor pump; (c) designed CAD model

4. Results and discussion

4.1 Rheological results

Table 7 shows rheological test results that were further analyzed via the ANOVA, which was adopted to investigate whether factors have statistically significant effects on responses. The analysis result enables researchers to identify major trends and to predict rheological properties according to derived models. The detailed analysis results are discussed in following sections.

Table 7 Rheological properties results measured via factorial design

No.	S/B	W/B	FA/OPC	SF/OPC	Fiber/%	Torque Viscosity /N·mm·min	Flow resistance /N·mm	Thixotropy/ (N·mm·min ⁻¹)
1	0.28	0.30	1	0.1	1	10.7	1088	813.0
2	0.28	0.30	2	0	1	9.03	141.4	2126
3	0.28	0.32	1	0	0	5.22	99.11	192.5
4	0.28	0.32	2	0.1	0	4.16	159.1	656.2
5	0.30	0.30	1	0.1	0	6.09	465.5	1484
6	0.30	0.30	2	0	0	14.3	122.1	4653
7	0.30	0.32	1	0	0	4.90	110.1	1008
8	0.30	0.32	2	0.1	1	8.75	438.3	4578

4.2 ANOVA analysis and derived models

A software of Design Expert [30] was used to carry out the ANOVA analysis based on the data listed in Table 7 and analyzed results are presented in Table 8, 9 and 10. F -value, F_0 , of each factor was then calculated. The significance of each factor was obtained with a confidence level by comparison of calculated F_0 and the control value in the F-distribution chart.

Table 8 ANOVA analysis result for flow resistance

Source	DOF	F-Value	Confidence level/%
S/B	1	1.08	64
W/B	1	0.81	57
FA/C	1	6.02	82
SF	1	9.74	94
F	1	12.33	97
Error	2	—	—
Total	7	—	—

$$F_{0.3,1,2} = 1.92; F_{0.2,1,2} = 3.55; F_{0.15,1,2} = 5.20; F_{0.1,1,2} = 8.52;$$

(Note: above F_{α, ν_1, ν_2} is acquired from the F-distribution chart. $1-\alpha$, ν_1 and ν_2 represent confidence interval, the degree of freedom (DOF) of investigated factor and error, respectively.)

Table 9 ANOVA analysis results for torque viscosity

Source	DOF	F-Value	Confidence level/%
S/B	1	1.11	64
W/B	1	0.84	57
FA/C	1	1.01	60
SF	1	0.36	40
F	1	2.36	80
Error	4	—	—
Total	9	—	—

Table 10 ANOVA analysis results for thixotropy

Source	DOF	F-Value	Confidence level/%
S/B	1	3.82	80
W/B	1	7.62	89
FA/C	1	4.25	82
SF	1	1.53	66
F	1	1.22	62
Error	4	—	—
Total	9	—	—

Table 8 illustrates that the dosage of fiber and SF/OPC have significant influence on the flow resistance at a very high confidence level, 97 % and 94 %, respectively, followed by FA/OPC with a confidence interval of 82 %. To state that S/B and W/B have significant influence on flow resistance, the confidence level is only 64 % and 57 %, respectively. Table 9 indicates the dosage of fiber has significant effects on the torque viscosity at a high confidence interval (80 %), followed by S/B, FA/OPC and W/B with confidence intervals of 64 %, 60 %, and 57%, respectively. However, the confidence level of SF/OPC is at 40 %. Table 10 shows S/B, W/B and FA/OPC possess significant influence on thixotropy at a high confidence level (above 80 %), followed by SF/OPC and the dosage of fiber with confidence intervals of 66 % and 62 %, respectively.

Table 11 Effect of each factor on flow resistance, torque viscosity and thixotropy

Factors	Intersection	S/B	W/B	FA/OPC	SF/OPC	Fiber
Parameters	a	A	B	C	D	E
Torque viscosity	7.89	1.74	-3.70	3.96	-0.49	2.73
Flow resistance	327.89	2.71	-194.03	-145.99	419.18	287.42
Thixotropy	3072	-2125	-822.2	-185.1	1345	397.5

Changes in responses induced by changes of a given factor are shown in Table 11. Positive or negative numbers indicate that the factor possesses positive or negative influence on the responses, respectively. The greater a number, the more influence a factor has on a response. Obtained parameters of derived models were based on effects of each factor as shown in Table 11.

Table 11 presents the dosage of fiber have positive effects on flow resistance, torque viscosity and thixotropy, whereas increasing W/B declines flow resistance, torque viscosity and thixotropy. Larger SF/OPC raises flow resistance and thixotropy but reduces torque viscosity, whereas FA/OPC presents an opposite trend. S/B possesses positive influence on torque viscosity and flow resistance while negative influence on thixotropy. From the analysis above, SF/OPC seems to be a type of critical ingredients for 3D printable cementitious materials. It not only has significant effects on flow resistance, thixotropy with a high confidence level but also shows a desirable trend for flow resistance, torque viscosity and thixotropy, i.e. positive for flow resistance and thixotropy but negative for torque viscosity.

Effect values of each factor on responses also represents the coefficient of each factor in the derived models. Therefore, the numbers in Table 11 are also the coefficients of statistical rheology models, as shown in Eqs. (8-10). These models correlate rheological properties with various factors, where A, B, C, D, E correspond to S/B, W/B, FA/OPC, SF/OPC and dosages of fiber, respectively.

$$\text{Flow resistance} = 327.99 + 2.71 \times A - 194.03 \times B - 145.99 \times C + 419.18 \times D + 287.42 \times E \quad (8)$$

$$\text{torque viscosity} = 7.89 + 1.74 \times A - 3.70 \times B + 3.96 \times C - 0.49 \times D + 2.73 \times E \quad (9)$$

$$\text{Thixotropy} = 3072 - 2125 \times A - 822.2 \times B - 185.1 \times C + 1345 \times D + 397.5 \times E \quad (10)$$

293

294 The accuracy of models was estimated by statistical analysis. The results of prediction models
295 are listed in Appendix. Fig. 6, 7, and 8 show the normability tests, where all the points scattering
296 around a straight line. The result indicates that residuals followed a normal distribution, and flow
297 resistance, torque viscosity and thixotropy cannot be improved by adopting other curve fitting
298 methods.

299

Fig. 6 Normability test: (a) flow resistance; (b) torque viscosity; (c) thixotropy

To exclude the influence of run order on the statistical rheological models, the correlation between the residuals and run order should be confirmed. Fig. 7 depicts that the points scatter randomly (no grouping) with the run order and the result indicates that the run order has no significant effects on the responses. Ideally, the model error (residuals) will consist of normally distributed random variation from the experimental process and measurement systems. Fig. 7 shows the normal probability plots of the residuals for flow resistance, plastic viscosity, and thixotropy. Fig. 7 exhibits straight lines for the normal probability plots of the residuals with no outliers or groupings in the data. The result implies that the residuals are normally distributed describing the experimental noise.

Fig. 7 Residuals analysis: (a) flow resistance; (b) torque viscosity; (c) thixotropy

Fig. 8 presents the correlation between prediction results and actual rheological test results based on Tables A.1 to A.3. Apparently, the models can still largely capture the correlation between

factors and responses (flow resistance, torque viscosity and thixotropy) while second-order interaction effects are ignored.

Fig. 8 Predicted vs actual result: (a) flow resistance; (b) torque viscosity; (c) thixotropy

Afterwards, three additional formulations were randomly selected to verify the accuracy of the derived models. The formulations and rheological test results are shown in Table 12 and 13, respectively. Table 13 implies the model can largely predict experimental results well despite the random selection of formulations. Nevertheless, the accuracy of these first-order models can still be improved, and more accurate models with higher-order interactions incorporated are required to predict rheological properties. However, this is beyond the scope of this study.

Table 12 Formulations selected for verification

No.	S/B	W/B	FA/OPC	SF/OPC	Fiber/%
1	0.3	0.3	1	0.05	0
2	0.28	0.3	2	0	1
3	0.28	0.32	2	0.1	0

Table 13 Rheological test results

Parameters	No.	Experiment	Model	Error/%
Viscosity / N·mm·min	1	9.17	9.37	2.18
	2	10.69	13.81	29.2
	3	6.91	6.41	7.24
Flow resistance/ N·mm	1	384.5	383.3	0.312
	2	225.5	241.6	7.14
	3	93.23	117	25.5
Thixotropy/ N·mm·min ⁻¹	1	506.7	609.3	20.3
	2	4146	4587	10.6
	3	6024	5534	8.13

4.3 Demonstration of 3D printable FRCC

Finally, miniature structures were printed for verification and demonstration purpose via a robotic printing system [7]. No.1 formulation in Table 7 was selected for the purpose as it possesses the highest flow resistance among all formulations, 1088 N·mm, and low torque viscosity, 10.7 N·mm·min. No.3 formulation, possessing lowest flow resistance among all formulations, was also selected as the control. Raw ingredients were firstly mixed in a Hobart

341 mixer. The paste was then conveyed from a concrete pump to a nozzle which was fixed on a
342 robotic arm. The robotic arm then printed the paste with the intended shape as the CAD model.
343 The robotic arm movement speed and pumping speed were 100 mm/s and 2.5 L/min,
344 respectively. A conveyance hose used in the printing process was 25.4 mm in diameter and 3 m
345 in length. A nozzle with 8 mm in diameter was used for the printing test.

346
347 The final printed hexagon part of formulation No. 1 is shown in Fig. 9. The printed part is with
348 an edge length of 74.83 mm and height of 84.33 mm. The final printed of formulation No.3 is
349 plotted in Fig. 10, which indicates that material yield stress was insufficient to sustain
350 subsequently printed layers such that serious deformation occurred.

352
353 Fig. 9 Final printed part (formulation No. 1): (a) top view; (b) side view

Fig. 10 Final printed part (formulation No.3): (a) top view; (b) side view

5. Conclusions

Factorial design was adopted in this research to minimize the experimental runs while the experimental results still retain essential information for data analysis. The method is particularly useful when one has to deal with a great number of variables in an experiment, e.g. in search of printable materials with desirable rheological properties. Confidence interval, first-order models and the effect values were attained via the factorial design that possesses several advantages compared with a traditional variable-controlling method, e.g. the method can save time and resources as it minimizes the number of experimental runs. The method can also be used to derive statistical models for further predictions.

The influences of material constituents on flow resistance, torque viscosity and thixotropy are explored in this work. Results indicate that flow resistance and torque viscosity are improved with the increment of the dosage of fiber. S/B possesses positive influence on torque viscosity and flow resistance while negative influence on thixotropy. Flow resistance, viscosity and thixotropy are dropped with the rise of W/B. Torque viscosity is boosted while flow resistance and thixotropy are declined with raising FA/OPC. However, compared with the influence of FA/OPC on rheological properties, the opposite trend remains with the increment of SF/OPC, which is desirable for 3DP materials. Finally, silica fume is an appropriate ingredient to adjust rheological properties for the 3D cementitious materials printing process.

The dosage of fiber and SF/OPC have significant influence on flow resistance at a very high confidence level (more than 90 %), followed by FA/OPC with a confidence interval of 82 %. However, the confidence level of S/B and W/B on flow resistance is only at around 60 %. The dosage of fiber dosage has significant effect on the torque viscosity at a relatively high confidence interval (80 %), followed by S/B, FA/OPC and W/B with confidence intervals at around 60 %. However, the confidence level of SF/OPC is only at 40 %. S/B, W/B and FA/OPC show significant influence on thixotropy at a relatively high confidence level (above 80 %), followed by SF/OPC and the dosage of fiber with confidence intervals at around 65 %.

High flow resistance and low viscosity are necessary for the 3D printing process. Rheological models are highly needed for researchers to design their material formulations according to the rheological properties required for the 3D printing process. Although the first-order models derived in this work can be further improved, the models can still act as initial guidance for the control of the rheological properties of printable materials to achieve a successful printing process. At the next stage, second-order models will be developed to improve the degree of prediction accuracy of rheological properties, which is beyond the scope of this study.

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Conflicts of interest

The authors declare no conflict of interest.

Appendix A. Analysis results of prediction models

Table A.1 Statistical analysis results of a prediction model of flow resistance

Run order	Actual value	Predicted value	Residual	Internally studentized residuals
1	141	225	-83.9	-0.536
2	466	382	83.9	0.536
3	110	194	-83.9	-0.536
4	122	37.8	83.9	0.536
5	99.1	-57.6	157	1.87
6	1088	931	157	1.87
7	122	279	-157	-1.87
8	438	595	-157	-1.87

Table A.2 Statistical analysis results of a prediction model of torque viscosity

Run order	Actual value	Predicted value	Residual	Internally studentized residuals
1	9.03	8.77	0.256	0.143
2	6.09	6.34	-0.256	-0.143
3	4.90	4.64	0.256	0.143
4	6.82	7.07	-0.256	-0.143
5	5.22	7.00	-1.78	-6.94
6	10.7	12.0	-1.78	-6.94
7	14.3	12.5	1.78	6.94
8	8.75	6.96	1.78	6.94

Table A.3 Statistical analysis results of a prediction model of thixotropy

Run order	Actual value	Predicted value	Residual	Internally studentized residuals
1	813	1257	-444	-0.209
2	2126	1682	444	0.209
3	193	637	-444	-0.209
4	656	212	444	0.209
5	1485	3614	-2129	-4.79

6	4654	6783	-2129	-4.79
7	10077	7948	2129	4.79
8	4578	2449	2129	4.79

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