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<td><strong>Author(s)</strong></td>
<td>Ruan, Shaoqin; Qiu, Jishen; Yang, En-Hua; Unluer, Cise</td>
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Fiber-reinforced reactive magnesia-based tensile strain-hardening composites

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Abstract:
This study focuses on the development of a new strain-hardening composite (SHC) involving carbonated reactive MgO cement (RMC) and fly ash (FA) as the main binder. Rheological properties of the developed composites were investigated by varying FA and water contents to achieve desirable fiber dispersion. A suitable mix design, in which polyvinyl alcohol (PVA) fibers were introduced to provide tensile ductility, was determined. The effect of key parameters such as w/b ratio and curing age on the mechanical properties of carbonated RMC-SHC was evaluated. Adequate binder content and w/b ratio was necessary for desirable fiber dispersion. Lower water contents and longer curing ages contributed to the strength development of RMC-SHC by improving the fiber-matrix interface bond and enhancing the formation of a dense carbonate network.

Keywords: MgO; Rheology; Carbonation; Tensile properties; Fiber reinforcement
1. Introduction

Portland cement (PC)-based concrete is the most frequently used construction material. The annual global production of PC exceeds 4 billion tons [1]. The production of PC is associated with a high energy consumption and ~8% of the total global anthropogenic carbon dioxide (CO₂) emissions [2]. The notable environmental impact of PC production has driven the search for alternative binders with potentially lower environmental footprints. Reactive magnesia (MgO), obtained via the calcination of magnesite (MgCO₃) at a temperature of 700-1000 °C (dry route) [3], magnesium silicates (e.g. serpentine) [4] or extracted from seawater or brine (wet route) [5], is of interest as a new binder.

Several studies have looked into the use of reactive MgO cement (RMC) as the main binder with or without the use of PC, fly ash (FA) or slag [6-12]. Most of these studies reported the strength development of RMC-based mixes via the carbonation of MgO, following its hydration into brucite (Mg(OH)₂). When in the presence of CO₂, these Mg-phases carbonate and form a range of hydrated magnesium carbonates (HMCs), such as nesquehonite (MgCO₃·3H₂O), hydromagnesite (4MgCO₃·Mg(OH)₂·4H₂O), dypingite (4MgCO₃·Mg(OH)₂·5H₂O) and artinite (MgCO₃·Mg(OH)₂·3H₂O). The hardened dense carbonate network reduces sample porosity and provides binding strength within RMC-based samples [13-15].

In addition to laboratory-scale samples reported in many studies, the use of RMC in the production of commercial-scale masonry units has been demonstrated in earlier studies [16, 17], highlighting its feasibility to be utilized in various non-structural applications. Recent studies on the use of RMC as a binder focus on understanding the factors that affect the carbonation and the associated strength development of RMC formulations, including mix design [18], curing conditions [14, 19] and use of additives [10, 11, 13, 19, 20], leading to 28-day concrete strengths as high as ~60 MPa [10]. Although continuous research on RMC formulations has achieved significant improvements in terms of mechanical performance, RMC-based concrete is still considered as a brittle material, which could highly benefit from the use of reinforcement in the development of structural...
members. Nevertheless, the relatively low pH (i.e. ~10) of carbonated RMC formulations presents a challenge in the use of steel reinforcement [9], which can face a risk of corrosion due to the loss of the passivated surface at such relatively low alkalinites, thereby potentially creating structural safety issues.

To increase the application spectrum of RMC within the construction industry, it is curial to develop alternative methods that will enhance the ductility of RMC-based formulations. The inclusion of a small amount of short polymeric fibers has been proven to be very effective in eliminating the brittleness and enhancing the tensile ductility of PC-based composites [21]. One of the successful derivatives involving the use of fibers is engineered cementitious composites (ECC), which adopt polymeric fibers at a fraction of typically ~2% by volume. Unlike the strain-softening conventional PC, ECC demonstrate strain-hardening behavior as their tensile stress continues to increase even in the presence of cracks. ECC samples can achieve a tensile ductility of 1-5%, enabled by the formation of multiple fine cracks (< 100 μm) with very small spacing (1-5 mm) [22-24]. The tensile properties of ECC can be further tailored with micromechanics to achieve multiple attributes such as high impact resistance [25, 26] and fatigue resistance [27-29]. Therefore, to reduce the dependence of RMC-based structural members on steel reinforcement, the use of short polymeric fibers, which can provide similar tensile strain-hardening behavior and high ductility within RMC formulations, can be explored.

One of the key factors to be considered in increasing the ductility of RMC-based strain-hardening composites (SHC) is the provision of desirable fiber dispersion. Previous studies on the rheology of ECC mixtures have shown that a relatively low yield stress (i.e. to ensure the flowability of the mixture) and high plastic viscosity (i.e. to provide sufficient shear force for the separation of the micro-fibers, which are bound together in their original form) are desirable properties [30]. The required yield stress and viscosity can be achieved by adjusting both the water/binder (w/b) ratio and FA content. Specifically, while a lower w/b ratio can lead to a high viscosity, it can also cause an undesirably high yield stress [31]. Alternatively, the replacement of cement by FA can
significantly reduce the yield stress without compromising viscosity due to the distinct spherical shape and similar particle size of FA as ordinary cement. The relationship between the rheology of cement pastes and tensile ductility, assessed via the measurement of the uniformity of fiber dispersion, has revealed that an increase in viscosity can lead to higher ductility [32].

While a desirable fiber dispersion is critical, it does not necessarily guarantee strain-hardening behavior or high ductility. As shown in Fig. 1, the hardened composites must satisfy the two strain-hardening criteria [33]. Specifically, the complementary energy of the fiber-bridging curve, $J_b'$ (i.e. the hatched area), must be larger than the crack tip toughness of the matrix, $J_{tip}$, as shown in Equation 1. Furthermore, the fiber-bridging strength, $\sigma_0$ (i.e. the curve peak), must be higher than the tensile cracking strength of matrix, $\sigma_c$, as shown in Equation 2.

$$J_{tip} \leq \sigma_0 \delta_0 - \int_0^{\delta_0} \sigma(\delta) d\delta \equiv J'_b \quad (1)$$

$$\sigma_c < \sigma_0 \quad (2)$$

As the fiber-bridging and matrix crack tip toughness are influenced by factors such as the w/b ratio [13], it is important to study the effect of these factors on the tensile performance of RMC-SHC. Until now, no studies have been reported on the development of RMC-based fiber-reinforced samples. In this respect, this paper presents a pioneering study on the development of RMC-based SHC by investigating the influence of key parameters throughout this process. The first part of the study focuses on the effect of w/b ratio and FA content on the rheological properties of RMC pastes, with the goal of determining a suitable mix design that leads to a desired viscosity and sufficient flowability for good fiber dispersion. The second part focuses on the inclusion of fibers within the mix design determined in the first stage and investigates the effect of certain factors such as the w/b ratio, fiber aspect ratio, fiber surface treatment and curing age on the performance of the developed formulations.
2. Materials and Methodology

2.1. Materials

Reactive MgO cement (RMC), obtained from International Scientific Ltd. (Singapore), was the main binder used in this study. Class F fly ash (FA), obtained from Bisley Asia Ltd. (Malaysia), was used to adjust the rheology of the fresh mixtures and function as a binder via its limited reaction with brucite [34]. The chemical composition and particle size distribution of these two binders are provided in Table 1 and Fig. 2, respectively. Sodium hexametaphosphate (Na(PO3)6), acquired from VWR International Ltd. (Singapore), was added to the prepared formulations as a water reducer at 10% of the water content [35]. Polyvinyl alcohol (PVA) fibers, one of the most common types of fibers used in ECC [36], were obtained from Kuraray Ltd. (Japan) and included in selected RMC-SHC samples. The properties of the PVA fibers used in this study are listed in Table 2.

2.2. Methodology

The experimental program designed in this study consisted of two parts. The first part studied the effect of w/b and FA/b ratios on the rheology of fresh mixtures before the inclusion of fibers. The goal of this part was to determine a suitable mix design (i.e. w/b ratio and FA content) that led to a desired plastic viscosity and flowability for good fiber dispersion. In the second part, during which PVA fibers were introduced into the mix design determined in the first part, the effects of the w/b ratio and curing age, whose increase can lead to a higher degree of carbonation and improve the strength development of RMC mixes [14], on the mechanical properties of RMC-SHC were studied.
The details of the seven different mix proportions prepared in the first part of the study are provided in Table 3. The notation for sample names followed a format of FA(X)-(Y), where X represented the FA content (i.e., as a percentage of the total binder) and Y represented the w/b ratio. A range of w/b (0.47-0.58) and FA/b (0-0.6) ratios were determined from preliminary samples prepared for each formulation. The sample preparation process started with the dissolving of Na(PO₃)₆ in the pre-determined amount of water. This solution was then added to the dry mix of RMC and FA during the mixing process. A stopwatch was set to notify two minutes from the moment the solution was added to the dry RMC-FA mix. After two minutes of mixing, one spoon of the fresh mixture was placed onto a 39 mm proliferated base plate on the rheometer, equipped with a 39 mm P35 TiL top plate pressed against the mixture at 0.5 N. The rheology measurements were performed via a HAAKE MARS III 379-0400 rheometer, which was used to measure the shear resistance of the fresh mixtures at a designated rotation speed according to the test setup shown in Fig. 3. Three rheology measurements were taken at 6, 12 and 18 minutes. For each measurement, the top plate was first rotated at a relatively high speed (50 /s) for 30 seconds to prevent any agglomeration [37], followed by an increase in the shear rate from 1 to 100 /s within 180 seconds. The shear resistance, \( \tau \), was recorded throughout the total 210 seconds. The sample was kept in-situ between measurements. Both the sample preparation and subsequent measurements were conducted under an ambient temperature (25 °C).

As a certain level of plastic viscosity is crucial for good fiber dispersion and tensile ductility [30], results from the first part of the study were used in the selection of w/b and FA/b ratios to be incorporated in the mix designs in the second part. Previous literature [32] has shown that while the fiber dispersion coefficient increased with plastic viscosity, tensile ductility stabilized after a threshold fiber dispersion coefficient. This value corresponded to a plasticity of about 3.5 Pa·s, which was also adopted in this study for the preparation of samples used in the testing of tensile properties. Referring to the results obtained in the first part of the study to identify samples that demonstrated good fiber dispersion (e.g., FA30-0.53), the three mix proportions listed in Table 4 were prepared to assess the tensile performance of RMC-SHC. For all the mix designs, the
FA/b was fixed at 0.3, whereas the w/b ratio was kept at ≤ 0.53 according to the outcomes of the rheology measurements. A constant fiber fraction corresponding to 2% of the binder content was utilized in line with the previous literature on ECC [36].

To initiate sample preparation, RMC and FA were dry mixed in a planetary mixer for over 3 minutes, after which the prepared Na(PO₃)₆ solution was slowly added to form the fresh mixture. The mixing process continued for an additional 2-3 minutes until a uniformly mixed homogenous mixture was achieved, after which the PVA fibers were slowly added within 2 minutes. During the entire mixing process, the blade rotation speed was kept constant at 6.4 rad/s. The prepared mixture was cast into cubic (50x50x50 mm) and dogbone shaped molds, whose dimensions are shown in Fig. 4(a). The cast samples were stored in a sealed container, where silica gel was used for removing the excess moisture from the pastes to facilitate the penetration of CO₂ within the sample pore structure in the subsequent stage [38]. After 3 days, samples were removed from their molds and cured in a carbonation chamber set at a CO₂ concentration of 10%, temperature of 30±1.5 °C and relative humidity (RH) of 85±5% (i.e. ambient levels in Singapore) for 7 days [13, 20]. The effect of curing duration was studied by exposing one of the prepared samples (RMC-0.41) to carbonation curing for a total of 28 days under the same conditions.

Once curing was completed, the compressive strength of cubic samples were measured in triplicates in accordance with the specifications of ASTM C109/C109M-13 [39]. The equipment used for compression testing was Landmark 370.25, operated at a loading rate of 0.25 mm/min. The uniaxial tensile tests were conducted on at least three specimens for each mix design by using an Instron 5569, as shown in Fig. 4(b). During this test, the loading rate was set at 0.02 mm/min and two LVDTs were used to determine the extension of the gauged length (60-70 mm). After the uniaxial tensile test, the crack width and spacing (i.e. gauge length/crack number) on each specimen were determined with a Nikon DS-Fi2 high resolution camera equipped with NIS Elements (Nikon) software and an OPTEM Zoom 70XL microscope at a magnification of 420x. For each mix design, three to six samples were analyzed to evaluate their crack
patterns, which involved capturing the image with the camera, and counting and recording the crack number within the gauge length of sample under the microscope.

In addition to their compressive and tensile strengths, the CO₂ uptake of selected samples during carbonation curing were assessed by obtaining representative powders from each sample. These powders were ground to pass through a 75 μm sieve and vacuum dried before analysis. The quantitative measurements were performed via thermogravimetric analysis/differential scanning calorimetry (TGA/DSC) conducted on a Perkin Elmer TGA 4000 equipment. During TGA/DSC, the samples were heated from 40 to 900 °C at a heating rate of 10 °C/min under nitrogen flow.

3. Results and Discussion

3.1. Rheological properties

Fig. 5 illustrates the typical relationship between the shear resistance τ (Pa) and shear rate $N$ (/s) of a fresh RMC mixture at different elapsed times. The curves show that the fresh mixtures were Bingham liquids, in which the shear force exceeded the initial mixture resistance to initiate rotation, after which the shear resistance increased linearly with the rotation speed $N$ (/s), showing no shear thinning or thickening effect. The relationship between τ and $N$ of Bingham liquids is quantitatively described by Equation 3, where $g$ (Pa), the intercept on the y-axis, is the yield stress needed to break the network of interactions between particles and initiate rotation; and $h$ (Pa s), the slope of the curve, is the plastic viscosity [40]. For any given fresh mixture at a designated time, $g$ and $h$ are constants that represent the rheological properties of that mixture.

$$\tau = g + Nh$$

In regular cement-based fresh mixes, shear resistance, which varies with shear rate ($N$) and solid volume fraction ($V_s$), depends on several types of particle interactions (i.e. Van
der Waals forces, direct contact forces between particles and hydrodynamic forces, for
which the friction between fluid layers increases with velocity) [41]. At relatively low
shear rates (i.e. whose threshold depends on the w/b ratio [42, 43]), the yield stress \( (g) \)
is mainly determined by Van der Waals or direct contact forces. Previous studies on the
yield stress of C₃S pastes [44] showed that a critical \( V_s \) existed at 0.38, beyond which
the solid particles became so compacted that the dominant particle interaction shifted
from Van der Waals to direct contact forces. As C₃S and RMC have a comparable
particle size distribution [9], and they both conform to Bingham model, the information
on C₃S was used to evaluate the behavior of RMC-based samples. It must be noted
that the yield stress can also be related to the surface chemistry of particles, which was
not differentiated here. Most of the mix designs presented in this study, except for FA0-
0.58 \( (V_s = 0.35) \) and FA30-0.58 \( (V_s = 0.37) \), had \( V_s \) values that were larger than 0.38,
indicating the dominance of direct contact forces. At relatively high shear rates, the yield
stress is majorly determined by hydrodynamic forces, while the effects of Van der Waals
and direct contact forces still exist.

The measured values of the yield stress \( (g) \) and plastic viscosity \( (h) \) of all samples at
different elapsed times are provided in Table 5. For some mixes, the values of \( g \) and \( h \)
at 12 and 18 minutes were not listed due to the loss of contact between the top plate
and the fresh mixture, resulting in the underestimation of shear resistance. A generally
increasing trend in \( g \) and \( h \) was observed with elapsed time. This was possibly
associated with the precipitation of brucite (Mg(OH)₂) on the surface of RMC particles
[45], which increased the direct contact between particles by enlarging the solid particle
size and leading to additional drag between fluid layers.

The effects of water and FA contents on the rheological properties of RMC samples are
shown in Fig. 6, where a declining trend in \( g \) and \( h \) was observed with increasing water
content at both FA/b ratios of 0.53 and 0.58. The reduction in plastic viscosity and yield
stress with increasing w/b ratios could be attributed to the higher liquid content that
decreased direct contact amongst particles. Regarding the effect of FA content, the
trend in \( g \) and \( h \) values at different FA contents ranging between 0% and 60% was
shown at a w/b of 0.58. An increase in the FA content from 0 to 30% (i.e. \( V_s < 0.38 \), Van der Waals forces dominate) led to an increase in plastic viscosity, while it did not have a profound effect on the yield stress. This increase in the plastic viscosity could be due to the enhanced Van der Waals forces via the reduction of the distance between particles as \( V_s \) increased from 0.35 to 0.37. The constant yield stress of samples FA0-0.58 and FA30-0.58 could be associated with the loose compaction of particles within these two mixes, thereby limiting the effect of particle shape on Van der Waals forces. A further increase in the FA content from 30% to 60% (i.e. \( V_s > 0.38 \), direct contact force dominates) resulted in the decline of both \( g \) and \( h \) at all w/b ratios. Although an increase in the solid volume fraction with an increase in the FA content could be expected due to the lower density of FA than RMC (2400 vs. 3230 kg/m\(^3\)), the reduction of the yield stress and plastic viscosity with increasing FA content could be attributed to the spherical geometry of FA particles, which reduced the contact force between particles.

### 3.2. Mechanical performance

Based on the rheological results mentioned in Section 3.1, samples containing 30% FA (i.e. FA/b = 0.3) with different w/b ratios (0.41-0.53) and curing ages (7 and 28 days) were prepared for compression and uniaxial tensile tests. The obtained results are presented in Table 6. The tensile stress recorded at the occurrence of the first crack is referred to as the “first cracking strength”, whereas the tensile stress and tensile strain approaching specimen failure are referred to as the “ultimate tensile strength” and “tensile strain capacity”, respectively. In addition to the first cracking strength, ultimate tensile strength, and tensile strain capacity, crack spacing and average crack width of each sample are also provided in Table 6.

The relationship between the tensile stress and strain within each mix are shown in Fig. 7. Distinct elastic and strain-hardening stages were observed within each sample. In the elastic stage, the tensile stress developed linearly with the strain. The continuous increase in the load led to the introduction of the first crack, which marked the beginning
of the strain-hardening stage. During the strain-hardening stage, tensile stress increased slowly with the strain, accompanied with the progressive generation of multiple fine cracks that indicated ductility. Towards the end of strain-hardening, tensile stress started to drop dramatically due to the deterioration of fiber bridging followed by damage localization with increasing load. At this point, the load was released immediately after specimen failure, leading to the shrinking of a majority of the cracks due to the spring effect of fiber-bridging. Fig. 8 shows the typical crack distribution on a failed specimen after unloading, highlighting the occurrence of multiple cracking in RMC samples. The typical fracture surface can be seen in Fig. 9(a), where the layout of fibers at the location of the crack at failure is revealed. The pulled-out fiber and the leftover fiber tunnel at a fracture point are shown in Fig. 9(b) and (c), respectively. As can be seen from these images, the fiber surface was smooth with a very small amount of matrix debris attached on it, showing that a majority of the fibers were pulled out instead of ruptured. This suggests the fiber strength was not fully utilized in the developed formulations and the limiting phase was the fiber-matrix interface, which may be further strengthened.

3.2.1. Effect of w/b ratio

The results obtained after the mechanical testing of samples RMC-0.53, RMC-0.47 and RMC-0.41 are listed in Table 6. The influence of w/b content on the performance of each sample was observed both in terms of compressive and tensile strengths. As could also be seen in Fig. 7, the mechanical properties were enhanced when the w/b reduced from 0.53 to 0.47, where a less pronounced change was observed as the w/b further reduced to 0.41.

The initial reduction in the w/b from 0.53 to 0.47 led to a compressive strength that was almost four times higher (4.6 vs. 16.3 MPa). This was accompanied with a 50% increase in the first tensile crack strength, which mainly depended on the matrix performance. This improved performance could be associated with the increased

diffusion rate of CO\textsubscript{2} in a less saturated pore system, as well as the higher initial density provided by the lower water content within sample RMC-0.47. The increased diffusion of CO\textsubscript{2} could translate into a higher degree of carbonation and hence the formation of strength providing HMCs [14]. The other tensile properties, such as the ultimate tensile strength and ductility, which were mainly determined by the fiber-bridging, were also improved under lower water contents. The ultimate tensile strength was more than doubled, while the ductility was increased by about 50\%. The reduction in crack spacing indicated that strain-hardening became more robust as the w/b ratio decreased. The enhanced fiber bridging, which was clearly indicated by the reduced crack width, could also be attributed to the higher degree of carbonation and denser microstructure at the fiber/matrix interface [46, 47]. When compared to RMC-0.53, the enhanced fiber-bridging of RMC-0.47 also led to a higher complementary energy $J'_b$, resulting in a more saturated multiple cracking. The reduced crack width would be especially important for any potential crack healing [48].

A further decrease in the w/b ratio from 0.47 to 0.41 (i.e. at 7 days) led to improved mechanical performance, albeit at a lower rate than previously observed. When compared to the increase in performance when the w/b changed from 0.53 to 0.47, the lower rate of increase observed in the transition of the w/b from 0.47 to 0.41 could be explained by the limited hydration of RMC due to the lower availability of water, which could have also hindered the formation of HMCs in the longer term. On the other hand, as the diffusion of CO\textsubscript{2} is faster in drier environments, the relatively low w/b of 0.41 could have provided the right medium for increased carbonation, which can explain the higher compressive strength results of sample RMC-0.41 [14]. Another factor that may have contributed to the relatively higher strength results of this sample was its reduced porosity within the interfacial zone of fiber-matrix under the low water content, thereby resulting in a denser structure than samples RMC-0.47 and RMC-0.53. The reduction in crack spacing and average crack width were also an indicator of improved strain-hardening and fiber bridging.
3.2.2. Effect of curing age

While a robust strain-hardening and ultra-high ductility were achieved in RMC-SHC samples, their mechanical properties were still not at the same level as typical PC-based SHC, whose ultimate tensile strength and ductility can easily reach 5 MPa and 3-5%, respectively [49]. Accordingly, samples with the highest ductility (RMC-0.41) were chosen to be exposed to additional CO\(_2\) curing for up to 28 days to explore the full potential of RMC-SHC samples in terms of tensile strength enhancement. A comparison of samples RMC-0.41-7d and RMC-0.41-28d revealed an increase in the ultimate tensile strength by ~40%, which could be attributed to the continued formation of carbonate phases that strengthened the fiber/matrix interface over the 28-day curing period [14]. On the other hand, there were no notable changes in the tensile ductility, crack spacing and crack width. These results indicated the feasibility of enhancing the ultimate tensile strength of RMC-SHC formulations via the effective use of additional CO\(_2\) curing, during which their strain-hardening robustness was not compromised.

3.3. CO\(_2\) sequestration

The amount of CO\(_2\) sequestered during the curing process was quantified via TGA/DSC, a representative curve for which is shown in Fig. 10. The mass loss < 100 °C due to the loss of hydroscopic water was followed by two distinct endothermic peaks. The first peak at around 320 °C corresponded to the removal of water of crystallization in Mg-carbonates that formed during carbonation curing and the decomposition of uncarbonated hydrates (Mg(OH)\(_2\)) into MgO. The second peak at around 460 °C corresponded to the decarbonation of carbonate phases, leaving MgO at the end of the analysis. The quantification of the mass loss at > 460 °C, which was associated with the loss of CO\(_2\) from the carbonated RMC system, revealed an average carbonation degree of around 10%. A similar outcome can be expected under ambient carbonation conditions, albeit at a much slower rate due to the low concentration of CO\(_2\) in the atmosphere (0.04%). These results indicate that the sequestration of CO\(_2\) in the form of
stable carbonates within the prepared formulations can not only provide a safe storage for atmospheric CO$_2$, but also contribute to the development of a new type of strain-hardening composite that does not necessitate the use of any PC.

4. Conclusions

This study presented a preliminary investigation focusing on the development of ductile carbonated reactive MgO cement (RMC)-based strain-hardening composites (SHC) that did not involve the use of any PC. The effect of key factors such as the mix design (i.e. binder component and water content) on the rheological properties of fresh RMC mixtures was studied. The obtained results revealed the ideal binder composition and w/b ratios that led to a relatively high plastic viscosity and sufficient flowability for desirable fiber dispersion. Compressive and tensile strength tests were conducted to evaluate the effect of water content and curing age on the mechanical performance of the developed formulations. The use of carbonation curing enhanced the strength development of RMC-SHC by improving the fiber-matrix interface bond, but not ductility. Lower water contents led to increased tensile strength and ductility, which was attributed to the strengthening of the bond between the fibers and the matrix. The prepared RMC-SHC samples, which presented a considerable CO$_2$ sequestration capability during the curing process, achieved an ultimate tensile strength and ductility of up to 3.7 MPa and 3.3%, respectively. The RMC-FA formulations developed in this study, with a potentially lower environmental impact than corresponding PC-based mixes due to their ability to gain strength via carbonation, are considered as a promising candidate for strain-hardening composites that can be used in various building applications.

Acknowledgement
The authors would like to acknowledge the financial support from the Singapore MOE Academic Research Fund Tier 1 (RG 95/16) for the completion of this research project.

References


List of Tables:

**Table 1** Chemical composition of RMC and FA.

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<tr>
<th>Material</th>
<th>MgO</th>
<th>CaO</th>
<th>SiO&lt;sub&gt;2&lt;/sub&gt;</th>
<th>Fe&lt;sub&gt;2&lt;/sub&gt;O&lt;sub&gt;3&lt;/sub&gt;</th>
<th>Al&lt;sub&gt;2&lt;/sub&gt;O&lt;sub&gt;3&lt;/sub&gt;</th>
<th>K&lt;sub&gt;2&lt;/sub&gt;O</th>
<th>TiO&lt;sub&gt;2&lt;/sub&gt;</th>
<th>Others</th>
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<td>RMC</td>
<td>97%</td>
<td>1.3%</td>
<td>1.3%</td>
<td>0.2%</td>
<td>0.2%</td>
<td>-</td>
<td>-</td>
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<tr>
<td>FA</td>
<td>0.8%</td>
<td>1.2%</td>
<td>58.6%</td>
<td>4.7%</td>
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Table 2 Properties of the PVA fibers used in this study.

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<th>Length (mm)</th>
<th>Diameter (μm)</th>
<th>Fiber aspect ratio, i.e. length/diameter</th>
<th>Density (kg/m³)</th>
<th>Nominal tensile strength (MPa)</th>
<th>Surface oil-content (%)</th>
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<td>12</td>
<td>39</td>
<td>307</td>
<td>1300</td>
<td>1600</td>
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Table 3 Mix proportions of fiber-free RMC mixtures prepared for rheology measurements.

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<thead>
<tr>
<th>Sample</th>
<th>RMC (kg/m³)</th>
<th>FA (kg/m³)</th>
<th>Water (kg/m³)</th>
<th>Na(PO₃)₆ (kg/m³)</th>
<th>FA (%)</th>
<th>w/b</th>
<th>Solid (RMC+FA) volume fraction</th>
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<tr>
<td>FA0-0.58</td>
<td>1102</td>
<td>0</td>
<td>639</td>
<td>64</td>
<td>0</td>
<td>0.58</td>
<td>0.35</td>
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<tr>
<td>FA30-0.47</td>
<td>840</td>
<td>360</td>
<td>570</td>
<td>57</td>
<td>30</td>
<td>0.47</td>
<td>0.42</td>
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<td>FA30-0.53</td>
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<td>338</td>
<td>595</td>
<td>59</td>
<td>30</td>
<td>0.53</td>
<td>0.39</td>
</tr>
<tr>
<td>FA30-0.58</td>
<td>744</td>
<td>319</td>
<td>617</td>
<td>62</td>
<td>30</td>
<td>0.58</td>
<td>0.37</td>
</tr>
<tr>
<td>FA60-0.47</td>
<td>462</td>
<td>693</td>
<td>548</td>
<td>55</td>
<td>60</td>
<td>0.47</td>
<td>0.44</td>
</tr>
<tr>
<td>FA60-0.53</td>
<td>435</td>
<td>653</td>
<td>573</td>
<td>57</td>
<td>60</td>
<td>0.53</td>
<td>0.42</td>
</tr>
<tr>
<td>FA60-0.58</td>
<td>411</td>
<td>616</td>
<td>596</td>
<td>60</td>
<td>60</td>
<td>0.58</td>
<td>0.39</td>
</tr>
</tbody>
</table>

* The rheology measurements performed on samples FA0-0.47 and FA0-0.53 were not presented as these fresh mixtures hardened too fast, resulting in incorrect results. Specifically, once the plate-to-plate spinning started, these samples immediately dislodged and were no longer in full contact with the upper plate.
Table 4 Mix proportions prepared for testing hardened tensile properties.

<table>
<thead>
<tr>
<th>Sample</th>
<th>RMC (kg/m³)</th>
<th>FA (kg/m³)</th>
<th>Water (kg/m³)</th>
<th>Na(PO₃)₆ (kg/m³)</th>
<th>PVA fiber (kg/m³)</th>
<th>w/b</th>
<th>Curing duration (days)</th>
</tr>
</thead>
<tbody>
<tr>
<td>RMC-0.53</td>
<td>799</td>
<td>342</td>
<td>601</td>
<td>59</td>
<td>26</td>
<td>0.53</td>
<td>7</td>
</tr>
<tr>
<td>RMC-0.47</td>
<td>853</td>
<td>365</td>
<td>573</td>
<td>53</td>
<td>26</td>
<td>0.47</td>
<td>7</td>
</tr>
<tr>
<td>RMC-0.41</td>
<td>864</td>
<td>370</td>
<td>510</td>
<td>51</td>
<td>26</td>
<td>0.41</td>
<td>7 and 28</td>
</tr>
</tbody>
</table>
Table 5 Rheological test results.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Elapsed time = 6 min</th>
<th>Elapsed time = 12 min</th>
<th>Elapsed time = 18 min</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>g (Pa)</td>
<td>h (Pa·s)</td>
<td>g (Pa)</td>
</tr>
<tr>
<td>FA0-0.58</td>
<td>60.6</td>
<td>1.15</td>
<td>-</td>
</tr>
<tr>
<td>FA30-0.47</td>
<td>153.1</td>
<td>5.55</td>
<td>-</td>
</tr>
<tr>
<td>FA30-0.53</td>
<td>172.8</td>
<td>3.66</td>
<td>183.3</td>
</tr>
<tr>
<td>FA30-0.58</td>
<td>48.0</td>
<td>2.59</td>
<td>76.0</td>
</tr>
<tr>
<td>FA60-0.47</td>
<td>26.2</td>
<td>2.24</td>
<td>40.3</td>
</tr>
<tr>
<td>FA60-0.53</td>
<td>6.1</td>
<td>1.13</td>
<td>11.0</td>
</tr>
<tr>
<td>FA60-0.58</td>
<td>2.1</td>
<td>0.43</td>
<td>1.6</td>
</tr>
</tbody>
</table>

* "-" indicates g and h were underestimated due to the loss of contact between the top plate and the fresh mixture.
Table 6 Mechanical test results.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Curing age (days)</th>
<th>Compressive strength (MPa)</th>
<th>Results of uniaxial tensile test</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>First cracking strength (MPa)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Ultimate tensile strength (MPa)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Tensile strain capacity (%)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Crack spacing* (mm)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Average crack width (μm)</td>
</tr>
<tr>
<td>RMC-0.53</td>
<td>7</td>
<td>4.60 ±0.07</td>
<td>1.09 ±0.11</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1.32 ±0.09</td>
<td>1.40 ±0.66</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>12.80 ±3.50</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>146.80 ±22.90</td>
</tr>
<tr>
<td>RMC-0.47</td>
<td>7</td>
<td>16.27 ±0.47</td>
<td>1.60 ±0.13</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>2.89 ±0.26</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>2.13 ±1.02</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>2.66 ±1.10</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>49.74 ±12.88</td>
</tr>
<tr>
<td>RMC-0.41</td>
<td>7</td>
<td>18.97 ±0.48</td>
<td>2.19 ±0.48</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>2.61 ±0.48</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>2.64 ±1.22</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>1.45 ±0.14</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>47.21 ±15.08</td>
</tr>
<tr>
<td></td>
<td>28</td>
<td>-</td>
<td>2.38 ±0.24</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>3.67 ±0.35</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>2.70 ±0.96</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>1.74 ±0.27</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>63.77 ±10.38</td>
</tr>
</tbody>
</table>

*Crack spacing (mm) = Gauged length extension/crack number; a smaller crack spacing is usually associated with a higher degree of strain-hardening.
Fig. 1 Illustration of the fiber-bridging constitutive law, showing the tensile stress-crack opening displacement $\sigma(\delta)$ curve and strain-hardening criteria [33]
Fig. 2 Particle size distribution of RMC and FA
Fig. 3 Measurement of sample rheology, showing (a) rheometer used and (b) test setup
Fig. 4 Illustration of uniaxial tensile test, showing (a) dimensions of the dogbone specimen and (b) test setup
Fig. 5 $\tau$-$N$ curve of sample FA60-0.53 at 6, 12 and 18 minutes, showing (a) overall test results and (b) section selected in (a)
Fig. 6 Effects of the water and FA contents on the (a) plastic viscosity and (b) yield stress of RMC samples
Fig. 7 Tensile stress vs. strain curves of samples (a) RMC-0.53-7d, (b) RMC-0.47-7d, (c) RMC-0.41-7d and (d) RMC-0.41-28d
Fig. 8 Crack pattern of a typical sample (RMC-0.41 at 7 days)
Fig. 9 Illustration of a typical failed specimen (RMC-0.41 at 7 days), showing the (a) fracture surface with several pulled fibers, (b) FESEM image of a pulled-out fiber and (c) FESEM image of a left-over tunnel from the pulled fiber.
Fig. 10 A typical TGA/DSC curve of a RMC-SHC sample (RMC-0.53)