Journal of Building Engineering 74 (2023) 106814

Contents lists available at ScienceDirect

Journal of Building Engineering

journal homepage: www.elsevier.com/locate/jobe

Effect of Magnetorheological additives on the buildability of 3D concrete printing

Sasitharan Kanagasuntharam, Sayanthan Ramakrishnan *, Shravan Muthukrishnan, Jay Sanjayan

Centre for Sustainable Infrastructure and Digital Construction, School of Engineering, Swinburne University of Technology, Hawthorn, Australia

ARTICLE INFO

Keywords:
Additive manufacturing
Set-on-demand
Magnetorheological (MR) fluid
Active rheology control (ARC)
Active stiffening control (ASC)

ABSTRACT

This study investigates the effect of magneto-rheological additives in 3D concrete printing as a stimuli-responsive technique for buildability enhancement. The carbonyl iron powder (CIP) was used as the magnetorheological additive. Two distinct magnetic activation strategies of print-bed magnetisation and print-head magnetisation were considered. The fresh properties including static yield strength development and viscosity representing the buildability and pumpability characteristics of magnetorheological cement mortar (MRC) mixtures were assessed for both strategies. Besides, the compressive strength and volume of permeable voids (VPV) were studied to assess the effect of CIP on the hardened properties of MRC mixtures. It was found that the addition of CIP had a significant enhancement in the static yield strength of fresh MRC mixtures when the magnetic field is continuously present (i.e., print-bed magnetisation). However, short-duration exposure to the magnetic field (i.e., print-head magnetisation) resulted in a slight improvement in static yield strength, revealing that the residual effect of MRC mixtures is insignificant. For instance, the MRC mixture with a CIP dosage of 10% by weight exhibited a static yield strength of 15 kPa at 20 min when subjected to continuous magnetisation, whereas the same mixture showed a static yield strength of 5 kPa at 20 min for short-duration magnetisation (the exposure time of 60 s). The hardened properties of CIP incorporated MRC mixtures showed the enhancement in compressive strength and reduction in volume of permeable voids for CIP dosage up to 5% with further increase in CIP dosage lead to strength reductions.

1. Introduction

3D Concrete printing (3DCP) is gaining enormous attention among construction industries and research institutes due to its various advantages, such as formwork-free construction, design flexibility, sustainability, etc. 3DCP also has immense potential to be a fast and economical construction method given the technology reaches its maturity stage by overcoming the current challenges in concrete technology. The 3D printing process can be divided into four major stages – mixing, pumping, extrusion, and building [1–3]. During the first three stages, the mixture is prepared, transported to the print head and extruded from a nozzle for printing respectively. In these three stages, the mixture should exhibit low viscosity and static yield strength for smooth operation. Here, static yield strength and viscosity represents the resistance to flow initiation and flow propagation respectively. Low viscosity and static yield strength mixture will have minimum resistance during the mixing, pumping and extrusion stages.The fourth stage is the building or placement

* Corresponding author.
E-mail address: sramakrishnan@swin.edu.au (S. Ramakrishnan).

https://doi.org/10.1016/j.jobe.2023.106814
Received 26 March 2023; Received in revised form 4 May 2023; Accepted 9 May 2023
Available online 11 May 2023

of concrete to create 3D objects. At this stage, the concrete needs to exhibit contradictory rheology to the previous stages. This is partially achieved by the thixotropic behaviour of the concrete. Thixotropy is defined as the ability of the mixture to develop viscosity and static yield strength with time when kept at rest. During pumping, mixing and extrusion stage, the concrete is subjected to shearing which reduces the viscosity and (dynamic) yield strength of the concrete. However, during building stage, the concrete develops its viscosity and static yield strength with time due to reflocculation and hydration reaction. This assists the concrete mixtures to attain the contradictory rheological properties.

The buildability stage plays a vital role in determining the productivity of 3DCP. For fast building rates or high productivity, the printable mixture needs to exhibit sufficient buildability. Buildability is defined as the ability of the fresh concrete to sustain its weight and weight from the subsequent layers without failure until the required height is built. Failure in buildability can occur due to plastic collapse, elastic buckling, or a combination of both [4]. Plastic collapse is governed by the static yield strength of the material, whereas failure due to buckling is related to the mixture's stiffness. Nevertheless, static yield strength is directly related to stiffness, therefore, a mixture with sufficiently high static yield strength during printing would ensure good buildability. On the other hand, the setting time of concrete mixtures can also be related to the buildability such that the short setting time of the mixtures will lead to high buidaiblity. However, the setting time only represents two measurements of needle penetration resistance (initial setting and final setting time), which cannot be used to compare the failure of structures throughout the printing process. Moreover, the measurement of static yield strength and its growth with time can be used to ensure the stability of printed strucutres since the static yield strength should be higher than the self weight induced stresses on the bottom layers of printed structures.

The buildability enhancement in 3D concrete printing can be primarily categorized into two methods [4]:

1) Introducing the additives during initial mixing – The mixture design is adjusted during the initial mixing stage using additives that increase the thixotropic behaviour and/or setting rate of the fresh mixture. Some of the methods are reported in Refs. [5–8]. These methods are relatively simple and widely used in 3DCP, however, the buildability enhancement is restricted by pumpability limitations in these methods [4]. That means a higher dosage of such additives will cause difficulty in pumping and reduce the open time of the mixture, resulting in low printing duration per batch, thus, affecting productivity.
2) Interventions at the print head – In this technology, a printable mixture with a long open time is pumped to the print head and the setting rate is accelerated at the print head with specific interventions such as chemical, heat, ultrasonic waves, etc [9–14]. Amongst various approaches, the most developed and widely used approach in 3DCP is the chemical activation with the addition of set accelerators at the print head [4,15–18]. As the setting rate of the mixture is only accelerated at the print head (i.e., shortly before the extrusion), the pumpability of the mixture is not affected. Therefore, this enables enhancement in buildability without pumpability constraints.

Even though the chemical intervention using the set accelerators is a widely used method for buildability enhancement, this method is still in its early stages of development. Researchers are finding solutions to the issues associated with this method such as non-uniform mixing of accelerators during print head mixing, long-term durability concerns and the possible flash setting of concrete in the print head. Attaining uniform mixing of set accelerators at a short residence time is challenging and requires precise engineering of the print head design [10,15,19].

In recent years, On-demand control of rheology using magnetorheological (MR) fluids have been investigated as a novel technology in 3DCP. MR fluid is a combination of magneto-responsive materials suspended in a carrier fluid and it provides an on-demand control over the carrier fluid's rheological behaviour by applying a magnetic field of required strength [20]. Previous studies have investigated the application of MR fluid in the on-demand stiffening of cementitious mixtures [20–22]. In these studies, the cement paste (cement + water) was considered as a carrier fluid, whereas the responsive constituents were Carbonyl Iron Powder (CIP) and nano-Fe_3O_4 particles. According to these studies [20–25], the mechanism governing the on-demand stiffening of magneto-responsive paste (MRP) under the application of a magnetic field can be explained as follows.

During the mixing process, the responsive particles are randomly distributed within the carrier fluid (cement paste). The homogeneous dispersion of magneto-responsive particles can be achieved by mixing at an appropriate speed and duration. When a magnetic field of appropriate strength is applied, the responsive particle moves and forms clusters. These clusters are arranged in the direction of the magnetic field and restrict the fluid flow (tangling effect), thus, increasing the strength of the fresh MRP mixture. The size of clusters is dependent on the concentration of responsive particles in the carrier fluid. High concentration leads to a larger cluster formation which in turn increases the resistance to deformation (enhanced tangling effect), resulting in high static yield strength. During the magnetic field application, initially, the movement of responsive particles breaks the early-age C–S–H bridges, resulting in a reduction in the static yield strength for a certain period [23]. This period is known as percolation time [22]. This is followed by an increment in the static yield strength due to the tangling effect. The percolation time is an important factor that governs the print head design/magnetisation zone for 3D printing. Theoretically, the residence time of the mixture inside the print head (magnetisation time) should be higher than the percolation time.

Schutter et al. [26] studied the residual effect of magnetisation of cement paste containing magnetorheological additives. It was found that the storage modulus (proportional to static yield strength) of the mixture increases with the application of the magnetic field. Interestingly, once the magnetic field was removed the storage modulus remained unaffected. Therefore, it can be inferred that the mixture is only required to reside in the magnetisation zone for a short duration, equivalent to percolation time, to obtain a permanent increment in the static yield strength. This knowledge can be utilised to attain on-demand buildability enhancement of the printable mixtures, where the fresh magneto-responsive concrete (MRC) is magnetised for a pre-defined duration at the print head to attain accelerated static yield strength development after deposition.

Many studies have reported the possibility of actively controlling the rheology of cement pastes (MCP). However, its feasibility in concrete 3D printing application is yet to be investigated. In particular, the reported studies [20–24] have used cement paste as the carrier fluid. Therefore, the movement of CIP particles has minimal disruptions. However, the 3D printable mixtures usually consist of fine aggregates (sand) with particle sizes significantly larger than cement particles. The movement of CIP particles could be affected by the inclusion of sand. According to the authors' best knowledge, there are no known studies assessing the performance of CIP particles in 3D printable mixtures with sand.

This study utilises Carbonyl Iron Powder (CIP) as the responsive constituent in developing the MRC mixture for 3DCP applications. CIP particles are added during the initial mixing stage, eliminating the need for an in-line mixer at the print head. The MRC mixture after its preparation is pumped to the print head, where the mixture is exposed to a magnetic field for buildability enhancement as shown in Fig. 1. The buildability enhancement is dependent on factors such as CIP concentration, the residence time of the MRC mixture in the magnetisation zone (print head) etc. Moreover, the CIP concentration might also affect the pumpability and hardened properties of the MRC mixture. Therefore, this study conducts a comprehensive experimental analysis, where the CIP dosage that exhibits required pumpability, buildability and hardened properties is first identified. After that, the identified mixture design was used to print laboratory-scale specimens for validation.

2. Materials and methods

2.1. Cement, aggregates and admixtures

General-purpose cement complying with Australian standard - AS3972, sourced from Cement Australia was used as the binder, whereas, two types of graded silica sands – Coarse Sand (CS) and Fine Sand (FS) supplied by Holcim Australia were used as aggregates in developing printable MRC mixture. Polycarboxylic ether based superplasticiser (masterGlenium SKY 8379) and retarder (MasterSet RT 122) supplied by BASF were added in a precise dosage to enhance the pumpability of the MRC mixture. Carbonyl Iron Powder (CIP), supplied by the Chemical Store Inc., was used as the magneto-responsive additive in the MRC mixture for buildability enhancement. The specification of CIP provided by the supplier states that the median diameter of CIP particles was 5.2 μm with the 10% passing (D10) and 90% of passing (D90) of particles were 2.7 μm, and 9.2 μm respectively.

2.2. Magnets

Two permanent magnets made from Neodymium were used to generate the magnetic field required to stimulate the MRC mixture. The magnets, having a pulling force of 4 kN, were supplied by Factoryfast, Australia. The measured magnetic field on the surface was 0.1 T. Each magnet has a diameter of 100 mm and a thickness of 20 mm. These magnets were attached to a frame as shown in Fig. 2. The distance between the magnets can be adjusted using the frame corresponding to the required field strength. When the distance between the magnets is reduced, the magnetic field strength is increased. Therefore, the frame was designed to ensure the magnets were stable even at a low separating distance.

2.3. Preparation of printable MRC mixture

The mixture design of the carrier fluid was the same for all the mixtures. Only CIP dosage was varied amongst the mixtures studied as shown in Table 1. The dry ingredients (cement, aggregates and CIP) were weighed according to the mixture design and transferred to the Hobart mixer. After that, the dry mixing was performed for 1 min at a low speed (61 RPM). Meanwhile, the superplasticiser and retarder of the required amount were measured and dispersed in the mixing water. Once the dry mixing was complete, the wet ingredients (water, SP and retarder) were transferred to the mixing bowl and the mixing was continued for 1 min at low speed (61 RPM) followed by medium speed (124 RPM) mixing for 1 min. After 2 min of wet mixing, the mixer was stopped to scrape the unmixed material from the surface of the mixing bowl and blade to ensure homogenous mixing. It was then followed by 1 min of mixing at low speed.

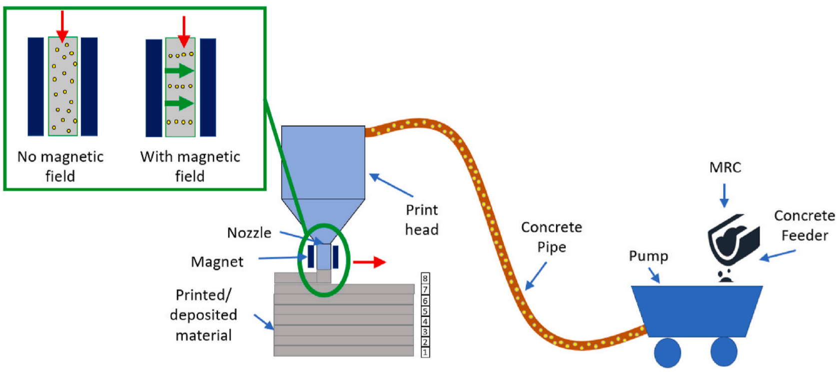

Fig. 1. Illustration of 3DCP system that utilises MRC mixture for buildability enhancement at the print head.

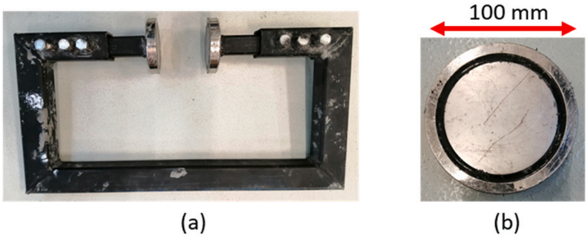

Fig. 2. (a) Magnet attached to a frame (b) magnet with dimensions.

Table 1
Mixture design of the MRC mixtures used for this study.

	Cement	CS	FS	CIP	SP[a]	RT[a]	Water
	in kg	in kg	in kg	in kg	in ml	in ml	in kg
Control	1	1	0.5	–	4	5	0.31
MRC_2.5	1	1	0.5	0.025	4	5	0.31
MRC_5	1	1	0.5	0.05	4	5	0.31
MRC_7.5	1	1	0.5	0.075	4	5	0.31
MRC_10	1	1	0.5	0.1	4	5	0.31

[a] SP – Superplasticiser, RT - Retarder.

3. Experimental program

In this study, the MRC mixtures were prepared with varying dosages of CIP to understand its effect on the mixture's printability and hardened properties. The printability of the MRC mixture was assessed by analysing its pumpability and buildability properties. The pumpability of the mixture was characterised by determining the variation in viscosity with shearing rate, whereas buildability was assessed by determining the evolution of static yield strength with time.

3.1. Apparent viscosity

The pumpability of cementitious materials is characterised by their viscosity [27,28]. The presence of CIP particles may affect the viscosity due to the nucleation effect or formation of CIP clusters. Therefore, the viscosity of the MRC mixture was determined using a rotational rheometer (Viskomat XL) with a 6-blades vane in a cup setup. Fresh MRC mixture after preparation was transferred to the rheometer cup and then sheared according to the protocol shown in Fig. 3. The shearing rate was increased in steps from 0 s^{-1} to 13 s^{-1} and then reduced to 0 s^{-1} in a total of 16 steps (8 steps for ramping up and 8 steps for ramping down). Each step was continued for 30 s to ensure the shear stress corresponding to the applied shearing rate reaches equilibrium. After the test was completed, the equilibrium shear stresses corresponding to the applied shearing rates was used to develop the flow curve. The descending section of the curve was utilised to determine viscosities at different shearing rates. It is important to note that the mixture was pre-sheared for 60 s at 13s^{-1} (maximum shearing capacity) to ensure a uniform starting point for all the mixtures under investigation.

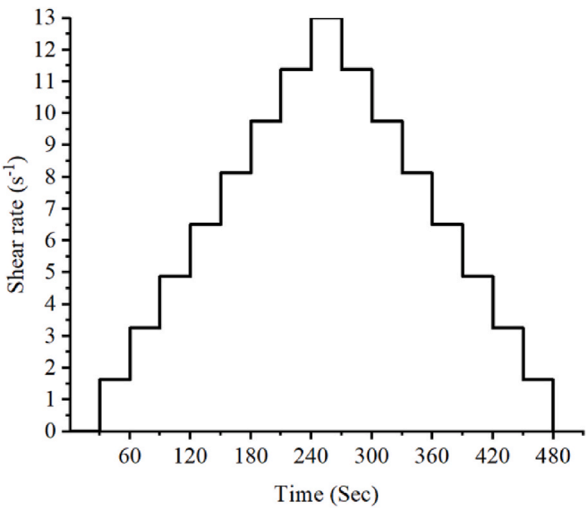

Fig. 3. Shearing protocol used to determine the viscosity of MRC mixture.

3.2. Evolution of static yield strength with time

To investigate the effect of CIP dosage on the buildability of the MRC mixture, the development of static yield strength with time was determined using a continuous penetrometer. The continuous penetrometer consists of a needle, a load cell and an actuator as shown in Fig. 4 (a). The needle with the dimensions shown in Fig. 4 (b) was connected to a 500 N load cell and was slowly penetrated inside the fresh mixture at a fixed rate of 10 mm/h. The resistance of penetration was continuously measured and converted into the static yield strength using the equation below [29].

$$Yield\ strength\ (\tau) = \frac{Penetration\ resistance\ (F)}{\pi * Cone\ radius\ (R)\sqrt{(R^2 + Cone\ height\ (H)^2}}$$

In this study, two buildability enhancement approaches using the MRC mixture and permanent magnets were investigated (see Fig. 5):

(1) Print-head activation technique: In this approach, the magnets were attached to the moving print head, providing design flexibility. Comparatively, the MRC mixture resides in the magnetisation zone (at the print head) for a significantly shorter period. Buildability enhancement attained using this approach will be due to the residual changes in the static yield strength of the MRC mixture from the short exposure to the magnetic field. This approach was simulated by exposing the MRC mixture to a magnetic field for a pre-defined duration before determining its static yield strength development rate. The duration of the exposure was limited to 60 s as the longer exposure time corresponds to larger print heads with magnets.

(2) Print-bed activation technique: This approach magnetises the MRC mixture after the deposition. This allows a significantly longer exposure time compared to the previous approach. The buildability failure is governed by the strength of the initial (bottom) layers [30]. Therefore, a long exposure of the first few printed layers to the magnetic field could result in a fast static yield strength development rate of the exposed layers which in turn enhances the buildability. However, this approach is limited by the design of the structure to be printed. For instance, while printing intricate structures, the magnets attached to the print bed can obstruct the subsequent print path. This approach was simulated by exposing the fresh MRC mixture to the magnetic field throughout the test duration while determining the static yield strength.

3.3. Compressive strength at 7 and 28 days

While the CIP particles accelerate the cement hydration by providing nucleation sites, high concentrations of these particles may negatively influence the strength development. In addition, the CIP particle contains a layer of carbon on the external surface. Although carbon is chemically inert towards hydration, carbonaceous fine particles at the right dosage have enhanced concrete's hardened properties and durability [31,32]. Meanwhile, excessive incorporation of carbonaceous components has shown a detrimental effect on the hardened properties [33]. Therefore, MRC mixtures with varying CIP dosages were cast into 50 × 50 × 50 mm^3 cubes to determine the compressive strength at 7 and 28 days. It is important to note that the MRC mixtures were not exposed to the magnetic field for this study. Cubes after de-moulding were cured in a water bath at 23 ± 0.5 °C until the test date. The compressive strength was determined by applying the load at a rate of 0.33 kN/s until failure. Three specimens were tested for each mixture design to check the repeatability.

3.4. Volume of permeable voids

The volume of permeable voids (VPV) was determined according to AS 1012.21. For this test, 50 × 50 × 50 mm^3 cubes with different CIP dosages were used after 28 days of the curing period. The cube samples were first oven dried for 24 h at 105 ± 5 °C and the dry weight was measured as W_1 in g. The samples were then submerged in the boiling water for 5.5 h and the weight of the boiled specimen was measured as W_2. Following the boiling process, the suspended weight of the samples was measured as W_3. Three cubes

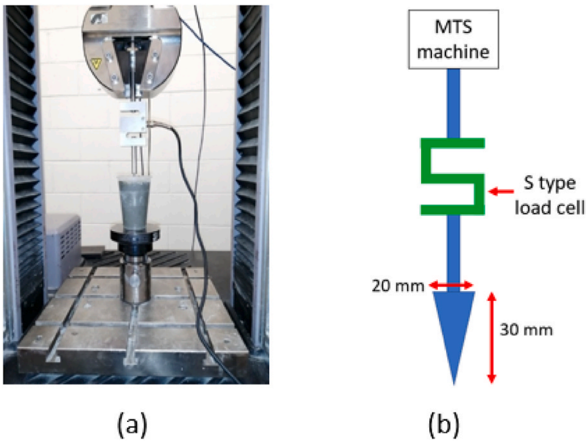

Fig. 4. (a) Penetrometer used to measure the evolution of static yield strength with time. (b) Dimensions of the penetrating needle used in this study.

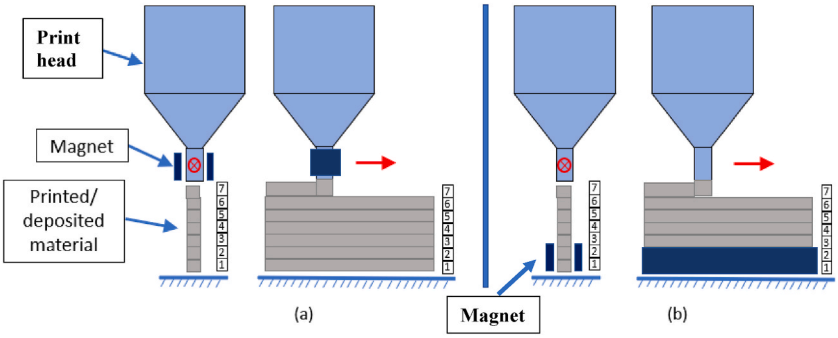

Fig. 5. Schematic representation of approach (a) print-head activation and (b) print-bed activation.

were tested for each CIP dosage and all weights were measured using an electronic balance with an accuracy of 0.01 g. The VPV of the mixtures were determined as per Equation (1).

$$VPV\ (\%) = \frac{W_2 - W_1}{W_2 - W_3}\ x\ 100 \qquad \text{Equation 1}$$

4. Results and discussions

4.1. CIP dosage on the pumpability of MRC mixture

The pumpability of a mixture is governed by its viscosity. The mixture with high viscosity requires high pumping pressure which in turn reduces the pumpability of the mixture. Fig. 6 shows the effect of various CIP dosages ranging from 0 to 10% by weight on the viscosity of the MRC mixture. It was found that up to a CIP dosage of 5%, the viscosity at low shear rates ($<5s^{-1}$) slightly reduced with the increment in CIP dosage. However, further increments in the CIP dosage slightly increased the viscosity at low shear rates. For instance, the viscosity of MRC mixture at a shear rate of 1.6 s^{-1} with CIP dosage of 0% (control), 2.5%, 5 %, 7.5% and 10% of cement were 62.1 Pa s, 55.6 Pa s, 50.6 Pa s, 65.6 Pa s and 79.1 Pa s respectively. It is known that the addition of CIP particles increases the total solid content, reducing the interparticle distance and the effective free water for lubrication. This increases the viscosity of the mixture [23]. On the other hand, the round shape of the CIP particle reduces the viscosity of the mixture due to the ball-bearing effect. Most likely, at low CIP dosages (up to 5% of cement) the ball-bearing effect of the round-shaped CIP particles supersedes the contradictory effect of increased solid content in the mixture. Meanwhile, at higher dosages (>5% of cement), the fine CIP particles could agglomerate due to their magnetic properties, providing resistance to the flow of concrete. The agglomeration of CIP particles in addition to the reduced free water content for lubrication significantly increases the viscosity of the mixture. Therefore, for CIP dosages above 5% of cement, the viscosity increases with the increment in the dosage. Moreover, the ball-bearing effect of the CIP particles is governed by their homogenous dispersion in the mixture. At higher dosages, the CIP particles are most probably not well dispersed due to agglomeration.

Meanwhile, previous studies reported that the superplasticiser helps the CIP particles to disperse, restricting the formation of agglomerates [34]. A similar effect can be seen in Fig. 6. Even though fine CIP particles have the tendency to agglomerate due to their

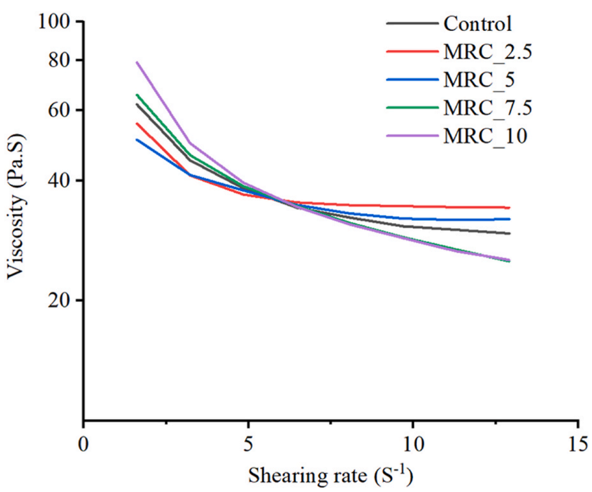

Fig. 6. Viscosity of the MRC mixtures at various shearing rates.

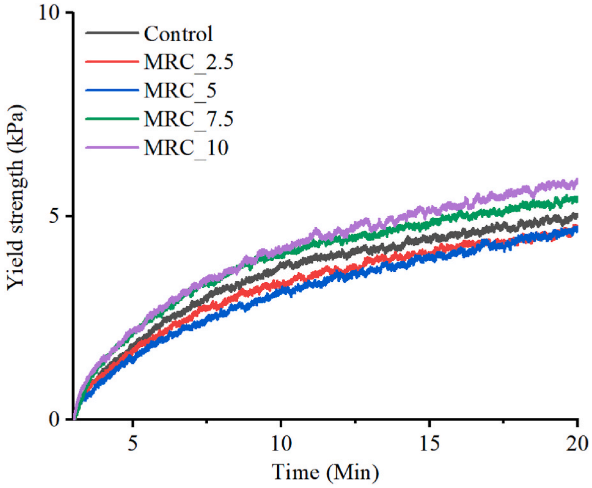

Fig. 7. Evolution of static yield strength with time of the MRC mixtures studied.

magnetic properties, the superplasticizer was effective in dispersing the CIP particles in the MRC mixture until a dosage of 5%. This can be identified by the reduction in viscosity of the MRC mixture with the increment in CIP dosage up to 5%. Interestingly, the superplasticizer was also found to be effective for CIP dosages above 5% at a higher shearing rate. While an increment in CIP dosages from 5% to 10% increased the viscosity at low shear rates, it reduced the mixture's viscosity at higher shear rates. For instance, the MRC mixture with a CIP dosage of 10% of cement exhibited a viscosity of 79.1 Pa s at a shear rate of 1.6 s^{-1}, whereas, with the increment in shear rate up to 12.9 s^{-1}, the viscosity of the mixture was determined as 25.3 Pa s (i.e., a reduction of 68%). In the case of MRC mixture with CIP dosage of 2.5%, the viscosity determined at 1.6 s^{-1} shear rate was 55.6 Pa s, whereas, at a shear rate of 12.9 s^{-1}, the viscosity of the mixture was determined as 34.2 Pa s (i.e., a reduction of 38%). This is most likely due to the combined effect of superplasticiser and high shearing rate destroying the CIP agglomerates at high dosages and dispersing the particles in the mixture. This enables the ball-bearing effect of round CIP particles at high dosages, resulting in low viscosity of the mixture. Therefore, it can be concluded that the buildability enhancement by the addition of CIP particles is not constrained by pumpability limitations, which is advantageous for the current study.

It is worth mentioning here that the viscosity of the MRC mixtures can be further reduced with the application of an alternating electromagnetic field [35,36]. It is reported that the application of an alternating electromagnetic field vibrates the magnetic particles, resulting in a vigorous micro-agitation of the MRC mixture which in turn reduces the viscosity and static /dynamic yield strength of the mixture [35,36]. Therefore, the addition of CIP particles to the cementitious mixture enables active control on the pumpability of the mixture which is essential to avoid blockages in the case of intermediate stoppages during 3D printing or long pipe pumping [35]. If required, the MRC mixtures can be exposed to electromagnetic pulses at strategized intervals to enhance pumpability by reducing the pressure required to pump the mixture at a steady state (propagation) or while initiating the pumping from intermediate stoppages [35]. However, the application of alternating electromagnetic fields for pumpability improvement is still in the early stage of development. The implementation of this technology requires further investigation, which is not in the scope of this study.

4.2. *CIP dosage on the buildability of MRC mixture*

4.2.1. *Without magnetic field*

The buildability of the MRC mixture was investigated by determining the evolution of static yield strength with time. In this section, the effect of CIP dosage on the static yield strength development was investigated without the presence of a magnetic field to assess the ability of CIP particles to provide filler effect, nucleation effect etc. Fig. 7 shows the static yield strength development of MRC mixtures determined using the slow penetrometer test. It was found that the addition of CIP particles showed a negligible effect on the static yield strength development with time. The MRC mixture with 0% and 10% dosage of CIP particles showed a similar static yield strength of approximately 5 kPa at 20 min respectively. Past studies have reported that CIP particles provide nucleation sites for C–S–H formation, thus increasing the static yield strength of the mixture [37,38]. Moreover, as the CIP particles fill the interparticle voids, the initial static yield strength was also found to be higher compared to the mixture without CIP particles [23,37]. It is important to note that in these studies the mixture did not contain superplasticizers and retarders (admixtures), which are required for 3D printable mixtures. Fig. 7 suggests that the dispersion and retarding effect provided by the admixtures lead to insignificant changes in the static yield strength of MRC mixtures. Therefore, the CIP particles alone cannot significantly enhance the static yield strength of printable MRC mixtures investigated in this study.

Meanwhile, the static yield strength of a mixture also relates to the pumpability of the mixture as it characterises the flow initiation of concrete during the pumping [39,40]. In large-scale 3D printers, the mixture is generally pumped in two stages, i.e., from the mixing chamber to an intermediate hopper followed by the hopper to the print head. This is termed as a separated pumping system by Xiao et al. [41]. In such pumping systems, the hopper is attached to the print head to provide an extra degree of freedom while printing. For instance, while printing an intricate structure, the extrusion rate needs to change frequently and precisely. In cases where concrete mixing occurs far from the print head, it is difficult to achieve such frequent and precise changes in the extrusion rate. The intermediate hopper assists in precise flow rate with frequent changes in extrusion rate. The fresh mixture usually resides in the hopper for a certain period before getting pumped to the nozzle. Therefore, an energy proportional to the static yield strength is required for the flow initiation. The static yield strength and viscosity properties of MRC mixtures imply that CIP dosage is not limited by both aspects of pumpability (flow initiation and propagation) in the absence of a magnetic field.

4.2.2. With magnetic field

This section describes the effect of CIP on the static yield strength development of the MRC mixture when exposed to the magnetic field. In this section, two types of buildability enhancement approaches are discussed. In the first approach, the deposited filaments are continuously exposed to the magnetic field to assess the effect of print-bed magnetisation on buildability enhancement. Here, the buildability enhancement is determined by measuring the evolution of static yield strength with time for the MRC mixtures exposed to the magnetic field throughout the testing duration. The results are shown in Fig. 8. It was found that the static yield strength development rate increased with the increment in CIP dosage. The reason for the high static yield strength at high CIP dosage is the formation of clusters during magnetisation that eventually restricts deformation. When the MRC mixture is exposed to a magnetic field, the CIP particles orient themselves to form clusters or chain-like structures in the direction of the magnetic field. These clusters increase the friction and restrict the flow when stress is applied, thus measuring high static yield strength [23]. With the increment in CIP dosage, the particles contributing to the cluster formation increase. This results in the enlargement of cluster (increase in the size of the cluster) followed by an increment in the cluster's density (closer packing of CIP particles in a cluster) [23].

When the CIP dosage was increased from 2.5% to 5% of cement, the static yield strength at 20 min of magnetisation increased from 7.9 kPa to 12.1 kPa, equivalent to a ~50% increment. On the other hand, when the CIP dosage was increased from 5% to 10% of cement, the static yield strength at 20 min of magnetisation was only increased by 17%. Therefore, it can be inferred that when the CIP dosage was increased from 2.5% to 5% of cement, the increment in static yield strength was a combined effect of cluster enlargement and its densification, resulting in a 50% increment in the static yield strength. However, further addition of CIP particles was majorly used to densify the cluster formed under magnetisation, limiting the static yield strength development rate. It is worth mentioning here that the acceleration in the static yield strength development at a high CIP dosage of cement can also be associated with the nucleation sites provided by excess CIP particles for the formation of C–S–H [23]. However, the static yield strength results shown in Fig. 7 confirm the contribution of CIP particles as nucleation sites towards static yield strength development are insignificant. Therefore, the changes in static yield strength development of the MRC mixture with the increment in CIP dosage are primarily governed by cluster enlargement and its densification under a magnetic field. Moreover, the influence of cluster enlargement on static yield strength development could be higher than the densification of the cluster.

In the second approach, the magnetic field was applied at the print head to assess the print-head magnetisation on the buildability enhancement. This was simulated by exposing the MRC mixture to the magnetic field for a pre-defined duration before measuring the development of static yield strength with time. The exposure time of the MRC mixture corresponds to the residence time of the mixture inside the print head. Therefore, the exposure time was limited to 60 s for the investigation. Moreover, a CIP dosage of 10% of cement was used in this study as it exhibited maximum static yield strength development in the previous section. Fig. 9 depicts the static yield strength development of MRC mixtures exposed to 20s and 60s as denoted by MRC_10_M_20s and MRC_10_M_60s respectively. The static yield strength development of the MRC mixture containing 10% of CIP particles with continuous magnetisation (i.e print-bed magnetisation) is also included to compare the different magnetisation strategies. As one could observe from Fig. 9, the print head magnetisation of the MRC mixture was found to be an ineffective strategy for buildability enhancement. The static yield strength of the MRC mixture after magnetisation was similar to the mixture without any magnetisation. Moreover, the increment in residence time up

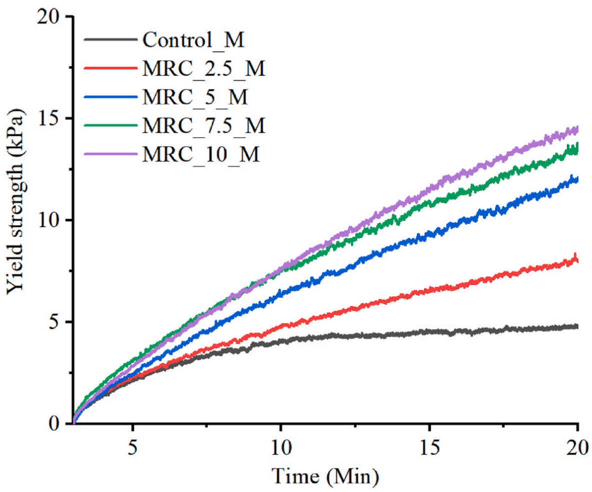

Fig. 8. Evolution of static yields strength with time of the MRC mixtures with print-bed magnetisation.

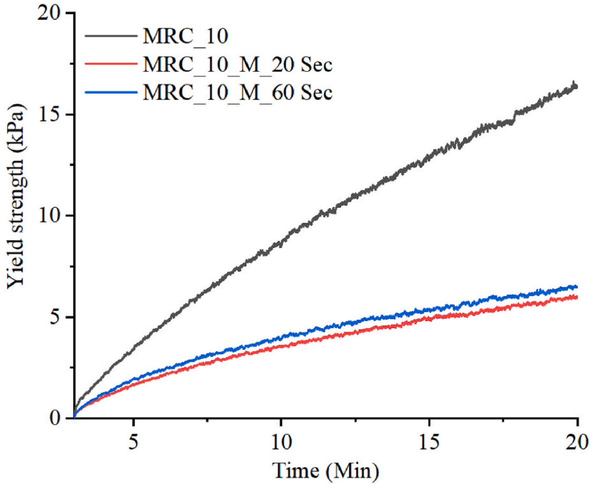

Fig. 9. Evolution of static yields strength with time of the MRC mixtures with print-head magnetisation.

to 60 s showed a negligible effect on the static yield strength development rate.

This could be due to the high percolation time in these MRC mixtures with the presence of superplasticiser such that the 60 s of residence time is insufficient to form the clusters of CIP particles. However, further investigation is required on the interactions between the superplasticiser and magneto-responsive CIP particles to understand the extended percolation time. According to Jiao et al. [34], the mixture with and without the superplasticiser showed a similar storage modulus (proportional to static yield strength) up to a magnetisation duration of 300 s. This implies the percolation time was more than 300 s for the mixtures studied by Jiao et al. [34]. For the MRC mixtures investigated in this study, the percolation time will vary due to different mixture design parameters, however, it is more than 60 s as shown in Fig. 9. This implies the design of a print head will be difficult as the residence time required is higher than 60 s. Perhaps the percolation time can be reduced with stronger magnets, such that the residence time of MRC mixture inside the print head can reduced. The authors' future study will investigate the factors reducing the percolation time of MRC mixtures to achieve a short residence time and compact print head designs.

Following the static yield strength assessment of MRC mixtures with two magnetisation strategies, 3D concrete printing of MRC mixtures was conducted with the layer dimensions of 200 mm (Length) x 50 mm (width) x 10 mm (thickness) until failure to validate the results obtained in Fig. 9. The printing was performed with the mixture containing a CIP dosage of 10% of cement. A pair of

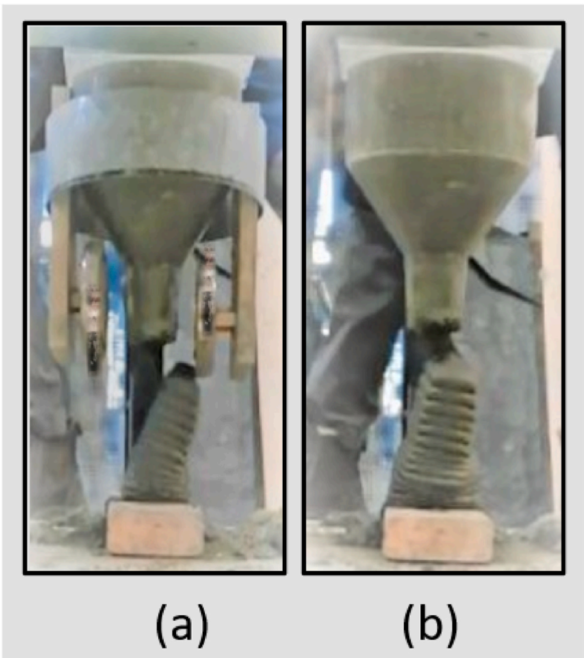

Fig. 10. Printing (a) with MRC mixture exposed to a magnetic field at the print and (b) without any magnetic field.

permanent magnets were attached to the print head using a metallic frame that ensures the stability of the fixture. Then the print head was attached to a computer controlled 3-axis gantry system for 3D printing. Meanwhile, a screw-type extruder was used to extrude the mixture through a circular nozzle of 30 mm diameter. The residence time of the mixture inside the magnetising zone was maintained for 10 s by adjusting the extrusion and printing rate. The printing rate used in this study is 10 mm/s. Fig. 10 shows the printed elements with and without print-head magnetisation for 10 s. It was found that the average failure height for MRC mixture both with and without magnetisation at the print head was 14 layers. This confirms that the print head magnetisation proposed in this study is an ineffective buildability enhancement strategy for 3DCP.

4.3. Mechanical and physical properties of MRC mixtures

The mechanical properties of MRC mixtures, measured by the compressive strength of cube specimens at 7 and 28 days, are shown in Fig. 11. As can be seen, the compressive strength of all MRC mixtures was higher than the control mixture at both ages (7 and 28 days). For instance, the control mixture exhibited 7- and 28-days strength of 50.7 MPa and 57.6 MPa respectively, whereas, with the addition of CIP particles at 5% of cement, the 7- and 28-days compressive strengths were increased by 16.9% and 16.4% respectively. The possible explanation for this is the increased particle packing density and filler effect by the CIP particles. In addition, the high toughness of the carbon layer on the CIP particles could also attribute to the improvement in the hardened properties of the mortar specimens [33,42]. However, within the MRC mixtures, the variation in compressive strength was insignificant for different CIP dosages. The mixture with a CIP dosage of 2.5% showed 7 days and 28 days strength of 56.7 MPa and 65.0 MPa respectively while increasing the CIP dosage from 2.5% to 10% showed a negligible increment of ∼0% and 1.5% at 7 and 28 days respectively. Among all studied mixtures, the MRC mixture containing 5% of CIP dosage showed the maximum compressive strength at both ages.

On the other hand, the volume of permeable voids (VPV) of MRC mixtures with varying CIP dosages was measured to understand the porosity and durability properties. Fig. 12 shows the void volume percentage of MRC mixtures prepared by conventional mould cast techniques and 3D printing techniques. Regardless of the preparation technique, the volume of permeable voids of the MRC mixtures is reduced with the addition of CIP particles. For mould cast specimens, the mixture without CIP particles showed a void volume of 10.01%, whereas the addition of CIP particles at a dosage of 2.5%, 5%, 7.5% and 10% reduced the void volume by 2.5%, 4%, 4.6% and 5.7% respectively. The influence of CIP particles on the reduction of VPV was high for printed samples. For instance, 3D printed specimens without CIP particles showed a void volume of 12.1%, whereas the corresponding 3D printed specimens at 2.5%, 5%, 7.5% and 10% of CIP dosage reduced the void volume by 2.3%, 7.5%, 9.5% and 13.8% respectively. The comparison of VPV results of printed and mould-cast specimens reveals that the porosity of printed specimens is higher than the mould-cast specimens despite the higher reduction compared to their control mixture counterparts. This could be due to the layer-wise fabrication method causing the increased porosity at the interlayer.

Fig. 13 compares the effect of CIP particles at various dosages on the compressive strength at 7 days and VPV of mould cast specimens. The compressive strength usually has an inverse relationship with VPV. More precisely, the specimens exhibiting lower VPV should have higher compressive strength than high VPV specimens. This is because the lower VPV implies higher packing density and densified matrix aids in the improvement of the strength properties. As can be seen from Fig. 13, up to a 5% of CIP content, the inverse relationship was witnessed as the CIP particles filled the voids, reducing the volume of permeable voids within the matrix. However, beyond 5%, the trend changed between the VPV and compressive strength. Even though, CIP particles at high dosages (>5%) filled the voids and reduced the VPV, the compressive strength determined was lower than the mixture with a CIP dosage of 5%. the possible reason to the reduced compressive strength at high CIP dosages is the excess carbon in the mixture brought by the addition of CIP particles at high dosages (>5%).

5. Conclusion

3D printable concrete mixtures incorporated with magnetorheological additives (MRC mixtures) were studied as an on-demand buildability enhancement technique for the emerging automated construction technology of 3D concrete printing. The on-demand buildability enhancement technique was tested for two magnetisation strategies of print head magnetisation and print-bed magnetisation by exposing the MRC mixture to the magnetic field at the printhead during printing and post-printing respectively. The following conclusions were drawn from this study.

1) The addition of CIP particles has an insignificant effect on the pumpability and buildability of the cementitious printable mixture in the absence of a magnetic field.
2) In the print-head magnetisation technique, the MRC mixtures exposed to 60s of residence time showed a negligible increment in static yield strength indicating that the buildability enhancement is insignificant. The MRC mixtures must be exposed for a long duration to provide a reasonable increment in the static yield strength.
3) With the continuous presence of a magnetic field (i.e., print bed magnetisation technique), the static yield strength was significantly increased for printable MRC mixtures compared to the control printable mixture. In addition, the static yield strength was increased with the increment in CIP dosage.
4) The 3D concrete printing of MRC mixtures using print-head magnetisation for 10s residence time reveals that the failure layers are the same for control and MRC mixtures.
5) The high dosage of CIP (above 5%) showed a slight reduction in the 7- and 28-days compressive strength of the MRC mixture despite the volume of permeable voids are reduced compared to their control mixture counterparts.

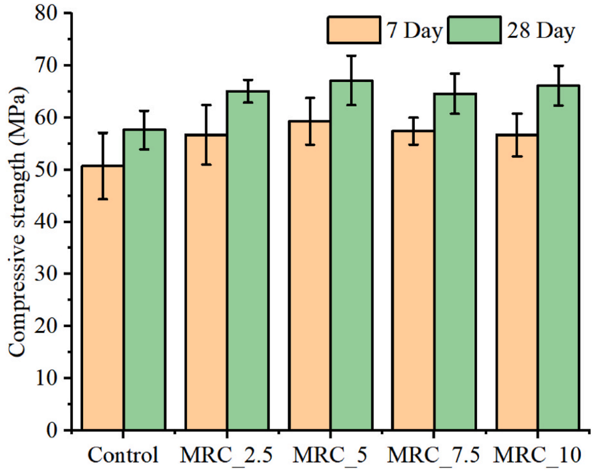

Fig. 11. 7- and 28-days compressive strength of MRC mixtures.

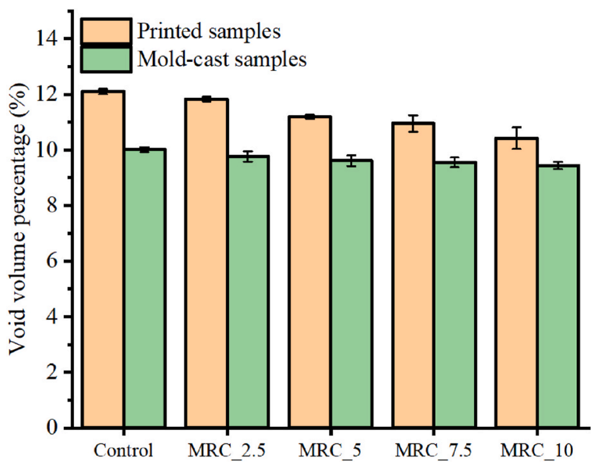

Fig. 12. VPV of MRC mixtures with various CIP dosages for printed and mould-cast samples.

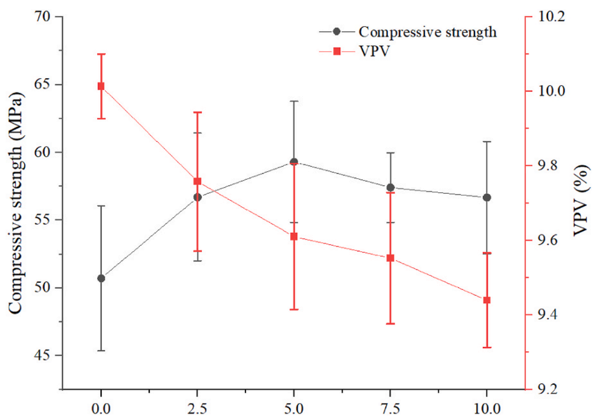

Fig. 13. 7-days compressive strength and VPV of MRC mixtures with various CIP dosages.

6. Future studies

This study suggests that to attain buildability enhancement in MRC mixtures, the print-bed magnetisation is crucial to ensure the MRC mixture is exposed to the magnetic field for a sufficiently long duration (longer than the percolation time). Even though this approach looks feasible, it limits the design flexibility of 3DCP (as discussed in 3.2). On the other hand, although the print-head magnetisation allows design flexibility, the buildability enhancement determined with this approach was insignificant. Therefore, our future study will be progressed in two directions:

1) Identifying the magnet with sufficient field strength and a suitable print head design to enhance the static yield strength of the MRC mixture using print-head magnetisation technique. The feasibility of electromagnets will be analysed. The electromagnets can produce a pulling force of more than 4 kN. In addition, new print heads will be developed that allow a longer residence time for the MRC mixture in the magnetisation zone without significantly increasing the size of the print head.
2) Understanding the relationship between CIP fine particles and the chemical admixtures in printable MRC mixtures. This study has been deemed important by Jiao et al. [34] as well since it would provide the reasons behind the extended percolation time due to the incorporation of chemical admixtures. With this knowledge, the printable MRC mixtures can be modified with compatible admixtures that provide a short percolation time of the mixtures without affecting the pumpability.

CRediT authorship contribution statement

Sasitharan Kanagasuntharam: Conceptualization, Methodology, Formal analysis, Investigation, Writing – original draft. Sayanthan Ramakrishnan: Conceptualization, Methodology, Writing – review & editing, Visualization, Supervision, Funding acquisition. Shravan Muthukrishnan: Conceptualization, Methodology, Formal analysis, Writing – review & editing. Jay Sanjayan:Conceptualization, Writing – review & editing, Supervision, Project administration, Resources, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgement

The Authors would like to thank the Australian Research Council (ARC) grants of DP210101680, LE170100168, and DE190100646 for the financial support on the research work reported in this paper.

References

[1] R.A. Buswell, et al., 3D printing using concrete extrusion: a roadmap for research, Cement Concr. Res. 112 (2018) 37–49.
[2] M.K. Mohan, et al., Extrusion-based concrete 3D printing from a material perspective: a state-of-the-art review, Cement Concr. Compos. 115 (2021), 103855.
[3] Y. Chen, et al., A review of printing strategies, sustainable cementitious materials and characterization methods in the context of extrusion-based 3D concrete printing, J. Build. Eng. 45 (2022), 103599.
[4] S. Muthukrishnan, S. Ramakrishnan, J. Sanjayan, Technologies for improving buildability in 3D concrete printing, Cement Concr. Compos. 122 (2021), 104144.
[5] S. Muthukrishnan, et al., Fresh properties of cementitious materials containing rice husk ash for construction 3D printing, J. Mater. Civ. Eng. 32 (8) (2020), 04020195.
[6] B. Panda, et al., Investigation of the properties of alkali-activated slag mixes involving the use of nanoclay and nucleation seeds for 3D printing, Compos. B Eng. 186 (2020), 107826.
[7] C. Liu, et al., Influence of HPMC and SF on buildability of 3D printing foam concrete: from water state and flocculation point of view, Compos. B Eng. 242 (2022), 110075.
[8] V. Saruhan, et al., Effect of fiber reinforcement on extrudability and buildability of mineral additive modified Portland cement mortars: a rheometer based simulation analysis, J. Build. Eng. 54 (2022), 104636.
[9] S. Muthukrishnan, S. Ramakrishnan, J. Sanjayan, Effect of microwave heating on interlayer bonding and buildability of geopolymer 3D concrete printing, Construct. Build. Mater. 265 (2020), 120786.
[10] Y. Tao, et al., Stiffening control of cement-based materials using accelerators in inline mixing processes: possibilities and challenges, Cement Concr. Compos. (2021), 103972.
[11] V. Vaitkevičius, E. Šerelis, V. Kerševičius, Effect of ultra-sonic activation on early hydration process in 3D concrete printing technology, Construct. Build. Mater. 169 (2018) 354–363.
[12] S. Muthukrishnan, S. Ramakrishnan, J. Sanjayan, Set on demand geopolymer using print head mixing for 3D concrete printing, Cement Concr. Compos. 128 (2022), 104451.
[13] S. Kanagasuntharam, S. Ramakrishnan, J. Sanjayan, Set-on demand concrete by activating encapsulated accelerator for 3D printing, in: RILEM International Conference on Concrete and Digital Fabrication, Springer, 2022.
[14] S. Muthukrishnan, S. Ramakrishnan, J. Sanjayan, Buildability of geopolymer concrete for 3D printing with microwave heating, in: RILEM International Conference on Concrete and Digital Fabrication, Springer, 2020.
[15] S. Muthukrishnan, S. Ramakrishnan, J. Sanjayan, Technologies for improving buildability in 3D concrete printing, Cement Concr. Compos. (2021), 104144.
[16] S. Bhattacherjee, M. Santhanam, Investigation on the effect of alkali-free aluminium sulfate based accelerator on the fresh properties of 3D printable concrete, Cement Concr. Compos. 130 (2022), 104521.
[17] F. Boscaro, et al., Eco-friendly, set-on-demand digital concrete, 3D Print. Addit. Manuf. 9 (1) (2022) 3–11.
[18] Y. Tao, et al., Using limestone powder as a carrier for the accelerator in extrusion-based 3D concrete printing, in: RILEM International Conference on Concrete and Digital Fabrication, Springer, 2022.

[19] G.Y.G.v.d. Meijde, Accelerating Early Strength and Stiffness Development of a Portland Cement-Based Mortar for 3D Printing, Eindhoven University of Technology, 2019.
[20] S.D. Nair, R.D. Ferron, Set-on-demand concrete, Cement Concr. Res. 57 (2014) 13–27.
[21] D. Jiao, et al., Quantitative assessment of the influence of external magnetic field on clustering of nano-Fe3O4 particles in cementitious paste, Cement Concr. Res. 142 (2021), 106345.
[22] D. Jiao, et al., Structural build-up of cementitious paste with nano-Fe3O4 under time-varying magnetic fields, Cement Concr. Res. 124 (2019), 105857.
[23] D. Jiao, et al., Structural evolution of cement paste with nano-Fe3O4 under magnetic field-Effect of concentration and particle size of nano-Fe3O4, Cement Concr. Compos. 120 (2021), 104036.
[24] C. Chibulu, et al., Active stiffening control by magnetically induced blocking in confined flow of fly ash pastes, Construct. Build. Mater. 313 (2021), 125485.
[25] D. Jiao, et al., Effect of solid volume fraction on the magnetorheological response of nassno-Fe 3 O 4 incorporated cementitious paste, J. Mater. Civ. Eng. 34 (11) (2022), 04022280.
[26] G. De Schutter, et al., Vision of 3D printing with concrete—technical, economic and environmental potentials, Cement Concr. Res. 112 (2018) 25–36.
[27] Mohan, M.K., et al., Rheological and pumping behaviour of 3D printable cementitious materials with varying aggregate content. Cement Concr. Res.. 139: p. 106258.
[28] D. Kaplan, F. de Larrard, T. Sedran, Design of concrete pumping circuit, ACI Mater. J. 102 (2) (2005) 110.
[29] D. Lootens, et al., Yield stress during setting of cement pastes from penetration tests, Cement Concr. Res. 39 (5) (2009) 401–408.
[30] R. Jayathilakage, P. Rajeev, J. Sanjayan, Yield stress criteria to assess the buildability of 3D concrete printing, Construct. Build. Mater. 240 (2020), 117989.
[31] L. Restuccia, G.A. Ferro, Promising low cost carbon-based materials to improve strength and toughness in cement composites, Construct. Build. Mater. 126 (2016) 1034–1043.
[32] S. Gupta, S. Muthukrishnan, H.W. Kua, Comparing influence of inert biochar and silica rich biochar on cement mortar–Hydration kinetics and durability under chloride and sulfate environment, Construct. Build. Mater. (2020), 121142.
[33] S. Muthukrishnan, S. Gupta, H.W. Kua, Application of rice husk biochar and thermally treated low silica rice husk ash to improve physical properties of cement mortar, Theor. Appl. Fract. Mech. 104 (2019), 102376.
[34] D. Jiao, et al., Rheological behavior of cement paste with nano-Fe3O4 under magnetic field: magneto-rheological responses and conceptual calculations, Cement Concr. Compos. 120 (2021), 104035.
[35] G. De Schutter, et al., Introduction to the Concept of Active Rheology Control in Case of Pumping of Cementitious Materials, RheoCon2 & SCC9, 2019.
[36] I. Dreßler, et al., Effect of electromagnetic pulsation on the rheological properties of mortars during pumping, in: Rheology and Processing of Construction Materials, Springer, 2019, pp. 294–302.
[37] C. Krezinski, et al., Magnetorheological cements for plug setting control in oil and gas wells, J. Petrol. Sci. Eng. 200 (2021), 108257.
[38] T. Oey, et al., The filler effect: the influence of filler content and surface area on cementitious reaction rates, J. Am. Ceram. Soc. 96 (6) (2013) 1978–1990.
[39] S. Muthukrishnan, S. Ramakrishnan, J. Sanjayan, Effect of alkali reactions on the rheology of one-part 3D printable geopolymer concrete, Cement Concr. Compos. 116 (2021), 103899.
[40] B. Panda, C. Unluer, M.J. Tan, Investigation of the rheology and strength of geopolymer mixtures for extrusion-based 3D printing, Cement Concr. Compos. 94 (2018) 307–314.
[41] J. Xiao, et al., Large-scale 3D printing concrete technology: current status and future opportunities, Cement Concr. Compos. (2021), 104115.
[42] S. Gupta, H.W. Kua, Carbonaceous micro-filler for cement: effect of particle size and dosage of biochar on fresh and hardened properties of cement mortar, Sci. Total Environ. 662 (2019) 952–962.