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A facile approach of developing micro crystalline cellulose reinforced cementitious composites with improved microstructure and mechanical performance

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ABSTRACT

In the present study, microcrystalline cellulose (MCC) reinforced cementitious composites have been developed using a short and less energy intensive physical dispersion technique. MCC–cement mortar specimens were prepared through addition of aqueous MCC suspensions to the cement-sand mixture. Aqueous MCC suspensions (0.4%, 0.8%, 1.2%, 1.6% and 2% MCC, by weight) were prepared through magnetic stirring of pre-soaked MCC powder for only 45 min. The flow behaviour of freshly prepared MCC-mortar paste as well as bulk density, mechanical performance, microstructure, porosity, water uptake and hydration products of developed cementitious composites were characterized. It was noted that with the increase of MCC content, the flow of mortar paste decreased significantly. Maximum improvements of 20.5% in flexural strength, 19.8% in compressive strength, 100% in flexural modulus and 27.2% in fracture energy were achieved after 28 days of hydration. Mechanical performance was found to be better at lower MCC concentrations and at early hydration days. The addition of MCC significantly reduced the pore size of cementitious matrix, leading to increased dry bulk density and reduced water uptake as compared to the plain mortar specimens.

1. Introduction

Concrete is a widely used construction material to develop infrastructures worldwide. Although concrete is frequently used in construction industries, owing to its lower tensile strength and quasi-brittle behaviour, reinforcements are used to improve tensile strength and fracture behaviour. The reinforcements are generally steel rebars and fibres ranging from millimetre to micrometer scale. Recently, the use of nanometer scale reinforcements in cementitious matrix is receiving much encouragement in order to arrest the initial propagation of cracks at nanoscale [1–3]. However, the efficiency of such reinforcements depends on many factors, namely their mechanical properties, length, orientation, interface with the matrix, dispersion, and most importantly, the influence of reinforcements on the hydration behaviour of cement [1–3].

Nowadays, sustainability has become one of the primary concerns in construction industry. The importance of less energy consuming processes is gaining importance to reduce global warming and environmental pollution. Properties like biodegradability and renewability have become the essential demands in consumer and engineering sectors, paving the way for developing sustainable process and materials [4]. Plant based fibres, which were earlier replaced by steel rebars for the reinforcement of cement matrix, are again coming back into the scenario due to high energy consumption and associated high CO_2 emission during steel production. Although plant fibres are getting importance as reinforcement since early 1970´s, a number of demerits restricted their extensive use such as: difficulty in prediction of plant fibre composite performance due to variability of their properties, high moisture absorption and swelling, poor interface with matrix and inferior resistance to temperature, fire and environmental conditions shortening the life span of reinforced structures [5–9].

New technologies like nanotechnology are emerging to overcome these problems through modification and tailoring of materials at nano-scale. It has been possible to extract nano/micro dimensional cellulosic structures from plant fibres, which are more robust as well as less affected by degradation than the original plant fibres. Nano crystalline cellulose (NCC), nano fibrillated cellulose (NFC), micro crystalline cellulose (MCC), micro fibrillated cellulose (MFC) and bacterial cellulose (BC) are different types of cellulose nano and micro structures

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Table 1
Properties of CPV-ARI NBR 5733: 1991 cement.

Physical properties[a]	Residue in the sieve 75 μm: 6.00%, specific area: 300m^2/kg
Compressive strength (MPa)[a]	14, 24 and 34 at 1, 3 and 7 days, respectively
Chemical composition[a]	
Oxide composition	% by mass
SiO_2	18.91
Al_2O_3	4.35
Fe_2O_3	2.69
CaO	60.59
MgO	4.74
SO_3	2.86
LOI	2.89
Alkalies (Na^2O equivalent)	0.62

[a] Source: www.cimentoitambe.com.br.

with interesting mechanical properties, and are therefore finding numerous applications in food, cosmetics, medical and hygiene products, emulsions, etc. [10–15]. These micro/nano materials have been already used for reinforcement of polymeric matrices [15] and recently, are also being explored for cementitious composites. For example, the flexural strength and compressive strengths of cementitious composites improved up to 30% and 42–45%, respectively through reinforcement with NCC obtained from acid hydrolysis of plant cellulose [16, 17]. NCC particles remain well dispersed in suspensions obtained from acid hydrolysis due to high surface charge and zeta potential and therefore, resulted in strong improvement in the mechanical properties of cementitious matrix. NFC was also used to reinforce cementitious composites and was found to significantly enhance the yield stress of cement slurry, degree of hydration and mechanical properties (up to 20.7% improvement in flexural strength using acid hydrolyzed NFC and 15% and 20% improvements in flexural and compressive strengths, respectively using TEMPO oxidized NFC) [18, 19]. The use of BC coating on plant fibres to improve interface with cementitious matrix and to reduce fibre mineralization has also been reported [20].

In contrary to nano cellulose, less research has been conducted on MCC for reinforcement of cementitious composites, although it is commercially available and easier to apply in the current construction industry. In a previous research, commercially available MCC powder was saturated with water and added to the cement mixture for developing cementitious composites. However, MCC addition through this process could not improve the mechanical performance owing to inhomogeneous MCC dispersion within cementitious matrix [21]. The dispersion of MCC and other cellulosic nanomaterials can be improved through chemical functionalization with various groups such as carboxylic, ester, silane, acetyl, etc. [22–24]. Improved MCC dispersion through surface modification with tetraethyl orthosilicate (TEOS) led to ~ 60% and 45% improvements in flexural and compressive strengths, respectively [25] The use of non-ionic surfactant was also found beneficial in the authors' previous research to improve MCC dispersion within cementitious composites and achieve strong improvement in mechanical performance [26]. The present work, however, focused on developing a simple and less energy intensive physical technique for achieving homogeneous MCC dispersion, as the above mentioned chemical techniques are associated with environmental problems, time consuming and not suitable for the construction industry in the current scenario. In the present research, commercially available MCC powder has been dispersed using magnetic stirring process and the suspensions were then added to the cement-sand mixture to develop cementitious composites. The bulk density, mechanical performance, porosity, water uptake, and hydration products of the resulting composites were characterized in order to find the optimum MCC content for achieving maximum property enhancements.

2. Experimental

2.1. Raw materials

MCC (Avicel® PH-101 grade) was purchased from Sigma Aldrich (Portugal). The Ordinary Portland cement, CPV-ARI (corresponding International standard: ASTM-C150, Type V) and sand, ABNT NBR 7214: 1982 (corresponding International Standard: EN196–1) used in this study were supplied by Cauê – InterCementBrasil and IPT (Technological Research Institute), respectively. Important properties of cement are listed in Table 1. The particle size distribution of cement and sand particles are presented in Fig. 1. The scanning electron microscopy (SEM) [performed using FEG-SEM, NOVA 200 Nano SEM, FEI equipment using secondary electron mode, acceleration voltage of 10 kV and after coating with 30 nm Au—Pd film] image of raw MCC presents clustered structures, as shown in Fig. 2. The particle size range of MCC was between 2 and 260 μm with mean diameter of 49.1 μm. The MCC crystals had variable shapes starting from larger fibrous rods to smaller irregular cuboids. The fibrous MCC crystals have a rod shaped morphology with average length of 137 μm, diameter of ~ 28 μm and aspect ratio of ~ 5 (determined through image analysis). The crystallinity of used MCC is approximately 92% [27]. The MCC powder had a solid density of 1.54 g/cc and moisture content of ~3 wt%. Due to presence of —OH groups on MCC surface, it formed hydrogen bonding with neighbouring molecules having oxygen atoms and led to formation of agglomerations or clusters. Thermogravimetric and DSC analyses (using Hitachi equipment at heating rate of 10 °C/min up to 600 °C) performed on raw MCC showed significantly lower quantity of lignin and hemicellulose when compared with the plant sources, as shown in

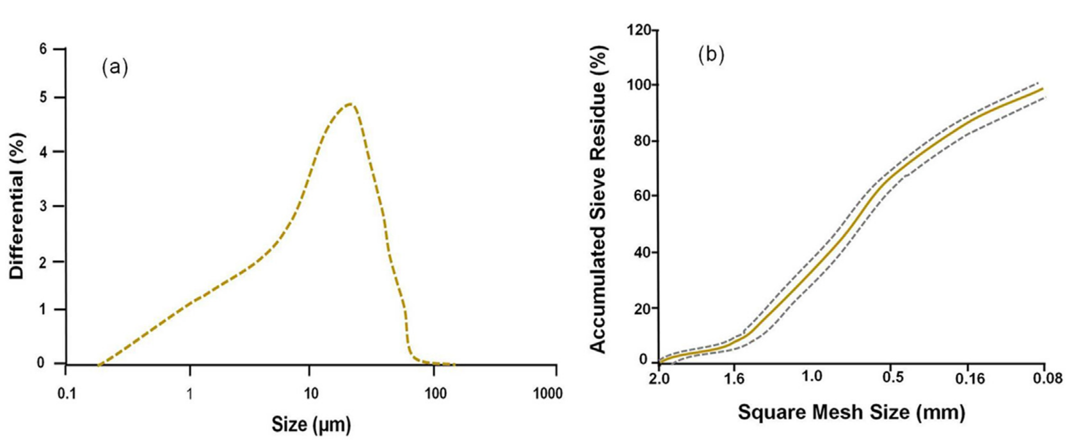

Fig. 1. Particle size distribution of cement (a) and sand particles (b) according to CPV-ARI and ABNT NBR 7214: 1982 standard, respectively.