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Effect of Volume Fraction of Fine Sand on Magnetorheological Response and Blocking Mechanisms of Cementitious Mixtures Containing Fe_3O_4 Nanoparticles

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Abstract: Active rheology control (ARC) or active stiffening control (ASC) is a concept with which the conflicting rheological requirements during different stages of concrete casting can be reconciled. For instance, formwork leakage could be reduced by actively controlling structuration at the formwork joints, without having the negative impact of increased structuration during pumping and form filling. Using the concepts of magnetorheology, an active control methodology was thus recently developed by the authors to study the control of formwork leakages under pressure. This was performed using a small-scale laboratory test setup, using cementitious pastes containing magnetisable particles. To upscale from paste to mortar, the effect of volume fraction of sand on the magnetorheological (MR) response and blocking mechanisms of mixtures containing Fe_3O_4 nanoparticles is thus investigated in the current study. The MR response is determined using storage modulus tests, and the impact of ASC for leakage reduction is investigated by measuring the flow rate. Experimental results show that increasing the sand volume beyond a threshold causes a reduction in mobility of the magnetic particles, and thus lowers the MR effect. Despite this reduction in the MR effect at high sand volume, the increased particle interactions induce clogging and filtration effects, drastically lowering the flow rate. Applying the ASC method refines the voids in the clog, thereby eliminating the filtration effect. It is concluded that ASC can be used on mortar, with the expectation that there would be a reduction in the magnetorheological effect with increasing volume of fine aggregates.

Keywords: active stiffening control; magnetic field; Fe_3O_4 nanoparticles; blocking; packing density; formwork leakage; flow rate

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1. Introduction

Active rheology control (ARC) or active stiffening control (ASC) is a developing concept that offers a solution to the conflicting rheological requirements during the different stages of the casting process [1–4]. During pumping, for example, the concrete should be fluid enough to facilitate the pumping process; however, once the concrete is in the formwork, high fluidity gives rise to increased formwork pressure and formwork leakage. The ability to adjust the fresh concrete properties to match the desired behaviour could thus be of advantage. The concrete can be fluid while flowing in the pumping pipes, and then actively stiffened while in the formwork to prevent formwork leakage. ARC or ASC can be achieved by applying an external signal to the concrete that triggers a change in the material behaviour [3]. Based on this principle of ARC, an innovative active control methodology has recently been proposed in which a magnetic field is applied to the cementitious material to invoke a change in the rheological behaviour due to the presence of 'active' ingredients in the mixture. The active ingredients used were magnetisable particles in the form of Fe_3O_4 nanoparticles [5–8] and fly ash [9,10]. Although magnetorheological control is a fairly new concept for cementitious mediums, this is a technology already widely used in other industrial applications such as brakes, hydraulic valves, and shock absorbers [11].

The interest in magnetorheological (MR) fluids for industrial applications arises from the rapid and reversible field-dependent rheological changes in the fluid [12]. When a magnetic field is applied, the magnetic particles acquire dipole moments and align with the magnetic field to form chains or clusters, giving the fluid an additional yield stress component. To initiate flow, these chains or clusters must be deformed or broken.

Building out from the principles of MR fluids, a magnetic-field-based active stiffening control methodology was developed to study the efficacy of ASC for formwork leakages of flowable pastes under high pressure. This was performed using a small-scale laboratory test setup in which a pressurised column with a narrow opening at the bottom was filled with cementitious material to simulate formwork leakage under pressure [9,13]. Using the ASC methodology, reductions in both the flow rate and net mass of leaked material were observed for pressures lower than 30 kPa. This was attributed to the formation of magnetic clusters due to the application of a magnetic field across the outflow width. Although the formation of clusters was less effective at higher pressures, the study showed that clusters formed under a pressure of 10 kPa were able to withstand pressures of up to 100 kPa [9]. Following these promising results obtained using cementitious pastes, the current study is aimed at studying the effects of fine aggregates on the ASC methodology, in a first step to upscaling to mortar phase.

Mortar can be considered as a concentrated suspension, with rigid particles of different sizes suspended in the bulk fluid. In such a system, there are strong interactions between particles due to their close proximity. The concentration, shape, size distribution, and surface properties of the particles all affect the strength of these interactions. In general, there is a net attraction between particles which leads to flocculation [14]. Convergence of such a fluid towards a bottleneck, such as in the case of formwork leakage through a gap, is likely to cause blockage of flow. In general, clogging of granular material is a complex phenomenon that is dependent on the outlet size and type, the physical properties of the particles, and the particle packing [15]. In the case of mortar, however, the presence of the cementitious pastes in which the grains are embedded also influences the contact network between particles [16]. The more paste available, the lower the frequency of collisions between coarser particles, and thus the lower the particle interlocking effect [17]. Additionally, due to the heterogenous nature of cementitious mortars, this blocking effect can induce filtration of the liquid phase through the granular skeleton, as observed in extrusion flows, for example [18], leading to a local change in properties at the flow confinement.

The presence of aggregates clearly has an influence on the nature of the flow, particularly when flowing through a small gap. It can also be expected that the presence of aggregates would influence the magnetorheological response of the fluid, since the magnetically induced particle interactions are strongly dependent on the distance between the magnetic particles within the matrix of the fluid [19]. As such, the current study investigates the effect of sand volume fraction on the magnetorheological response and active stiffening control methodology previously introduced by the authors.

2. Experimental Programme

2.1. Materials

The basic materials used in this study include Portland cement (CEM 52.5 N), spherical Fe_3O_4 nanoparticles (98% purity), a polycarboxylate ether superplasticiser (PCE), a cellulose-based viscosity-modifying admixture (VMA), and fine sand. The cement had a median particle size of approximately 7.2 µm and specific gravity of 3.2. The Fe_3O_4 nanoparticles used in the study had a particle size of 100 nm and specific gravity of 4.95, as stated by the manufacturer. The sand was a very fine quartz sand with median particle size of 170 µm and specific gravity of 2.65. A very fine sand was selected for testing to remain within the limit of maximum particle sizes permissible to conduct rheology experiments within the confined geometry of the plate–plate rheometer. The gap between the shearing plates should be at least 5–10 times the maximum particle size in the mixture [20]. Table 1 shows the chemical composition and Figure 1 shows the particle size distribution of the cement powder and sand.

Table 1. Chemical composition of cement and sand (wt. %).

Element	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	K_2O	SO_3	Others
Cement	19.4	6.04	4.12	61.5	1.25	5.35	0.48	1.86
Sand	99.0	0.06	0.70	0.02	-	0.30	-	0.06

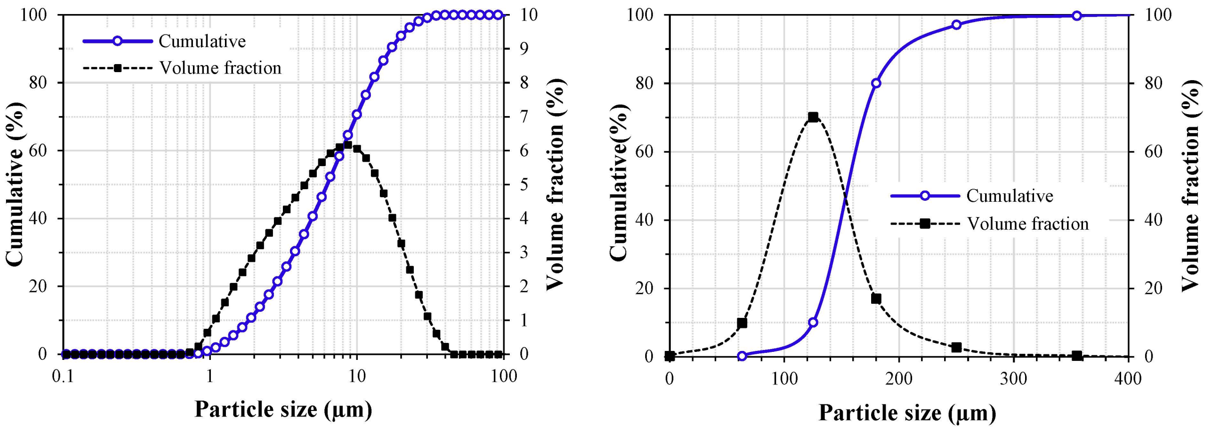

Figure 1. Particle size distribution of (**left**) cement powder; (**right**) sand.

The magnetic properties of the cement and magnetic nanoparticles (MNPs) were measured by a vibrating sample magnetometer (VSM) at 25 °C. The magnetisation curves of the materials are plotted in Figure 2, and the magnetic parameters are summarised in Table 2.

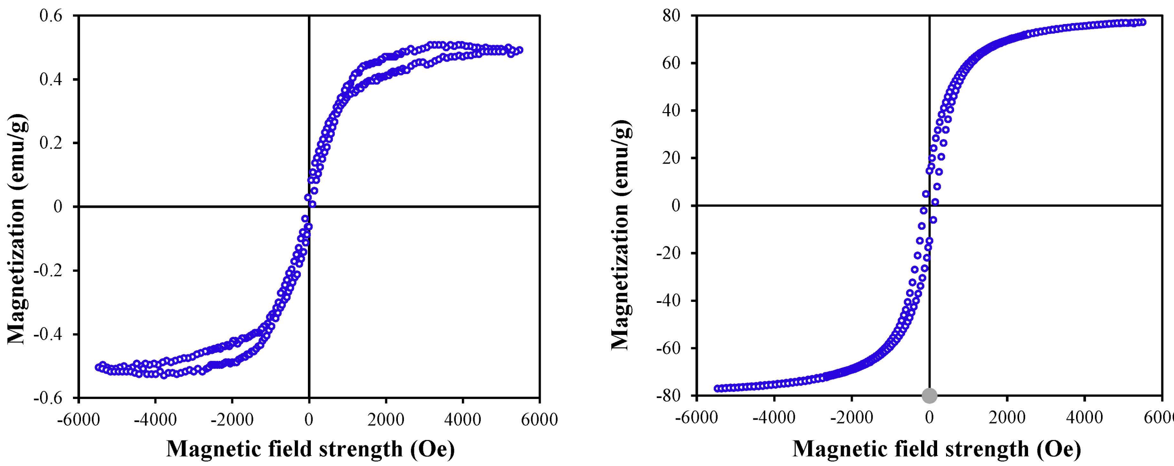

Figure 2. Magnetisation curves of (**left**) Portland cement, (**right**) MNPs.

Table 2. Magnetic properties of cement and Fe3O4 nanoparticles (MNP) used in the study.

Material	Saturation Magnetisation (M_s, emu/g)	Remnant Magnetisation (M_r, emu/g)	Coercive Field (H_c, Oe)
Cement	0.59	0.05	70.72
MNP	77.56	10.23	108.27

Five mixtures were prepared to study the effect of volume concentration of fine aggregates on the magnetorheological response and confined flow under magnetic field.

The water to cement ratio of the paste and the volume of MNPs used was kept constant. The quantity of superplasticiser, and in the instance of the reference paste the quantity of VMA, was adjusted to attain a mini slump spread diameter of 255 ± 5 mm. The sand amount was added to the paste in 10% volumetric replacements of the total volume of paste. Mixtures M0, M1, M2, M3, and M4 thus have 0%, 10%, 20%, 30%, and 40% total volume of sand. This is visually represented in Figure 3, and the mixture proportions of the pastes are shown in Table 3.

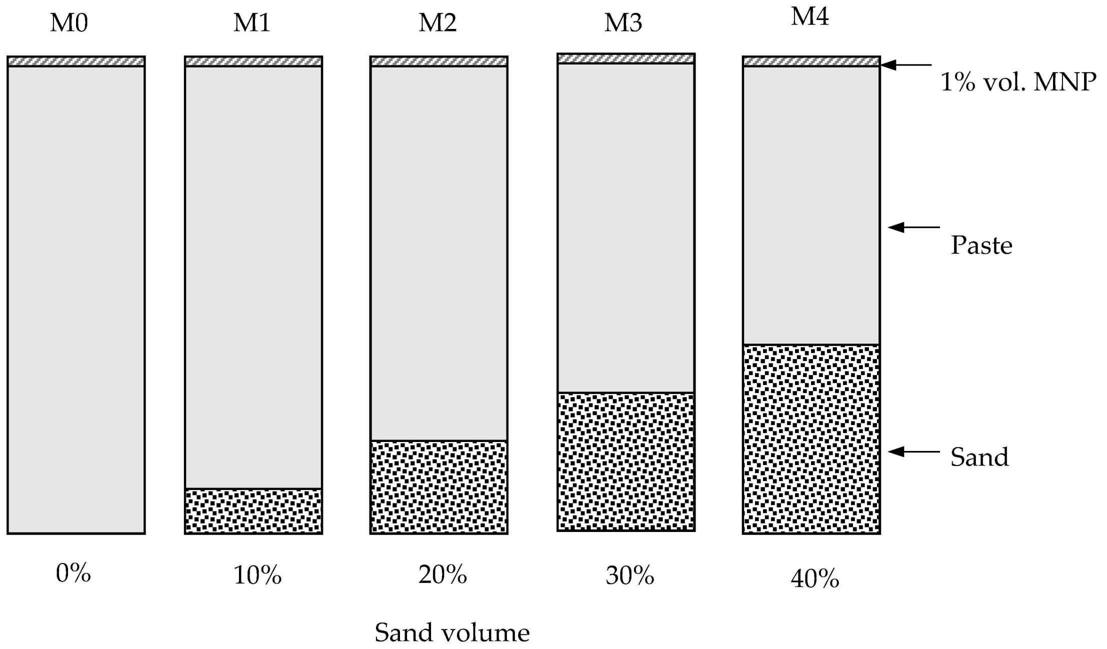

Figure 3. Schematic representation of mixture proportions.

Table 3. Mixture proportions and fresh properties.

Mix. No	W/C (-)	PCE (wt. % Cement)	VMA (wt. % Cement)	Sand (% Total Volume)	NP (% Total Volume)	Mini Slump (±5 mm)	Wet Density (g/cm^3)
M0	0.55	-	0.025	0	1	255	1.91
M1	0.55	0.05	0	10	1	255	2.01
M2	0.55	0.15	0	20	1	255	2.03
M3	0.55	0.4	0	30	1	255	2.13
M4	0.55	1.0	0	40	1	255	2.16

2.2. Methodology

2.2.1. Particle Packing Density

The packing density of solids in a granular system is a basic parameter that characterises the properties of the system. Since we can consider concrete or mortar as a granular system, the particle packing would thus also significantly influence the properties of these mixtures [21]. The packing density, minimum void ratio, and excess water ratio were determined using the centrifugal consolidation method [22,23]. The mixtures were placed in standard 50 mL centrifuge tubes and centrifuged at 3500 rpm for 10 min. When the test-tubes are centrifuged, the particles in the mixture are compacted, thereby reducing the amount of water needed to fill the voids in between the compacted particle matrix. After centrifuging, the excess amount of water thus forms a layer at the top of the compacted mixture. This excess water layer was removed using a pipette and measured, and the amount of water remaining in the compacted sample could be determined. The excess water ratio W_e could then be calculated according to Equation (1).

$$W_e = \frac{V_{w(excess)}}{V_s} \quad (1)$$

The packing density, φ_{max} was then calculated as shown in Equation (2):

$$\varphi_{max} = \frac{V_s}{V_s + V_w} \quad (2)$$

where V_s and V_w are the volume of total solids and the volume of the water remaining in the consolidated mass, respectively. The water remaining in the consolidated material was taken as the mass of the initial water in the mixture (mixing water), minus the excess water removed, assuming the particles did not absorb any water. The minimum void ratio V_{min} defined as the volumetric ratio of voids to solids can be determined from the packing density and is calculated as shown in Equation (3). Generally, the higher the packing density, the smaller the volume of voids needing to be filled [24].

$$V_{min} = (1 - \varphi_{max})/\varphi_{max} \quad (3)$$

2.2.2. Oscillatory Time Sweep Test

The oscillatory time sweep test was performed to evaluate the structural build-up with and without magnetic field application by using a rotational parallel-plate rheometer (Anton Paar MCR-102, Graz, Austria). Tests with magnetic field application were conducted with a magnetic field strength of approximately 0.65 T, to match the magnetic field applied in the pressure flow test. The magnetic field was applied at the start of the time sweep test. The strain amplitude and frequency during the time sweep tests were 0.001% (within the linear viscoelastic region) and 2 Hz, respectively. To determine the linear viscoelastic region (LVER), strain sweep tests were conducted without a magnetic field from 0.0001% to 10% and constant frequency of 2 Hz prior to the time sweep tests. The critical strain as shown in Figure 4 was approximately 0.004%, and thus a strain amplitude of 0.001% could be selected for the time sweep test.

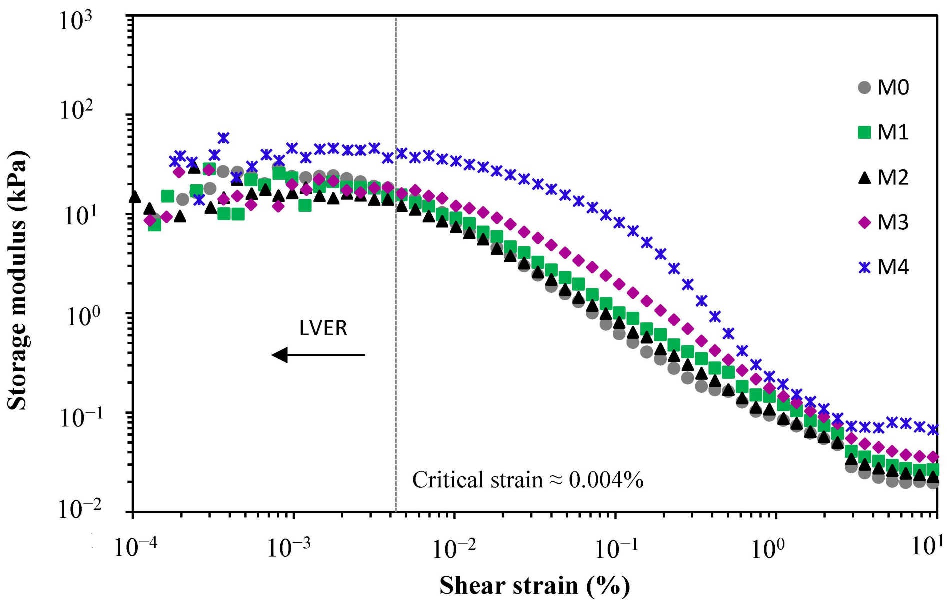

Figure 4. Strain sweep test results showing LVER of the mixtures.

2.2.3. Pressure-Driven Flow Test

A small-scale test setup that was previously developed [9,13] was used to study the flow rate of the mixtures through a confined geometry under the impact of applied pressure to imitate formwork leakage. The test setup includes a vertical pressurised cylinder with a flow outlet positioned at the bottom of the cylinder. The flow outlet used in this study had a rectangular cross-section, with dimensions of 2 mm × 20 mm, and height of approximately 7 mm. To apply a magnetic field, the bottom plate of the cylinder was replaced by a plate with similar geometry, but instead with static magnets (see Figure 5). This configuration creates a magnetic field across the flow width, similar to MR fluids in the valve mode [12,25]. This allows for the deposition of the magnetic particles at the walls, thereby pinching the flow width together and reducing the flow rate like an MR fluid in the magnetic gradient pinch mode [26].

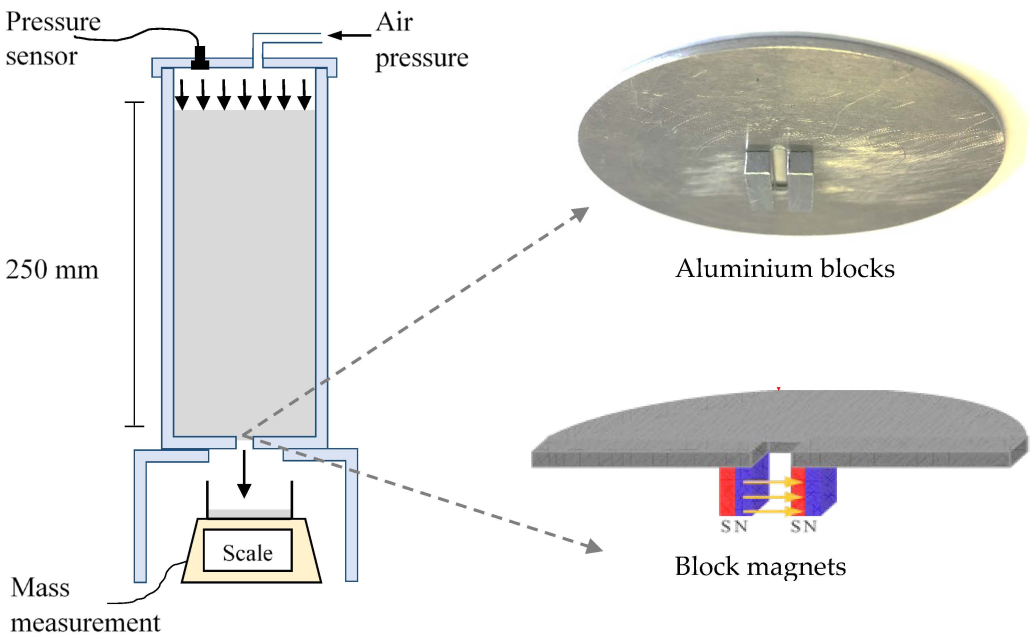

Figure 5. Test setup to study pressure-driven flow of cementitious paste [9].

Previous experiments showed a threshold pressure at approximately 30 kPa at which the active stiffening control was no longer sufficient to cause blockage of flow. The overhead pressure was thus kept constant at 30 kPa for each test and two tests were performed for each mixture to check repetition.

3. Results and Discussion

3.1. Particle Packing Density and Excess Water Ratio

The packing densities of the particles, which is defined as the ratio of the volume of solid particles to the bulk volume occupied by the particles [27], are given in the first column of Table 4 and plotted against the volume fraction of sand in Figure 6. As expected, the packing density, $\varphi_{max,}$ increases with increasing sand volume fraction. This is due to the occupying effect which follows when the volumetric fraction of coarser particles is less than the optimum value [27,28]. When the larger particles (sand grains) are added to smaller particles (cement and MNP), the larger particles occupy the space within the bulk and porous volume, thereby increasing the packing density. The minimum void ratio (V_{min}) and excess water ratio (W_e) were are also determined and are given in Table 4, and W_e is plotted against the volume fraction of sand in Figure 6.

Table 4. Particle packing and excess water ratio.

Sample	φ_{max}	V_{min}	W_e
M0	0.47	1.13	0.59
M1	0.54	0.85	0.46
M2	0.59	0.69	0.33
M3	0.64	0.57	0.22
M4	0.67	0.49	0.12

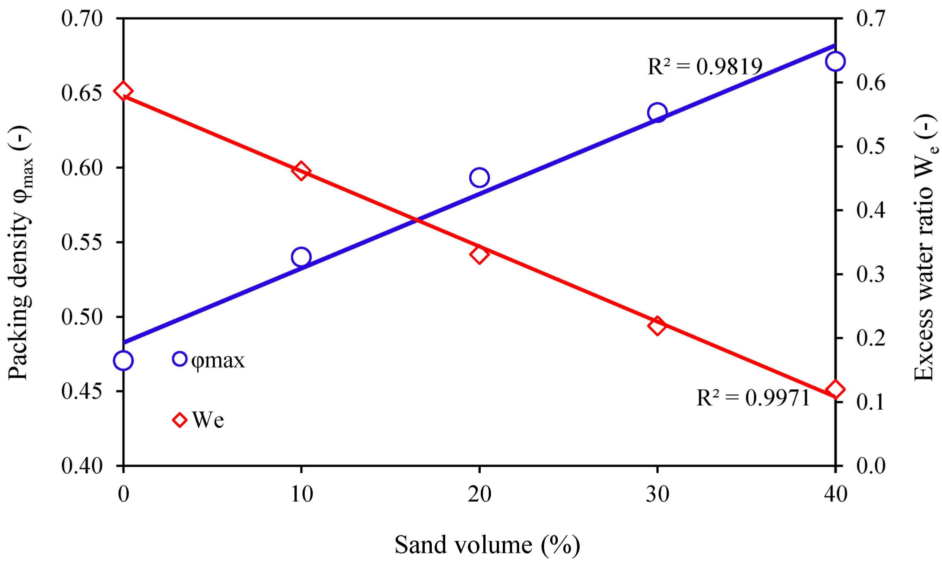

Figure 6. Packing density and excess water ratio as a function of sand volume fraction.

In a state of maximum packing density, the coarser particles are tightly packed together, with just enough fine particles available to fill the voids between the coarser particles. Since there are no excess fine particles separating the coarse particles from each other, the coarser particles would slide against each other, resulting in large interparticle interactions and interlocking. Conversely, in the state of maximum mass flow rate, the coarse particles are separated from each other by the excess fines since there are more than enough fines necessary to fill the voids. The fine particles act like ball bearings, lowering the particle interlocking action and the smooth movement of coarse particles within the fluid [17]. Extending this theory to cementitious pastes, the excess amount of water in the paste is responsible for dispersing the particles and lubricating the cement paste [27]. Increasing the sand volume in the mixture from 0 to 40% therefore moves the mixtures from a state of mass flow rate towards maximum packing density, which is evident by the increasing φ_{max} and decreasing W_e. This could arguably also lead to a reduction in mobility of the MNPs to form clusters due to the obstruction caused by the increasing presence of larger solid particles in the mixture as illustrated in Figure 7. The magnetorheological response of the mixtures was therefore investigated by measuring the storage modulus of the mixtures with and without magnetic field application and is discussed in the following section.

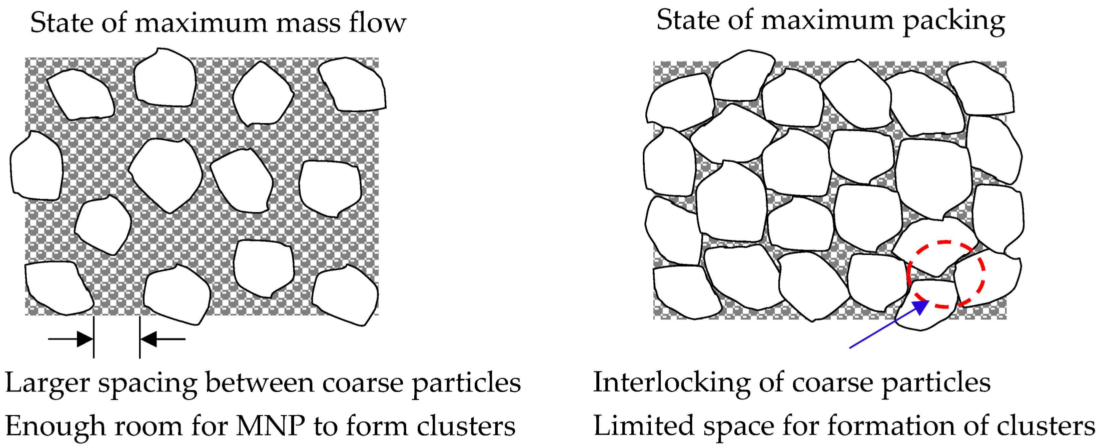

Figure 7. Effect of aggregate content (after [17]).

3.2. Storage Modulus

The effect of sand volume fraction on the magnetorheological response determined by measuring the storage modulus of the mixtures with and without magnetic field application is shown in Figures 8 and 9. In the absence of the external magnetic field, the mixtures all have a storage modulus in a similar range after 300 s (G'_{300s} < 150 kPa). When a magnetic field of 0.65 T is applied, the initial rate of increase in storage modulus (and hence the structural build-up) and final storage modulus after 300s of all the mixtures increases significantly to varying degrees. This is consistent with previous studies in which the storage modulus of cementitious pastes containing MNPs was measured with/out a magnetic field. The increase in storage modulus is due to the increased structuration caused by the formation of magnetic clusters within the paste. The magnitude of increase in the storage modulus under a magnetic field provides an indication of the degree of magneto responsiveness and ability to form clusters. Generally, a larger relative increase in storage modulus indicates a higher magneto response [5,7,8,29].

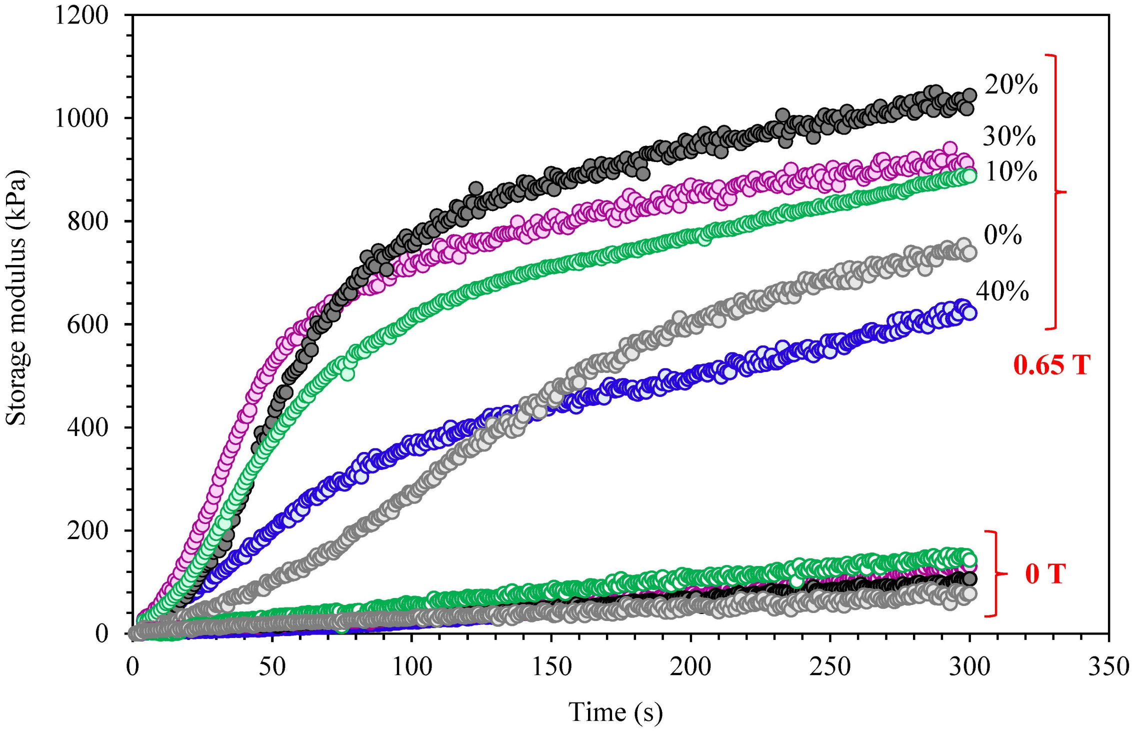

Figure 8. Storage modulus evolution with/out magnetic field application.

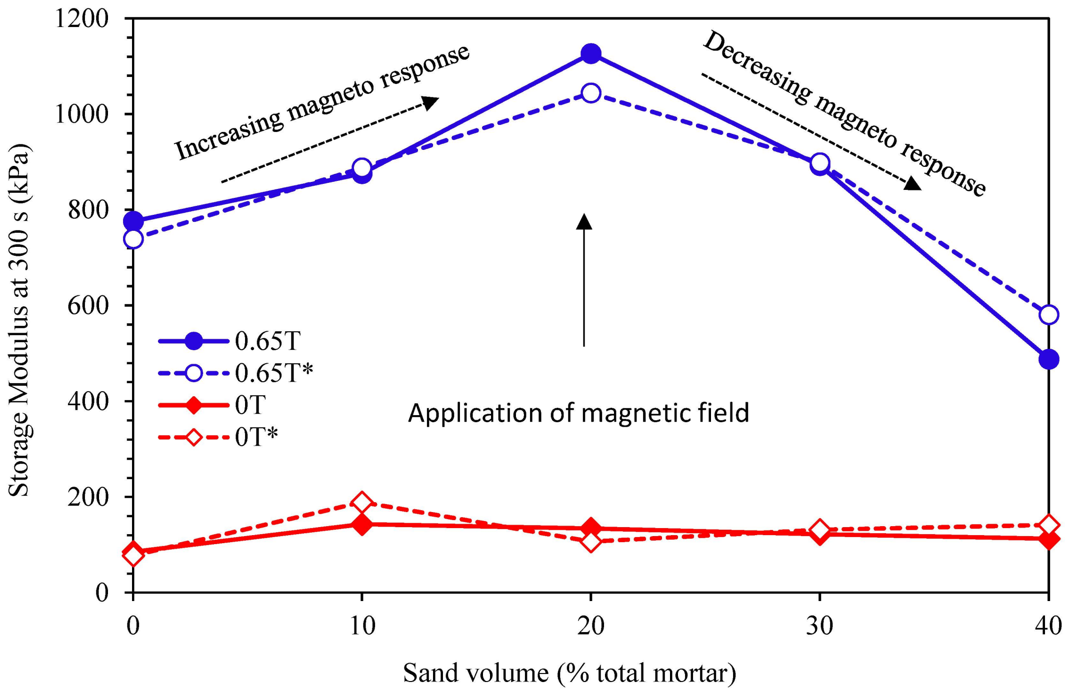

Figure 9. Effect of sand volume fraction on magneto response and increase in storage modulus at 300 s. * Repetition of test on a different batch.

Increasing the volume percentage of sand particles from 0 to 20% had the effect of increasing the magnetorheological response of the mixture, as shown in the storage modulus evolution in Figure 8 and final storage modulus after 300 s in Figure 9. It is hypothesised that in up to 20% volume of sand, the presence of sand grains in the mixture does not have a negative impact on the ability of the magnetic particles to form chains or clusters. Due to the low packing density, the sand grains are loosely packed within the cementitious paste, giving the magnetic particles more freedom to move and form clusters. As the percentage volume of sand is increased from 20 to 40%, it is theorised that the presence of the sand grains begins to inhibit the movement of the magnetic particles, thereby lowering the magnetorheological response. At 40% sand volume, the magnetorheological response is the lowest. The result thus suggests that there is a threshold volume percentage of fine aggregate that can be added to the paste after which the magnetorheological effects begin to decline due to the presence of aggregates. In practice, it is more desirable to formulate the mixture composition for maximum packing density to improve the strength and durability indexes of the concrete [30]. It can thus be hypothesised that for more practical mixture compositions containing fine aggregates of even larger sizes than used in this study, the magnetorheological response would not be as high as its paste counterparts. Nevertheless, despite the M4 having the lowest magneto response, the magnitude of increase in storage modulus is still noteworthy. The storage modulus increased by a factor of 4.2. Moreover, in comparison to the time sweep tests that are performed in nearly static conditions, formwork leakage flows occur in dynamic conditions, of which the particle concentration also plays a key role in the blocking mechanisms. The higher sand volume could have a positive effect on the blocking mechanisms despite the lower magneto response. This is discussed in the following section.

3.3. Flow Rate

The mass of material flowing out of the test cylinder was measured continuously during testing. The loss of mass from the test cylinder relative to the initial mass in the 250 mm height of fluid (M_{loss}) could thus be determined. The M_{loss} as a function of time under 30 kPa overhead pressure is plotted in Figure 10. Despite having different packing densities, the M0, M1, M2, and M3 mixtures exhibited similar flow behaviour, i.e., mass flow, with similar flow rates when no magnetic field was applied. The slope of the M_{loss} curve remained relatively constant until the end of the test when the test cylinder was almost completely emptied, except for the material in the dead zones. The M4 mixture, however, deviated from mass flow and exhibited significant filtration effects from the onset of flow. Only the liquid fraction was lost from the cylinder, and therefore the percentage mass loss was extremely low. However, the filtration process continued until most of the liquid fraction within the vicinity of the gap was lost.

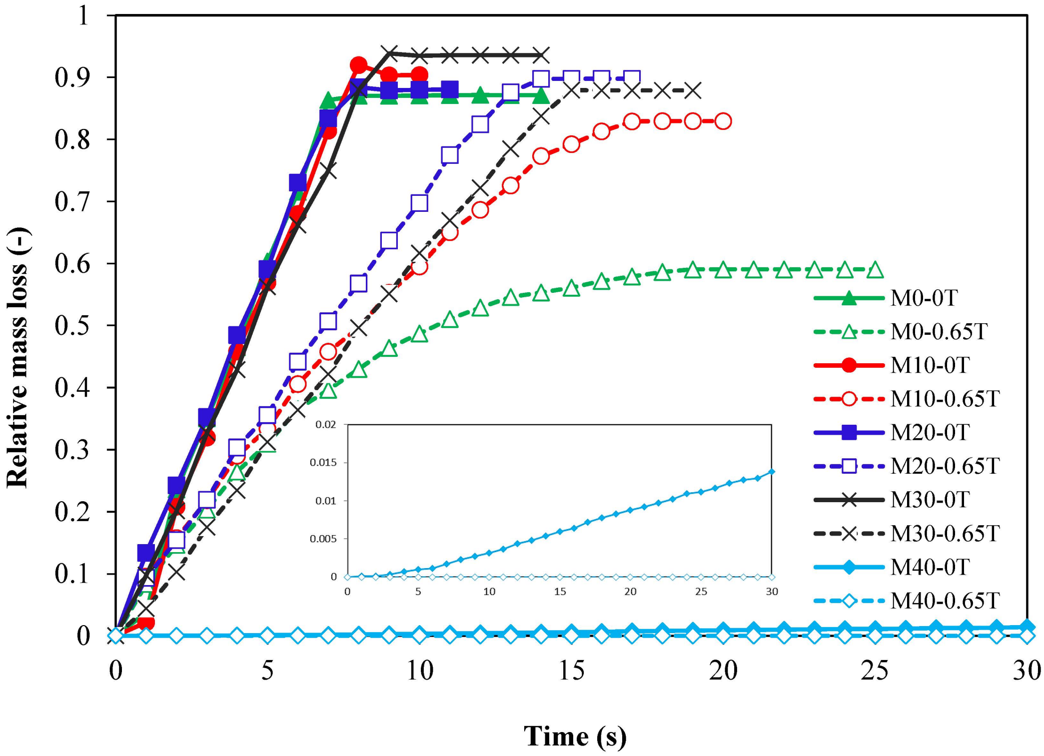

Figure 10. Relative mass loss as a function of time without ASC (solid lines) and with ASC (dashed lines).

On the contrary, when a magnetic field of 0.65 T was applied, the flow rates of all the mixtures decreased. The M1, M2, and M3 mixtures only show a reduction in slope, with no changes to the net mass of outflow at the end of the test. The M0 mixture, however, not only showed a reduction in the initial slope, but the slope continued to decrease until it reached a plateau, indicating that complete blockage occurred. This resulted in an approximately 40% reduction in mass loss. For the M40 mixture, the slope was effectively reduced to zero and the filtration effect was not detectable. The reduction in slope is due to the formation of magnetic clusters at the outflow region [9]. As the mixture flows through the magnetic field, the magnetisable particles are deposited onto the walls of the outflow channel that pinch the flow width inwards, like an MR fluid in the magnetic gradient pinch mode [26].

The mass flow rate $\dot{m}$ can be obtained by computing the slope of the M_{loss} curves. For comparative reasons, the mass flow rate was converted into a volumetric flow rate since the mixtures had different densities. The linear slope indicates a constant flow rate, with little to no blocking, and thus it was assumed that the density of the material flowing out of the cylinder remained unchanged. In the cases where blocking was observed, the flow rate was only computed during the initial flow stage when the slope of the M_{loss} curve was approximately constant [9]. In the case of filtration, since it was predominantly the liquid fraction that was lost from the sample, the density of material was assumed to be that of water. The computed flow rates with and without ASC are plotted in Figure 11 and show the effect of percentage sand volume on the magnetically induced flow reduction.

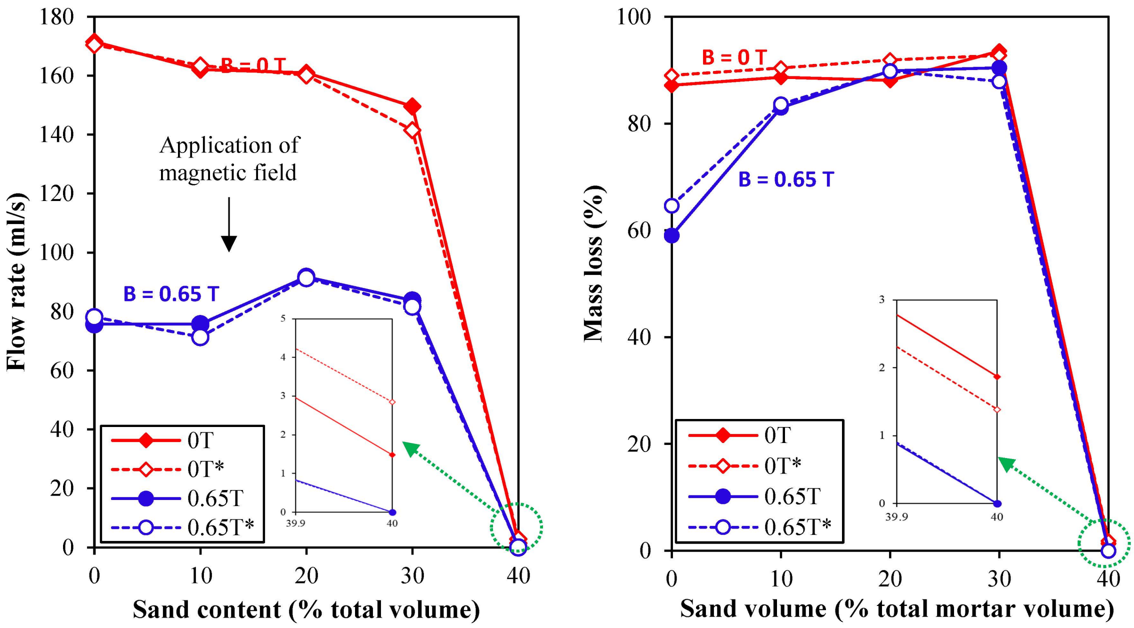

Figure 11. Effect of sand content on (**left**) mass flow rate, (**right**) mass loss (%). * Repetition of test on a different batch.

Our results showed that when there are no sand grains in the mixture, the packing density is lower, the excess water is higher, and the MNPs have more freedom to move and form clusters. As the clusters deposit on the walls of the outflow, they accumulate and lead to complete blockage of the flow. When the sand volume is increased up to 30%, the packing density is increased, and the excess water decreased. However, it is hypothesised that the percentage concentration of sand in the mixture is still sufficiently low, such that the interparticle interactions between the sand particles are still below the level at which large interlocking actions can be observed. There is still sufficient room for the aggregates to move freely within the paste. However, this is theorised to have a negative effect on the formation of clusters, in that the movement of the aggregates within the paste can lead to the breakage of magnetic clusters formed during the shearing action caused by the applied overhead pressure. This could explain why the M1, M2, and M3 pastes did not block completely despite having a higher or similar magnetic response in the storage modulus results in comparison to the M0 mixture. For the M4 mixture, however, it is assumed from the higher packing density and low excess water that there is sufficient interparticle interaction between the sand grains to cause interlocking effects, particularly as the fluid is flowing through the outflow region. The increased friction between particles leads to a jamming of the flow and leads to filtration effects. This is consistent with the theory that the probability of a granular blocking event increases when the particles are closer to each other, which becomes more probable when the volume fraction of particles is increased. In fact, if the mixture is stable such that there is no dynamic segregation that would locally increase the volume at the flow confinement, granular blocking during concrete casting is said to only fractionally be dependent on the number and size of the coarsest particles passing through the confinement, the volume fraction, and the size of the confinement (e.g., space between reinforcement bars), and not necessarily the rheological behaviour of the suspending fluid [31]. Considering that no visible segregation or bleeding was observed in the mixtures used in this study, it can be assumed that the blocking was caused by the increase in volume concentration of sand coupled with the flow confinement. Due to the non-homogenous nature of the mixtures, the blocking induced the filtration of the interstitial fluid through the granular skeleton. The applied overhead pressure enhances the friction between the grains, increasing the stresses and pressure gradients, which in turn increases the fluid filtration [32].

When a magnetic field was applied, however, the magnetic particles form clusters that further refine the voids in the clog created by the agglomeration of sand particles at the outflow. Due to this pore refinement, the loss of interstitial fluid is prevented, thereby preventing the formation of honeycombs in the finished surface, as shown in Figure 12, which shows the hardened surface of the M4 mixture after the tests with and without the ASC method. A previous study shows that if the fluid intrinsically exhibits blocking, the application of the magnetic field further enhances this blocking behaviour [9]. Thus, although the magneto response is decreased due to increasing sand content, this is overwhelmingly compensated for by the increase in particle interactions which lead to blocking.

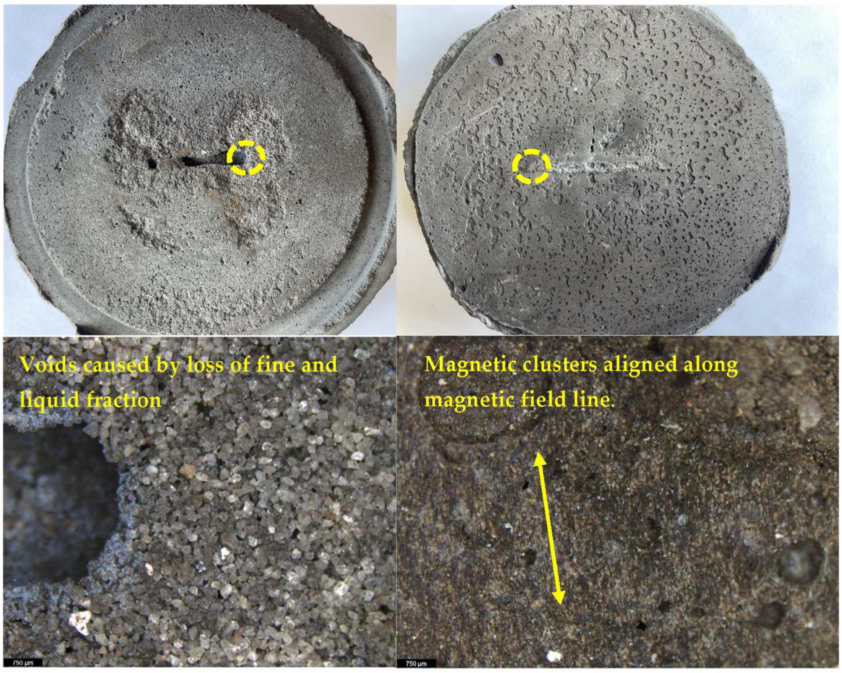

Figure 12. Hardened surface after the test: (**left**) no magnetic field application; (**right**) magnetic field of 0.65 T applied.

4. Conclusions

Effect of volume fraction of fine sand on the magnetorheological response and blocking mechanisms of cementitious mixtures containing Fe_3O_4 nanoparticles was investigated for the application of formwork leakage control. From the preceding results and discussions, the following conclusions can be drawn:

(1) The magnetorheological response, measured using the oscillatory time sweep tests, increases with increasing volume fraction of sand until a threshold value after which the presence of fine aggregates begins to limit the movement of the MNPs.
(2) Using ASC, the flow rate is significantly reduced for all mixtures due to the formation of magnetic clusters at the outflow. However, the flow reduction is reduced when the sand concentration is low in comparison to the reference paste.
(3) The filtration of the interstitial fluid caused by clogging effects at higher aggregate content can be reduced or eliminated when the ASC methodology is applied due to the pore refinement resulting from the formation of magnetic clusters.

In summary, the ASC methodology can be used on mortar, with the expectation that there would be a reduction in the magnetorheological effect with increasing volume of fine aggregates. Regardless, there is still great potential for the ASC method to be used for formwork leakage control due to the counteracting blocking effect caused by the increased particle interactions, which are further enhanced when the ASC method is applied. Further studies are needed to reveal the limits of the active stiffening control method to reduce or prevent formwork leakage.

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