Mechanical and Micro-structural Investigation of Multi-scale Cementitious Composites Developed Using Sisal Fibres and Microcrystalline Cellulose

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Abstract

This research work attempts, for the first time, to use sisal fibres and microcrystalline cellulose (MCC) in combination for developing multi-scale cementitious composites with improved strength, modulus as well as fracture energy. MCC (0.1% to 3.0 wt.%) was first dispersed in water with the help of Pluronic F-127 surfactant (20% of MCC wt.) using ultrasonication and subsequently, the MCC suspensions were added to cement-sand mixtures containing sisal fibers (0.25 to 2 wt.% of cement). Visual observation, UV-Vis analysis and optical microscopy suggested that ultrasonication for only 15 min led to homogeneous MCC suspensions without significant sedimentation and Pluronic significantly reduced the total agglomerated area and the average MCC particle size in the suspensions. The use of up to 2 wt.% sisal fibres (without MCC) increased the fracture energy of cementitious composites by 351% (after 28 days hydration) but reduced the compressive and flexural strengths by 38% and 13%, respectively. On the contrary, MCC (without sisal fibres) could strongly improve the compressive strength (20.5% using 0.1 wt.% MCC) but was not found so effective in improving the fracture energy (improved by 29% using 0.1 wt.% MCC). However, multi-scale reinforcements containing 0.1 wt.% MCC and 0.25-0.5 wt.% sisal fibres led to improvements of up to 18.4%, 30.1%, 30% and 100% in the compressive strength, flexural strength, flexural modulus and fracture energy of composites, presenting distinct advantages over only sisal fibre or MCC-based reinforcements. The results indicated the formation of higher amount of hydration products in multi-scale composites due to the positive effect of MCC on cement hydration. Superior hydration in the multi-scale composites resulted in a denser microstructure with a lower pore size and an improved fibre-matrix interface, improving significantly the strength, modulus and fracture energy.

Keywords: Microcrystalline cellulose, Sisal fibers. Pluronic F-127, Dispersion, Multi-scale cementitious composites, Mechanical properties, Fracture energy.

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1. Introduction

Concrete is the most widely used construction material on earth and it is known for its high compressive strength (Torgal and Jalali, 2011). The global production capacity of cement in 2017 was 4100 million metric tons and is expected to be around 4.4 billion metric tons in 2020 (Statista, 2017; TERI, 2017). However, concrete has some inherent limitations and problems such as low tensile strength and cracking (Torgal and Jalali, 2011). Nano-scale cracks are formed within concrete during production and during subsequent stages, they conjoin to form micro and macro-cracks (Parveen et al., 2013). Moreover, the hydration of cement releases heat and increases the temperature of concrete. In massive structures, heat cannot release readily causing expansion, stresses and thermal contraction, which lead to cracking (Hoyos et al., 2013).

The incorporation of macro fibers into cementitious composites was found effective in delaying or preventing crack propagation (Fu et al., 2017; Peters et al., 2010; Soltan et al., 2017). Macro fibers (such as carbon, glass, steel, etc.) absorb energy through plastic deformation, friction, and fiber pull-out and can bridge macro-scale cracks, leading to an improved toughness of composites. These fibres are often added to the cement mixture to develop ultra high performance concrete (UHPC) which presents superior strength, toughness and durability over the classical concrete (M. M. R. Khosravani et al., 2019; M. R. Khosravani et al., 2019). UHPC holds great promise in the architectures with thin complex shapes, curvature and textures, and eliminates (or at least reduces) the need for steel reinforcements in some applications. Nano and microfibers, on the other hand, are effective in bridging nano and micro-cracks, respectively hindering their subsequent transformation into macro-cracks (Fu et al., 2017). Plant-based cellulose fibers such as sisal, jute, hemp, jute, coir, etc. are low-cost, less hazardous over synthetic fibres, eco-friendly and possess high specific mechanical strength (Parveen, 2017a). These fibres were found effective in significantly improving the fracture properties of concrete (Peters et al., 2010; Soltan et al., 2017). Plant fibre reinforcements led to an increased energy absorption and ductility of cementitious composites, which showed the capability to deform and support loads after the initial cracking. For example, the addition of short sisal fibers (25 and 50 mm long, randomly distributed, 1-6 vol%) to cement mortars resulted in significant improvement of toughness but at the cost of peak stress, strain at failure and elastic modulus. This reduction in mechanical properties has been a major problem in using these fibres in cementitious composites (Lima et al., 2014). The reduction of

strength and modulus of sisal and other plant fibre-based cement composites could be attributed to the increased porosity of mortars resulting from insufficient compaction, fibre agglomeration, unbound fibres and poor fibre/matrix bonding (Lima et al., 2014). It was also observed that the elastic modulus and strength of cementitious composites did not deteriorate when sisal fibres of shorter lengths (e.g. 25 mm as compared to 50 mm and 20 mm as compared to 40 mm) were used (Lima et al., 2014). These results further indicate the possibility of using cellulose nano/micro-dimensional fibres such as nanocellulose or microcrystalline cellulose (MCC) in cementitious composites without negative influence on the mechanical properties of composites.

MCC has excellent mechanical properties and is currently being considered as a sustainable reinforcement for composite materials (Dong et al., 2013; Chen et al., 2019; Parveen et al., 2017). Research has shown that the addition of MCC reduced the adiabatic temperature of cement and can be useful in large volumes of concrete to reduce thermal contraction and cracking (Hoyos et al., 2013). A number of other studies also showed positive effects of MCC on the mechanical properties of cementitious composites (Anju et al., 2016; Parveen et al., 2017, 2017c; Silva et al., 2018). Research suggested that the nano/microcellulose particles could improve the hydration behaviour of cementitious composites by facilitating diffusion of water molecules to the unhydrated parts of cement. This phenomena is known as the short-circuit diffusion mechanism (SCD) (Cao et al., 2015). However, the direct addition of MCC powder to cementitious composites was not found much effective due to agglomeration of MCC (Hoyos et al., 2013). Therefore, MCC has been processed using a number of dispersion techniques to ensure a homogeneous MCC dispersion within cementitious composites (Anju et al., 2016; Parveen et al., 2017a, 2017b; Silva et al., 2018). For example, cementitious composites with MCC (0.2% to 1.0 wt.% of cement) were developed by dispersing MCC in water using a magnetic stirring process for 45 min before adding to the cement mixture (Silva et al., 2018). Improvements of up to 20.5% in the flexural strength, 19.8% in the compressive strength, 100% in the flexural modulus and 27.2% in the fracture energy of composites were achieved after 28 days of hydration (Silva et al., 2018). Ultrasonic energy (15, 30, 45 and 60 min) was also used for homogeneously dispersing MCC in water for developing cementitious composites, leading to 96%, 19.2%, and 51.4% improvements in the flexural modulus, flexural strength and compressive strength of composites, respectively (Parveen et al., 2018). Previous studies also used stabilising agents, e.g. carboxyl methyl cellulose (CMC) and

surfactants, e.g. Pluronic®F-127 along with ultrasonic treatments to improve MCC dispersion. As compared to CMC, superior results were achieved with Pluronic due to its ability to disperse both cement and MCC particles (Parveen et al., 2017b) and 106%, 31% and 66% improvements were noticed in the flexural modulus, flexural strength and compressive strength of cementitious composites, respectively (Parveen et al., 2017b).

However, it has been frequently observed that the use of MCC in cementitious composites, although found very effective in improving strength and elastic modulus, could not improve the fracture properties significantly (Parveen et al., 2017b). The strain capacity and fracture energy of cementitious composites rather reduced due to the crystalline and stiff structure of MCC which restricted the mobility of matrix materials (Cao et al., 2015; Parveen et al., 2018). To overcome this problem, recently a multi-scale reinforcement (i.e., combination of nano and micro-scale reinforcements) comprising MCC and carbon nanotubes (CNTs) was used in cementitious composites looking at the high capacity of CNTs in improving fracture properties of cement (Alshaghel et al., 2018). CNTs possess excellent mechanical properties, very high aspect ratio and surface area and if dispersed homogeneously, can lead to strong improvements in strength, elastic properties as well as fracture performance of composites (Liew et al., 2016; Parveen et al., 2015; Tarfaoui et al., 2016). Using the above multi-scale reinforcement, besides improvements in flexural strength, flexural modulus and compressive strength, strong improvements in flexural strain and fracture energy were also noticed (Alshaghel et al., 2018). Nevertheless, CNT is expensive, has toxicity and dispersion problems and therefore, to find an alternative sustainable material sisal fiber has been used for the first time in this research in combination with MCC to develop a new bio-based and eco-friendly multi-scale reinforcement.

In summary, plant fibres have been frequently used as eco-friendly reinforcing materials for cementitious composites with significant improvements in strain capacity and ductility but at the cost of mechanical strength. Plant-based micro-reinforcements such as MCC, on the other hand, was effective in improving strength and modulus of cementitious composites but significantly reduced the fracture energy. Development of multi-scale composites combining MCC with CNTs could solve this problem. However, CNTs can be difficult to apply in the construction industry due to their high cost and toxicity problems. Consequently, it was necessary to find an alternative

low-cost and eco-friendly reinforcement for developing multi-scale composites. To address this need, the present study investigated the use of sisal fibres in combination with MCC for developing multi-scale cementitious composites possessing improved fracture energy along with superior strength and modulus. Although the effect of sisal fibres and MCC has been separately investigated in previous studies, to the best of authors' knowledge, the combined effect of sisal fibres and MCC as a multi-scale reinforcement on the properties of cementitious composites has not been reported till date. The effects of this new multi-scale reinforcement on the flow behaviour, flexural and compressive properties as well as microstructure of cementitious composites have been thoroughly investigated and discussed.

2. Experimental

2.1 Raw materials

MCC (Avicel[®] PH-101) was purchased from Sigma Aldrich, Portugal and sisal fibres were supplied from a local firm located in the Northeast region of Brazil. Pluronic[®] F-127 surfactant was purchased from Sigma Aldrich (Portugal) and the defoaming agent, tri-butyl phosphate (TBP) was purchased from Acros Organics (Thermo Fischer Scientific). The Ordinary Portland cement, CEM I 42.5 R (corresponding International standard: ASTM-C150, Type V) was obtained from Secil, Portugal and the standard sand (EN 196-1) was supplied by Société Nouvelle du Littoral (France). The important properties of raw materials used in the present study are listed in **Table 1**.

(Table1)

2.2. Methods

2.2.1 Characterisation of MCC and sisal fibres

MCC and sisal fibres were characterised for their morphology by using Scanning Electron Microscopy (FEG-SEM, NOVA 200 Nano SEM, FEI) at an acceleration voltage of 15 kV. Prior to SEM analysis the samples were coated with a 30 nm film of Au-Pd in a high resolution 208HR Cressington sputter coater. X-ray diffraction (XRD) analysis was performed on both MCC and sisal fibres to study their crystal structure. XRD was carried out in Bruker D8 Discover diffractometer using an angle range of 5-70°, step size of 0.04 and 2 s per step. The crystallinity index (CI) of MCC and sisal fibres was calculated using the following equation:

$$CI (\%) = 100 \times \frac{I_{002} - I_{am}}{I_{002}} \quad (1)$$

Where, I_{002} is the total peak intensity of crystalline and amorphous regions calculated from the height of 002 peak and I_{am} is the peak intensity of the amorphous region calculated from the height of the minimum between 002 and 101 peaks.

2.2.2. Preparation of aqueous MCC suspensions

The aqueous suspensions of MCC and MCC with the surfactant were prepared by first mixing the required quantity of MCC with distilled water (20 mL) with the help of magnetic stirring. The amounts of MCC added to water were 0.1 wt.%, 0.2 wt.%, 0.4 wt.%, 0.6 wt.%, 0.8 wt.%, 1.0 wt.%, 1.2 wt.%, 1.5 wt.%, 1.6 wt.%, 2.0 wt.% and 3.0 wt.%. In case of surfactant-based suspensions, 20% (on the weight of MCC) Pluronic® F-127 was added to the MCC aqueous suspensions, which were previously stored for 2 days for soaking. Magnetic stirring of the suspensions was then carried out for 10 min, followed by ultrasonication in a bath sonicator (Sonica® Ultrasonic Clear) for 15 min. The suspensions prepared without Pluronic were also processed through the same method except using the magnetic stirring process.

2.2.3. Characterisation of aqueous MCC suspensions through visual inspection, optical microscopy and UV-Vis spectroscopy

The aqueous suspensions of MCC and MCC with 20% Pluronic, after 48 h of the preparation, were characterised for sedimentation, homogeneity, agglomeration, etc. using visual inspection, optical microscopy (Olympus BH-2) and UV-Vis spectroscopy (UV-2401PC, UV-Vis Recording Spectrophotometer, Shimadzu). The optical microscopy was performed using a drop of MCC suspension taken on a glass slide and observing under different magnifications and at different locations of each suspension. Image J software was subsequently used to quantify the total area of agglomerates (%) and the average size of MCC particles in each suspension. UV-Vis spectra of different suspensions were recorded and the relationships between absorbance (at 300 nm) and MCC concentration were plotted.

2.2.4. Preparation of plain mortars and multi-scale composites and characterisation of flow behaviour

The fabrication of plain mortar samples was performed by mixing OPC (450 g), standardized sand (1350 g) and distilled water (225 mL), in the proportion of 1:3:0.5, using Hobart mixer according to NP EN 196-1:2006 standard. The mix design used for the preparation of different samples is provided in Table 2.

(Table 2)

Multi-scale cementitious composites were fabricated using the same process but replacing distilled water with MCC aqueous suspensions. The amounts of MCC added to the cement mixture were 0.1%, 0.2%, 0.4%, 0.6%, 0.8%, 1.0% and 1.5% (on the wt. of cement). The amounts of sisal fiber in multi-scale composites were 0.25% and 0.50% (on the wt. of cement). The amounts of MCC and sisal fibres were selected based on the previous literature on MCC and sisal fibre-reinforced cementitious composites (Parveen et al., 2015; Lima et al., 2014)) and some initial trials performed in the present study. Pluronic (20 wt.% of MCC) was used as the surfactant during the preparation of MCC suspensions. Samples were also prepared with 0.25%, 0.50%, 1.0% and 2.0% sisal fibres (without MCC) to study the influence of sisal fibers on the performance of composites. Also, cementitious composites using 0.1% and 1.0 wt.% MCC (without sisal fibres) were prepared with and without ultrasonication to investigate the influence of MCC concentration and ultrasonication time. Pluronic produced foam in the cement mixture and to suppress that TBP was used (50 wt.% of Pluronic) following the recommendations from authors' previous research (Parveen et al., 2015). After proper mixing, the prepared mortar pastes were poured into rectangular moulds (160mm × 40mm × 40mm) and kept for 24 h in a humid chamber according to NP EN 196-1:2006 standard. The samples were next demoulded and kept in water for 28 days to carry out the hydration process. Subsequently, the samples were taken out from water, wiped with a dry cloth, kept in the laboratory atmosphere for 2 h and then tested for flexural and compressive properties. The flow behaviour of fresh pastes of plain mortar and multi-scale composites was measured by performing the mini-slump test, according to EN1015-3:2004 standard. The flow values were measured from the diameter of mortar pastes at two perpendicular directions after standardised

vertical impacts. The influence of MCC, sisal fibres and multi-scale reinforcements on the flow behaviour of cement pastes was investigated.

2.2.5. Characterisation of mechanical performance of plain mortars and multi-scale cementitious composites

Flexural and Compressive Strength

Plain mortars and multi-scale cementitious composites were characterised for flexural and compressive properties according to BS EN 196-1:1995 standard. Cementitious composites reinforced with only sisal fibres were also characterised for flexural and compressive properties to investigate the influence of sisal fibres on the performance of composites. The flexural and compressive test setups and the samples before and after the testing are shown in Figure 1.

(Figure 1)

Fibre-Matrix Interface

Single fibre pull-out tests were performed on plain mortars and multi-scale composites to determine the adhesion between sisal fibres and the cementitious matrix. A sisal fiber (200 mm) was inserted (at 20 mm depth) into cylindrical-shaped (25.4 mm diameter and 20 mm height) plain mortars and selected cementitious composite specimens. The samples prepared for pull-out testing and the testing setup are shown in Figure 2. The samples were hydrated for 7 and 28 days. Ten samples were prepared for each type of samples. After hydration, the specimens were tested in a dynamometer (Hounsfield H10 KS) with a 2500 N load cell and at a speed of 2 mm/min. Force was applied onto the sisal fiber until it was completely removed from the cementitious matrix or ruptured and the force vs displacement was recorded.

(Figure 2)

Fracture Behaviour

The fracture energy of plain mortars and multiscale cementitious composites was calculated from the area under the flexural load–displacement curves using Origin software. Fracture energy was calculated for plain mortars, cementitious composites reinforced with sisal fibres, 0.1 wt.% MCC

+ 0.25 wt.% sisal, 1.0 wt.% MCC + 0.25 wt.% sisal, 0.1 wt.% MCC + 0.50 wt.% sisal and 1.0 wt.% MCC + 0.50 wt.% sisal samples, all having 160 mm × 40 mm × 40 mm dimensions.

2.2.6. Characterisation of hydration behaviour of plain mortars and multi-scale composites

Plain mortars and selected multi-scale cementitious composites were characterised using Thermogravimetric Analysis (TGA, Hitachi STA 7200), performed in a nitrogen atmosphere at a heating rate of 10°C/min up to 900°C, in order to study the hydration products. The quantitative estimation of different hydration products such as C-S-H, Ca(OH)₂, CaCO₃, etc. was performed using the derivative thermogravimetry (DTG) curves of the specimens. The influence of MCC, sisal fibres and the multi-scale reinforcement on the degree of cement hydration was studied from this characterisation. Plain mortars and selected multi-scale cementitious composites (0.1 wt.% MCC + 0.50 wt.% sisal and 1.0 wt.% MCC + 0.50 wt.% sisal) were characterised using XRD analysis to investigate various hydration products. XRD analysis was carried out using Bruker D8 Discover diffractometer in the angle range of 5-70° and using a step size of 0.04 with 2 s per step.

2.2.7. Characterisation of density, porosity and pore-size distribution of plain mortars and multi-scale composites

The measurement of density of plain mortars and selected multi-scale cementitious composites was carried out using pycnometry by intrusion of helium gas. The density measurement was carried out in Micromeritics equipment (model AccuPyc II 1340). The samples were taken from the fractured parts of the composites after mechanical tests (flexural and compressive). The samples were then stored in hermetically sealed plastic containers and kept in the laboratory at room temperature. Before measurement, the samples were oven dried at 60°C for 24 h and then kept in a desiccator. The pore-size distribution and porosity of plain mortars and selected multi-scale cementitious composites were analysed using mercury intrusion porosimetry (MIP) in Micromeritics AutoPore IV 9500 V1.07 instrument according to BS ISO 15901-1:2005 standard. In MIP, the pore-size distribution can be determined by forcing mercury into a sample under an increasing pressure and measuring the intruded volume of mercury as a function of pressure. A pressure range of 0.0007 to 414 MPa allowed the pore size measurement in the range from 340 µm to 5 nm. The surface tension of mercury and advancing/receding contact angle used in the

analysis were 0.485 N/m and 130°, respectively. The samples (1 cm³) were prepared after 6 months to allow complete hydration.

2.2.8. Micro-structural characterisation of plain mortars and multi-scale cementitious composites

The micro-structural characterisation of plain mortars and multi-scale cementitious composites was carried out on fractured parts of samples (fractured in mechanical testing) using Scanning Electron Microscopy (FEG-SEM, NOVA 200 Nano SEM, FEI, using the secondary electron mode and an acceleration voltage of 10 kV, after coating with a 30 nm film of Au-Pd in a high resolution 208HR Cressington sputter coater). Cementitious composites and observed MCC particles were characterised using an integrated Energy Dispersive Spectrometer (EDS), with EDAX Si(Li) detector, an acceleration voltage of 15 kV to investigate the elements present in the scanned areas of the specimens.

3. Results and discussion

3.1 Characterisation of MCC and sisal fibres

The morphology of MCC is shown in **Fig. 3(a)** and **Fig. 3(b)**. It can be observed that MCC particles had different shapes and were agglomerated. The cross-section morphology of sisal fibres (**Fig. 3c**) showed parallel hollow cells, in which the cell wall was composed of lignocellulosic materials and cellulose micro fibrils (Bai et al., 2002). The outer surface of the cells (as can be seen in Fig. 3d) had a rougher morphology and composed of lignocellulosic materials and waxy substances (Bai et al., 2002). The XRD patterns of MCC (**Fig. 4a**) showed a strong peak at 2θ -22.5° and other two weak peaks at 2θ -15° and at 34.55°, which are the characteristic peaks of cellulose I crystals (Zhao et al., 2018). The XRD patterns of sisal fibres also showed crystalline peaks of cellulose I (2θ -22.5° and 15°) (Kaushik et al., 2012). The calculated crystallinity indexes of sisal fibres and MCC were 68.2% and 85.2%, respectively.

(Figure 3)

(Figure 4)

3.2. Dispersion State of MCC in Aqueous Medium

The aqueous suspensions of MCC prepared with and without 20% Pluronic, as observed through visual inspection, are shown in **Fig 5**. It can be noticed that the suspensions, whether prepared with or without Pluronic, were clear with only slight sedimentation even at high concentrations. So, the used ultrasonication process for 15 min was found effective in dispersing MCC in water. Also, no significant difference was noticeable from this visual observation in the clarity or sedimentation between the suspensions prepared with and without Pluronic.

(Figure 5)

The findings of visual observation were further supported by UV-Vis spectroscopy results, as shown in **Fig. 6**. It is clear from **Fig. 6(c)** that the use of Pluronic had insignificant influence on the absorbance and therefore, on the concentration of dispersed MCC. The absorbance increased with the increase in MCC concentration indicating that the added MCC became well dispersed in water without significant sedimentation. However, the deviation from linearity of absorbance-concentration curves at higher concentrations was attributed to the saturation in the MCC dispersion as well as increased MCC sedimentation.

(Figure 6)

The positive influence of Pluronic was observed from the optical micrographs of prepared suspensions, as presented in **Fig. 7**. A clear difference between the suspensions prepared with and without Pluronic can be observed at 2 different magnifications. The MCC agglomeration increased with the increase in MCC concentration in both Pluronic-based and non-Pluronic suspensions. However, the MCC agglomeration was considerably lower when Pluronic was used, as can be observed qualitatively in **Fig. 7** and also from the quantitative analysis of agglomerated area and particle size presented in **Fig. 8**.

(Figure 7)

(Figure 8)

Both total agglomerated area % and the average particle size of agglomerates were significantly higher in the suspensions prepared without Pluronic. Being a non-ionic surfactant, Pluronic helped in the wetting of MCC agglomerates, breaking down the agglomerates into smaller particles (or even into individual MCC crystals when dispersed at very low concentrations) with the help of ultrasonication energy and their subsequent distribution into the whole volume of the suspensions. Previous studies also demonstrated the positive influence of Pluronic surfactants on the dispersion of micro/nano cellulose in aqueous or other mediums (Tardy et al., 2017).

3.3. Flow Characteristics of Fresh Mortar Paste

The influence of sisal fiber content on the consistence or flow properties of the fresh mortar paste is shown in **Fig. 9(a)**. It can be noticed that the increase in the sisal fibre content reduced the flow values of fresh mortar pastes significantly. By using 2 wt.% of sisal fibres, the flow values of fresh mortars decreased by ~ 15%. Similarly, as shown in **Fig. 9(b)** and **9(c)**, the flow values of fresh mortars also decreased considerably with the increase in the MCC content in presence or absence of sisal fibres. The flow values reduced by ~18% using 1wt.% MCC in the absence of sisal fibres and by ~24% with 0.25% sisal fibres. The reason for the decreased flow values of fresh mortars due to the presence of sisal fibres and MCC was their hydrophilic character which led to absorption of water from the mortar pastes reducing their flow properties (Cao et al., 2015; Hospodarova et al., 2018). MCC had slightly higher effect on the flow properties due to their higher surface area and ability to absorb water more quickly. The use of both sisal fibres and MCC as the multi-scale reinforcement had a higher effect on the flow properties over the individual components due to their combined effect. However, the ultrasonication process did not show any noticeable effect on the flow values of mortar pastes due to the fact that the water absorption of MCC was not significantly influenced by its dispersion state. **Fig. 9(d)**, **9(e)** and **9(f)** show the picture of the flow table for plain mortar pastes and pastes containing 0.25 wt.% and 2 wt.% of sisal fibres, indicating that the workability of mortar pastes reduced drastically at 2 wt.% sisal fibres. Considering a target flow value of 180 mm $\pm$ 10 mm according to EN 1015-3 standard, it can be commented that the multi-scale reinforcement containing low concentration of MCC (0.2 wt.% or less) led to a recommended level of flow values (>170 mm) (**Fig. 9 c**).

(Figure 9)

3.4. Compressive and flexural strengths of plain mortar and cementitious composites

Table 3 summarises the compressive strength of plain mortars and mortars containing different percentages of sisal fibres, MCC and both MCC and sisal fibres. It can be clearly noticed that the addition of 0.25 wt.%, 0.50 wt.%, 1.0 wt.% and 2.0 wt.% sisal fibres reduced the compressive strength of mortars by 3.5%, 6.9%, 20.8% and 38.4%, respectively.

(Table 3)

Other studies also suggested significant reductions in the compressive strength and elastic modulus of cementitious composites by adding short vegetable fibres (Lima et al., 2014). This reduction in the compressive performance of mortars was mainly attributed to the increased porosity and void formation within cementitious matrices owing to the addition of vegetable fibres (Fujiyama et al., 2014; Soto and Ramalho, 2016), inherent porosity of the fibres and their hydrophilic nature leading to reduced cement hydration (Lima et al., 2014). Interestingly, although possessing a similar hydrophilic character to sisal fibres, MCC could significantly improve the compressive strength of plain mortars, i.e. by 20.5% using 0.1 wt.% MCC, primarily due to their higher mechanical properties, higher surface area that led to a better interface with the cementitious matrix and reduced pore size of mortars owing to MCC addition (Parveen et al., 2018, 2017b). The increase in the compressive properties of cementitious composites through incorporation of MCC has been previously observed (Parveen et al., 2018, 2017b; Silva et al., 2018). It is also interesting to note that the compressive strength improved significantly even when no ultrasonication treatment was used (Silva et al., 2018), though the improvement was lower as compared to the composites prepared using ultrasonication (Parveen et al., 2018; Silva et al., 2018). Nonetheless, higher concentrations of MCC resulted in more agglomeration and consequently, reduced the compressive strength improvement (Silva et al., 2018). The multi-scale composites containing the lowest amount of MCC (i.e. 0.1 wt.%) and sisal fibres (i.e. 0.25 wt.%) resulted in an intermediate effect on the compressive strength of cementitious composites, i.e. the compressive strength improved as compared to the composites prepared using only sisal fibres, but the improvement was lower as compared to the composites reinforced with only MCC. An improvement up to 12.1% was obtained in this case. However, a further increase in the MCC concentration reduced the compressive strength considerably, resulting in ~24% reduction in the compressive strength due

to addition of 1.5% MCC. Unexpectedly, although sisal fibres reduced the compressive strength of mortars an increase in the sisal fibre content in the multi-scale composites led to an improved compressive strength, indicating the synergistic effect of multi-scale reinforcements. The use of 0.5% sisal fibres in combination with 0.1% MCC improved the compressive strength of mortars by 18.4%. The synergistic effect resulted mainly from the positive influence of MCC on the porosity of mortars, fibre/matrix interface and microstructure (discussed in detail in the following sections), which reduced the negative influence of sisal fibres and increased their crack-bridging ability.

Similar to compressive strengths, the flexural strength of composites also decreased considerably with the addition of sisal fibres (as can be seen in **Table 4**) due to the reasons discussed earlier. However, the multi-scale reinforcement (0.1 % MCC+0.25% sisal) improved the flexural strength strongly, resulting in an improvement of 30.1%. Also, similar to compressive strengths, an increase in the MCC content decreased the flexural strength of multi-scale composites significantly. However, in contrast to compressive strengths, the increase in the sisal fibre content from 0.25 wt.% to 0.5 wt.% decreased the flexural strength of multi-scale composites, leading to only 5.1% improvement as compared to plain mortars. This could be attributed to the fact that the flexural strength was affected more strongly by the increased porosity resulting from sisal fibres as compared to the compressive strength (Tolêdo Romildo D. et al., 2003). The flexural strain of mortars was also influenced strongly by the hybrid reinforcements. It is clear that the addition of sisal fibres strongly increased the flexural strain of mortars. The flexural strain of plain mortars improved by ~27% using up to 2 wt.% sisal fibres. The improvement in flexural strains of 0.25 wt.% sisal fibre-reinforced mortars reduced from ~12% to ~5% when 0.1% MCC was used in combination in the multi-scale composites. The flexural strain further reduced with the increase in the MCC content and became significantly lower as compared to that of plain mortars. The reduction of strain and ductility of cementitious (as well polymeric matrices) composites due to addition of MCC has been previously noticed by several researchers (Liu et al., 2017) and was mainly attributed to the stiffer and crystalline structure of MCC restricting the mobility of cementitious/polymeric matrices. The use of higher amount of sisal fibres (0.5 wt.%) in multi-scale composites in combination with 0.1% MCC improved the flexural strain strongly (by ~84%). In this case also, a further increase in MCC % drastically reduced the flexural strain.

(Table 4)

The flexural modulus of cementitious composites was also influenced significantly by sisal fibres and multi-scale reinforcements, as can be noticed in **Fig. 10**. The use of sisal fibres strongly reduced the flexural modulus of composites, resulting in up to 31.6% reduction using up to 0.2 wt.% sisal fibres. The presence of MCC resulted in an initial steeper slope (indicated by arrows) in the flexural-strain curves and the higher MCC content increased the brittleness of composites. The initial stiffening effect was more pronounced at low MCC concentrations combined with a high sisal fibre content (i.e. 0.5 wt.%). An increase in the MCC content (up to 1.5 wt.%) in these composites resulted in a completely MCC dominated brittle flexural behaviour (see curve for 1.5% MCC+0-5% sisal). It is clear from Fig. 10 (c) that the increase in the MCC content strongly increased the flexural modulus of composites containing higher amount of sisal fibres, i.e. 0.5 wt.%. On the contrary, when multi-scale composites contained lower amount of sisal fibres, i.e. 0.25 wt.%, the flexural modulus increased up to 0.1% MCC and then, reduced probably due to MCC agglomeration. Overall, an increase in flexural modulus up to 30% was achieved using 0.1% MCC in combination with 0.25% sisal fibres.

(Figure 10)

The flexural stress-strain curves (shown in **Fig. 11**) clearly shows a post-cracking deformation when the sisal fibre content was 0.5% or more, either used alone or in combination with MCC. The remarkable improvement of flexural strains and the post-cracking behaviour of cementitious composites due to sisal fibres were attributed to the ability of sisal fibres in bridging micro and macro-cracks developed within the cementitious matrix during deformation (Silva et al., 2009). A strong interface between sisal fibres and the cementitious matrix in multi-scale composites was evidenced from the adherence of cementitious materials on the surface of sisal fibres, as shown in **Fig. 12(a)**. The strong interface helped in holding the cracks, restricting their fast growth and preventing the catastrophic failure.

(Figure 11)

(Figure 12)

Another interesting finding of the present research was the improvement of sisal fibre-cementitious matrix interfaces due to the presence of MCC, as can be noticed from the single fibre pull-out test results presented in **Fig. 13**. The pull-out forces for MCC containing mortars were similar to plain mortars at 7 days, but increased significantly after 28 days as the hydration progressed. The improvement of the plant fibre-matrix interface due to presence of MCC in the matrix has been recently observed in the polymeric composites and it was believed that MCC acted as a strong linking material between fibres and the matrix improving the interface between them (Pichandi et al., 2018). The use of nanocellulose coating on the plant fibre surface was also found effective in improving the plant fibre-cementitious matrix interface (Mohammadkazemi et al., 2015; Pichandi et al., 2018). This was mainly attributed to the better hydration at the plant fibre surface in the presence of nanocellulose, which accelerated the growth of hydration products (Mohammadkazemi et al., 2015; Pichandi et al., 2018). As can be seen in **Fig. 12 (b)**, the observed MCC particle was well coated with the cement hydration products, as probably their growth was favoured by MCC. The EDS spectra of the MCC particle (scanned in the indicated region in Fig. 12b) showed a higher amount of C as compared to the spectra of plain mortars or multi-scale composites (**Table 5**) due to its cellulose chemistry. It also confirmed the well coating of the MCC surface with the cement hydration products, as evidenced by the presence of Ca, Si and Al. The hydroxyl groups of MCC could form covalent bonds with the hydration products, i.e. C-S-H and Ca(OH)_2 leading to strong interfacial adhesion between them, as similar to the case of graphene sheets-reinforced cementitious composites investigated earlier (Zheng et al., 2017). As MCC particles were dispersed throughout the matrix, they were also expected to be present around sisal fibres. Therefore, the improved growth of hydration products by MCC could reduce the porosity around sisal fibres and consequently, improved the contacts between sisal fibres and the cement matrix. Further, hydroxyl groups present on sisal fibres could also form chemical bonding with the surrounding hydration products as similar to MCC but at a much lower extent due to their lower surface area. Therefore, the presence of MCC in the multi-scale composites could improve the fibre-matrix interface due to improved fibre-matrix contacts and bonding, as similar to nanocellulose noticed in the previous research studies (Pichandi et al., 2018).

(Figure 13)

(Table 5)

3.5. Fracture energy of plain mortar and multi-scale composites

The influence of sisal fibres on the fracture energy of plain mortars is shown in **Fig. 14**. It can be noticed that the increase in the sisal fibre content increased the fracture energy of cementitious composites linearly. The fracture energy improved by ~ 351% using 2 wt.% sisal fibres, as listed in **Table 6**. This was attributed to the ability of sisal fibres in bridging macro and micro cracks as discussed earlier (Silva et al., 2009). MCC also improved the fracture energy of cementitious composites, but the improvement was significantly lower as compared to sisal fibres (improved by 29% with 0.1 wt.% MCC and the improvement reduced to ~12% with 1 wt.% MCC due to agglomeration). The advantage of using multi-scale reinforcements for improving the fracture energy of cementitious composites can be clearly noticed. In multi-scale composites, the combinations with low sisal fibre % or high MCC contents were not found much effective in improving the fracture energy. However, the multi-scale reinforcement containing 0.5% sisal fibres with 0.1% MCC resulted in ~100% improvement in the fracture energy. Therefore, the multi-scale reinforcement proved to be highly effective in improving the fracture energy of cementitious composites, while also enhancing flexural and compressive strengths and moduli.

(Figure 14)

(Table 6)

3.6. Micro-structure of plain mortar and multi-scale composites

The microstructure of cementitious composites changed significantly due to the addition of sisal fibres and MCC. The density of plain mortars and a selected type of multi-scale cementitious composites (with 0.1 wt.% MCC + 0.5 wt.% sisal + Pluronic), as measured using Pycnometry by intrusion of Helium gas, is listed in **Table 7**. The addition of 0.1 wt.% MCC and 0.5 wt.% sisal fibres along with Pluronic increased the density of plain mortars significantly. The favourable effects of MCC and Pluronic on the cement hydration leading to a denser microstructure have been previously reported (Parveen et al., 2017b). The short circuit diffusion mechanism of MCC that favoured the diffusion of water to the unhydrated parts of cement particles resulted in better hydration and a denser microstructure (Cao et al., 2015). In addition, due to the hydrophilic nature MCC could retain water and released slowly as the hydration progressed, forming well controlled and better hydration products (Parveen et al., 2018). At lower concentrations such as 0.1%, MCC

crystals were well dispersed without agglomeration showing no negative effect on the microstructure and density of mortars.

(Table 7)

Sisal fibres could also have a similar effect on the density of mortars owing to their similar cellulose chemistry and hydrophilic nature to MCC. However, the effect could be much lower due to significantly lower surface area as compared to MCC. Moreover, clustering of sisal fibres could also lead to increase in the porosity and consequent decrease in the density of mortars, which has been frequently observed in previous studies (Fujiyama et al., 2014; Lima et al., 2014). Pluronic showed a positive effect on the density of mortars as it could help to disperse the cement particles through the chemical functional groups present in its structure (Parveen et al., 2015). Overall, a positive effect on the density was observed in the multi-scale composites. The average pore diameter, as determined by MIP analysis (**Fig. 15**), also decreased from 44.1 nm to 37.0 nm in case of multi-scale composites.

(Figure 15)

The increase in the intruded mercury volume of mercury at 40 nm and 7 nm pore diameters in multi-scale composites (**Fig. 15**) indicated that the relative volume of small capillary pores (10-50 nm) and gel pores (0.5-10 nm) (Faure et al., 2012) increased in case of multi-scale composites. This also confirmed that a denser microstructure was resulted due to the addition of MCC and sisal fibres. A slight increase in the overall porosity (from 13.7% to 15.0%) also indicated the generation of larger pores primarily due to the presence of sisal fibres which, however, did not affect the density of mortars.

The denser microstructure of multi-scale composites was mainly attributed to better hydration and the formation of higher amount of hydration products, which was evidenced from DTG and XRD analyses. The thermal degradation and DTG curves of plain mortars and multi-scale composites are presented in **Fig. 16**. It can be observed that all samples showed a similar degradation behavior in mainly 3 stages, as explained in the DTG curves (14b). Plain mortars and multi-scale composites

showed a similar degradation behaviour, as can be observed from TGA curves, except the residual weight which decreased slightly in case of multi-scale composites containing higher MCC wt.%.

(Figure 16)

The three main degradation stages can be noticed as three peaks in the DTG curves of the samples. The first weight-loss peak resulted from the evaporation of water (50–100 °C) and also due to a portion of non-evaporable water (105–200 °C) from the C-S-H phase of cement (Parveen et al., 2015). The decomposition of calcium hydroxide Ca(OH)_2 resulted in the second DTG peak at ~ 400°C. The third weight-loss peak appeared in the temperature range of 600–780 °C and was attributed to the decomposition of calcium carbonate (Parveen et al., 2015). It can be clearly observed that the height of peak 1 was significantly higher for multi-scale composites over the plain mortar composites. This can be attributed to higher retention of evaporable water in the multi-scale composites due to presence of MCC and sisal fibres as well as higher formation of C-S-H gel, which contained a higher amount of non-evaporable water. It can be also noticed that peak 2 became significantly stronger in case of multi-scale composites, indicating a higher formation of Ca(OH)_2 . These observations confirmed the fact that the presence of MCC and sisal fibres led to formation of a higher amount of hydration products, as also evidenced in earlier studies (Parveen et al., 2017b; Silva et al., 2018). A significant difference in peak 3 was also observed between plain mortars and multi-scale composite samples. The peaks were much suppressed in case of multi-scale composites due to a lower formation of CaCO_3 . A finer microstructure in multi-scale composites possibly allowed a lower penetration of atmospheric CO_2 into the pore structure of composites and therefore, formed a lower amount of CaCO_3 (Parveen et al., 2015).

The main XRD peaks due to different hydration products of plain mortars and a multi-scale composite containing 0.1 wt.% MCC and 0.50 wt.% sisal fibres are indicated in **Fig. 17**: the peaks due to Ca(OH)_2 or portlandite phase (p), (2) quartz (q) and gypsum (g), (3) tricalcium silicate [C_3S (Ca_3SiO_5)] or alite (a) (Parveen et al., 2015). The much shaper peaks of portlandite, i.e. p phase in multi-scale composites clearly indicated a higher formation of Ca(OH)_2 and better hydration, as previously suggested by the DTG analysis.

(Figure 17)

In summary, a number of reasons could be responsible for the improved mechanical properties of developed multi-scale composites, as also reported earlier with other nano/micro particle-reinforced cementitious composites ((Han et al., 2019; Zheng et al., 2017): (1) As observed in this study, the porosity of multi-scale composites reduced significantly as compared to plain mortars due to the favourable effects of MCC such as: (a) MCC could act as the nucleating agent for the growth of hydration products resulting in a dense microstructure. MCC could also retain water and release slowly through the short circuit diffusion mechanism as the hydration progressed. This resulted in a well-controlled hydration process and the formation of superior hydration products. (b) MCC particles present around sisal fibre surfaces led to a higher formation of hydration products and consequently, reduced the porosity surrounding the sisal fibres. However, the pore-filling mechanism reported in case of other nanostructures was not expected in the present study due to the bigger size of MCC (Zheng et al., 2017). (2) The reduced porosity around sisal fibres improved their adhesion with the cementitious matrix and the adhesion could be further improved due to formation of chemical bonding between sisal fibres and the cementitious matrix. MCC present around the sisal fibres could also act as a bridging material and enhance the interfacial adhesion by forming chemical bonding between both sisal fibres and the cement matrix. The improved interfacial bonding between sisal fibres and the cementitious matrix (as observed in the pull-out tests) improved sisal fibres' crack-bridging ability and therefore, improved the strength and fracture energy of cementitious composites. (3) The formation of well-dispersed and strong MCC networks within the cementitious matrix and their good interfacial adhesion improved the elastic modulus of the composites. However, the fracture energy absorption by crack-deflection and crack-bridging mechanisms, as observed in case of other nanoparticles, was not expected in case of MCC as the cementitious composites reinforced with only MCC resulted in a drastic reduction of the fracture energy.

The main results of the present study are summarised and compared with the published results obtained with only MCC or sisal fibres in **Table 8**. It is clear that the addition of sisal fibres proved quite effective in improving the toughness of cementitious composites (Lima et al., 2014). However, their incorporation was found detrimental to the strength and modulus (Lima et al.,

2014). On the other hand, the influence of MCC on the properties of composites was exactly opposite, i.e. MCC could significantly improve the strength and modulus but at the cost of toughness (Parveen et al., 2018, 2017b). Previously, a hybrid reinforcement consisting of MCC and CNT was found effective in improving the strength, modulus and toughness of cementitious composites (Alshaghel et al., 2018), as similar to the hybrid reinforcement consisting of sisal fibres and MCC investigated in the present study. However, it can be noticed that the improvement of composites' properties achieved with this new bio-based hybrid reinforcement was significantly higher as compared to the CNT-based reinforcement, which also required a longer dispersion treatment (1 hr), is relatively expensive and has toxicity issues. The positive effects of MCC on the porosity and microstructure of cementitious composites minimised the negative effects due to sisal fibres and further improved their crack-bridging capability through an improved fibre-matrix interface, leading to an improved modulus, strength and toughness of cementitious composites. Therefore, this newly developed bio-based hybrid reinforcement can have a great potential in the construction industry for developing high performance cementitious composites.

(Table 8)

4. Conclusions

In this research, multi-scale cementitious composites were developed for the first time by using a plant-based multi-scale reinforcement containing sisal fibres and MCC. It was observed that an ultrasonic treatment for only 15 min was able to provide homogeneous MCC suspensions, which, however, showed a considerable increase in the agglomerated area and particle size with the increase in MCC concentrations from 0.1 wt.% to 3 wt.%. Although Pluronic did not have any noticeable effect on the sedimentation and dispersed concentrations of MCC, the total agglomerated area and average particle size were significantly lower with 20% Pluronic. Both Pluronic and MCC reduced the flow characteristics of fresh mortar pastes. Although the multi-scale reinforcement containing 1 wt.% MCC and 0.25 wt.% sisal fibres reduced the flow values by ~24%, the flow value was within the recommended value in case of low MCC concentrations (0.1 wt.% or less). The use of sisal fibres strongly increased the fracture energy of cementitious composites (~ 351% improvement using 2 wt.% sisal fibres), however, at the cost of compressive and flexural strengths, which decreased by 38% and 13%, respectively. On the other hand, the use of MCC could strongly improve the compressive strength (20.5% using 0.1 wt.% MCC), although

the fracture energy improved only marginally (29% using 0.1 wt.% MCC). Multi-scale reinforcements with optimised combination of sisal and MCC were found highly effective in improving the strength, modulus and fracture energy of cementitious composites without affecting the flow properties of mortar pastes. A maximum improvement of 18.4% in the compressive strength (with 0.1 wt.% MCC and 0.5 wt.% sisal), 30.1% and 30% improvements in the flexural strength and modulus (with 0.1 wt.% MCC and 0.25 wt.% sisal) and 100% in the fracture energy (with 0.1 wt.% MCC and 0.5 wt.% sisal) were obtained using the multi-scale reinforcement. The multi-scale composites exhibited an improved microstructure with a refined pore structure and a lower average pore volume as compared to the plain mortar composites. This was attributed to the formation of a higher amount of hydration products in multi-scale composites due to the positive effects (such as nucleating effect and short circuit diffusion mechanisms) on the growth of the hydration products. The lower porosity around sisal fibres in multi-scale composites as well as bridging effect of MCC through chemical bonding improved the interfacial strength between sisal fibres and the cementitious matrix. The combined effect of reduced porosity, improved fibre-matrix interface and reinforcement by MCC (improving stiffness) and sisal fibres (crack-bridging) resulted in an improved strength, modulus and fracture energy of multi-scale composites. This research showed the distinct advantages of using plant fibre-based multi-scale reinforcements in cementitious composites and in future, efforts can be directed to disperse higher amount of MCC without agglomeration and therefore, to achieve higher property improvements for developing UHPC.

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Figure Captions

Fig. 1 Samples and test setups: (a) samples before flexural testing, (b) samples after flexural testing, (c) and (d): flexural test setup, (e) compressive test setup, (f) a sample before compressive test and (g) a sample after compressive test

Fig. 2 Pull-out test samples (a) and the test setup (b)

Fig. 3 Morphology of MCC (a, b) and sisal fibre (c, d), as characterized by SEM, at different magnifications

Fig. 4 XRD patterns of MCC (a) and sisal fibre (b)

Fig. 5 Aqueous suspensions containing 0-3 wt.% MCC prepared without and with Pluronic

Fig. 6 UV-Vis curves of MCC suspensions prepared without (a) and with Pluronic (b) and absorbance-concentration relationships (c)

Fig. 7 Optical micrographs of MCC suspensions prepared without and with Pluronic

Fig. 8 Influence of MCC content and Pluronic on total area of agglomerates (a) and average particle size (b)

Fig. 9 Flow characteristics of fresh mortar paste: (a) effect of sisal fibre content, (b) effect of MCC content and ultrasonication, (c) combined effect of MCC and sisal fibres and flow table test of (d) plain mortar, (e) with 0.25 wt.% sisal fibres and (f) 2 wt.% sisal fibres

Fig. 10 Pre-peak flexural behaviour of sisal reinforced and multi-scale cementitious composites (a), effect of sisal fibre and multi-scale reinforcement on flexural modulus (b, c)

Fig. 11 Flexural stress-strain curves of plain cement mortar and cementitious composites reinforced with sisal fibers and multi-scale reinforcements

Fig. 12 Fracture surface of cementitious composites showing (a) sisal fibre and (b) MCC

Fig. 13 Pull-out force of sisal fibres from plain mortar and mortar containing MCC

Fig. 14 Influence of sisal fibres (a), MCC and multi-scale reinforcement (b) on the fracture energy of cementitious composites

Fig. 15 Intruded volume of mercury at different pore diameters in plain mortar and multi-scale composites

Fig. 16 TGA (a) and DTG (b) curves of plain mortar and multi-scale composites

Fig. 17 XRD pattern of plain mortar (a) and multi-scale cementitious composites with 0.1 wt.% MCC + 0.50 wt.% sisal + Pluronic (b)

Table Captions

Table 1 Properties of materials used for fabricating multi-scale cementitious composites

Table 2 Mix design showing quantity of different components used for preparation of plain mortar and cementitious composites

Table 3 Compressive strength of plain mortar and multi-scale cementitious composites

Table 4 Flexural strength of plain mortar and multi-scale cementitious composites

Table 5 Elements present in plain mortar, multi-scale composite and MCC particles.

Table 6 Fracture energy of plain mortar and multi-scale composites

Table 7 Density of plain mortar and a multi-scale cementitious composite

Table 8 Comparison of present results with published results using sisal fibres and MCC

Table 1

Material	Properties	Values*
MCC (Avicel® PH-101)	Particle size range	2 to 260 µm
	Sauter mean diameter	49.1 µm
	Moisture content	~ 3 wt%
	Solid density	1.54 g/cm ³
	Particle shape	larger fibrous rods to smaller irregular cuboids
Sisal fibers	Size	20 mm - short fibers, α-cellulose: 73%, hemicellulose: 10.1%, lignin: 7.6%, extractives: 6.2, ashes: 3.1% **
Ordinary Portland Cement (CEM I 42.5 R)	Product composition	95-100% clinker + 0-5% additional components
	Loss on ignition	≤ 5.0%
	Insoluble residue	≤ 5.0%
	Sulphur trioxide (SO ₃)	≤ 4.0%
	Chloride (Cl ⁻)	≤ 0.1%
	Initial setting time	≥ 60 min
	Soundness	≤ 10 mm
	2 days compressive strength	≥ 20.0 MPa
	28 days compressive strength	≥ 42.5 MPa and ≤ 62.5 MPa
Sand (CEN-EN 196-1)	Moisture content	≤ 0.2%
	SiO ₂	≥ 95%
	Particle size distribution	
	Square mesh size (mm)	Cumulative sieve residue
	2.00	0
	1.60	7 ± 5
	1.00	33 ± 5
	0.50	67 ± 5
	0.16	87 ± 5
Surfactant Pluronic® F-127	0.08	99 ± 1
	Description	Non-ionic surfactant
	Product line	Bio-reagent
	Average molecular weight	12500 g/mol
	Critical micelle conc.	950-1000 ppm
	Moisture content	≤ 0.75%

*manufacturers' data, ** source: (Sydenstricker et al., 2003)

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Table 2

Mix	Cement wt. (g)	Sand wt. (g)	Water vol. (ml)	MCC* (wt.%)	Sisal Fibres* (wt.%)
Plain mortar	450	1350	225	-	-
0.25 % sisal	450	1350	225	-	0.25
0.50 % sisal	450	1350	225	-	0.5
1.0 % sisal	450	1350	225	-	1.0
2.0 % sisal	450	1350	225	-	2.0
0.1 % MCC	450	1350	225	0.1	-
1.0 % MCC	450	1350	225	1.0	-
0.1 % MCC + 0.25 % sisal	450	1350	225	0.1	0.25
0.2 % MCC + 0.25 % sisal	450	1350	225	0.2	0.25
0.4 % MCC + 0.25 % sisal	450	1350	225	0.4	0.25
0.6 % MCC + 0.25 % sisal	450	1350	225	0.6	0.25
0.8 % MCC + 0.25 % sisal	450	1350	225	0.8	0.25
1.0 % MCC + 0.25 % sisal	450	1350	225	1.0	0.25
1.5% MCC + 0.25% sisal	450	1350	225	1.5	0.25
0.1 % MCC + 0.50 % sisal	450	1350	225	0.1	0.50
0.2 % MCC + 0.50 % sisal	450	1350	225	0.2	0.50
0.4 % MCC + 0.50 % sisal	450	1350	225	0.4	0.50
0.6 % MCC + 0.50 % sisal	450	1350	225	0.6	0.50
0.8 % MCC + 0.50 % sisal	450	1350	225	0.8	0.50
1.0 % MCC + 0.50 % sisal	450	1350	225	1.0	0.50
1.5% MCC + 0.50% sisal	450	1350	225	1.5	0.50

882 *MCC and sisal fibre wt.% are based on the weight of cement

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Table 3

Reinforcements	Samples	Compressive strength (MPa)	% Change
-	Plain mortar	42.32 ± 0.83	-
Sisal fibre	0.25 % sisal	40.83 ± 1.35	-3.5
	0.50 % sisal	39.42 ± 0.78	-6.8
	1.0 % sisal	33.51 ± 0.85	-20.8
	2.0 % sisal	26.07 ± 1.29	-38.4
MCC	0.1 % MCC without ultrasonication	47.60 ± 0.70	12.5
	0.1 % MCC with ultrasonication	50.99 ± 0.90	20.5
	1.0 % MCC without ultrasonication	46.47 ± 1.10	9.8
	1.0 % MCC with ultrasonication	49.53 ± 0.70	17.0
Multi-scale	0.1% MCC + 0.25% sisal + Pluronic	47.44 ± 0.58	12.1
	0.2% MCC + 0.25% sisal + Pluronic	42.54 ± 3.45	0.5
	0.4% MCC + 0.25% sisal + Pluronic	37.84 ± 2.26	-10.6
	0.6% MCC + 0.25% sisal + Pluronic	34.10 ± 1.42	1.9
	0.8% MCC + 0.25% sisal + Pluronic	35.45 ± 3.30	2.0
	1.0% MCC + 0.25% sisal + Pluronic	41.04 ± 0.87	-3.0
	1.5% MCC + 0.25% sisal + Pluronic	32.26 ± 2.17	-23.8
Multi-scale	0.1% MCC + 0.50% sisal + Pluronic	50.09 ± 0.86	18.4
	0.2% MCC + 0.50% sisal + Pluronic	37.44 ± 2.69	-11.5
	0.4% MCC + 0.50% sisal + Pluronic	33.03 ± 2.94	-22.0
	0.6% MCC + 0.50% sisal + Pluronic	37.32 ± 1.10	-11.8
	0.8% MCC + 0.50% sisal + Pluronic	41.94 ± 2.37	-0.9
	1.0% MCC + 0.50% sisal + Pluronic	41.12 ± 0.77	-2.8
	1.5% MCC + 0.50% sisal + Pluronic	44.38 ± 1.42	4.8

Table 4

Samples	Flexural strength (MPa)	% Change	Strain	% Change
Plain mortar	6.64 ± 0.13	-	0.0045 ± 0.0002	-
0.25 % sisal	6.27 ± 0.73	-5.6	0.0050 ± 0.0006	11.8
0.50 % sisal	6.09 ± 0.45	-8.3	0.0057 ± 0.0006	27.3
1.0 % sisal	5.54 ± 0.83	-16.6	0.0055 ± 0.0006	22.2
2.0 % sisal	5.79 ± 0.33	-12.8	0.0054 ± 0.0005	19.3
0.1% MCC + 0.25% sisal	8.64 ± 0.88	30.1	0.0047 ± 0.0014	5.2
0.2% MCC + 0.25% sisal	7.52 ± 0.43	13.3	0.0046 ± 0.0003	3.9
0.4% MCC + 0.25% sisal	6.95 ± 0.73	4.6	0.0039 ± 0.0001	-11.4
0.6% MCC + 0.25% sisal	5.51 ± 0.51	- 17.0	0.0032 ± 0.0003	-26.8
0.8% MCC + 0.25% sisal	5.07 ± 0.31	- 23.7	0.0047 ± 0.0003	5.1
1.0% MCC + 0.25% sisal	5.16 ± 0.49	- 22.4	0.0046 ± 0.0004	3.9
1.5% MCC + 0.25% sisal	5.67 ± 0.25	- 14.6	0.0046 ± 0.0009	3.8
0.1% MCC + 0.50% sisal	6.98 ± 0.37	5.1	0.0082 ± 0.0015	84.3
0.2% MCC + 0.50% sisal	6.65 ± 0.47	0.1	0.0069 ± 0.0010	54.5
0.4% MCC + 0.50% sisal	6.35 ± 0.27	- 4.4	0.0058 ± 0.0002	29.4
0.6% MCC + 0.50% sisal	6.36 ± 0.63	- 4.2	0.0052 ± 0.0002	16.9
0.8% MCC + 0.50% sisal	6.25 ± 0.88	- 5.9	0.0025 ± 0.0003	-42.9
1.0% MCC + 0.50% sisal	4.99 ± 0.64	- 24.9	0.0023 ± 0.0003	-48.0
1.5% MCC + 0.50% sisal	4.99 ± 0.09	- 24.9	0.0021 ± 0.0007	-52.5

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Table 5

Samples	Elements (Wt.%)								
	C	O	Na	Mg	Al	Si	S	K	Ca
Plain mortar	3.69	46.62	-	0.63	1.31	10.09	1.11	1.20	38.34
Multi-scale composite	8.85	40.44	0.24	0.32	0.92	13.51	0.38	1.30	34.13
MCC particle	39.64	27.22	-	-	2.08	7.43	-	-	23.62

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Table 6

Samples	Fracture energy (J)	% Change
Plain mortar	0.26 ± 0.02	-
0.25 % sisal	0.27 ± 0.07	2.1
0.50 % sisal	0.51 ± 0.18	92.4
1.0 % sisal	0.69 ± 0.16	158.5
2.0 % sisal	1.20 ± 0.18	351.3
0.1 % MCC	0.34 ± 0.01	29.0
1.0 % MCC	0.30 ± 0.04	11.6
0.1 % MCC + 0.25 % sisal + Pluronic	0.25 ± 0.03	-7.5
1.0 % MCC + 0.25 % sisal + Pluronic	0.18 ± 0.02	-32.9
0.1 % MCC + 0.50 % sisal + Pluronic	0.53 ± 0.29	100.4
1.0 % MCC + 0.50 % sisal + Pluronic	0.33 ± 0.12	25.1

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Table 7

Samples	Density (kg/m³)	Average pore diameter (nm)	Porosity (%)
Plain mortar	2395.4	44.1	13.7
0.1% MCC + 0.5% sisal + Pluronic	2426.4	37.0	15.0

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Table 8

Research study	Reinforcements	Change in properties
(Lima et al., 2014)	25 mm, 3 vol.% sisal fibre added to the cement mixture.	3% sisal fibres reduced the peak compressive stress by about 25% (from 51.6 MPa to 37.5 MPa), elastic modulus by about 20% (from 25.9 GPa to 21.1 GPa) and toughness index by 80% (0.48 to 0.86).
(Parveen et al., 2018)	1 wt.% MCC dispersed using 30 min ultrasonication and added to the cement mixture	Flexural modulus, flexural strength and compressive strength improved by 96% (from 15.0 GPa to 29.4 GPa), 19.2% (7.2 MPa to 8.6 MPa) and 51.4% (35.6 MPa to 53.9 MPa). Drastic reduction in the fracture energy (from 804 Nm to 244 Nm).
(Parveen et al., 2017b)	0.5 wt.% MCC dispersed using 15 min ultrasonication and Pluronic F-127.	Flexural modulus, flexural strength and compressive strength improved by 106% (15.0 GPa to 30.9 GPa), 31% (from 7.2 MPa to 9.4 MPa) and 66% (from 35.6 MPa to 59.2 MPa), respectively. Drastic reduction in Fracture energy (804 Nm to 218 Nm).
(Alshaghel et al., 2018)	0.5 wt % MCC and 0.3 wt % MWCNT, dispersed using 1 h sonication and Pluronic	16.3% (from 30.4 MPa to 35.4 MPa), 12.3% (from 7.1 MPa to 8.1 MPa) and 24.1% (from 14.3 GPa to 17.7 GPa) increase in compressive strength, flexural strength and flexural modulus, respectively. 62% (from 0.26 J to 0.42 J) increase in fracture energy.
Present study	0.1% MCC and 0.25-0.5% sisal fibres, dispersed using 15 min ultrasonication and Pluronic	18.4% increase in compressive strength (from 42.3 MPa to 50.1 MPa), 30.1% increase in flexural strength (6.6 MPa to 8.6 MPa), 30% increase in flexural modulus (from 15.4 GPa to 20.0 GPa) and 100% increase in fracture energy (0.26 J to 0.53 J)

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Figure 1.

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(a)

(b)

Figure 2.

(a)

(b)

(c)

(d)

Figure 3.

Figure 4.

Figure 5.

Figure 6.

Figure 7.

Figure 8.

Figure 9.

Figure 10.

Figure 11

Figure 12.

Figure 13.

Figure 14.

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Figure 15.

Figure 16.

Figure 17.