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ORIGINAL ARTICLE

An investigation on the impact of combined microfiber-microcrystalline cellulose addition on the performance of Portland cement composites

Uma investigação sobre o impacto da adição combinada de microfibra de celulose e microcelulose cristalina no desempenho dos compósitos de cimento Portland

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Abstract: The study of the effects of cellulosic materials as additives in matrices based on mineral binders is essential for the development of high-performance and more durable construction materials. In this context, the present research aims to propose composite systems incorporating cellulose microparticles and microfibers into cementitious matrices. The proposed systems were developed using CP V ARI cement, with cellulose microfiber (FC) contents of 0.5%, 1%, and 1.5%, along with crystalline microcellulose (MCC) contents of 0.4%, 0.6%, and 0.8%. The impact of cellulose microfiber and crystalline microcellulose on compressive strength, flexural tensile strength, mineralogy, and microstructure of cementitious composites was evaluated. The gradual increase in the combined additions of cellulose microfiber and crystalline microcellulose led to a reduction in mechanical properties. The diffraction patterns of the FC-MCC cellulose-added composites were similar to those of Portland cement composites without additives. The combinations of FC 0.5-MCC 0.4, FC 1.0-MCC 0.4, and FC 0.5-MCC 0.6 contents promoted a higher degree of hydration, resulting in superior compressive strength performance compared to cementitious composites without these materials.

Keywords: cement composite, cellulose microfiber, crystalline microcellulose.

Resumo: O estudo dos efeitos de materiais celulósicos como adição em matrizes à base de aglomerantes minerais é essencial para o desenvolvimento de materiais de construção de alto desempenho e maior durabilidade. Nesse contexto, a presente pesquisa tem como propor sistemas compostos por micropartículas e microfibras de celulose integradas em matrizes cimentícias. Os sistemas combinados são propostos com o sistema cimentício tipo CP V ARI e teores de microfibra de celulose (FC) (0,5%, 1% e 1,5%), juntamente com estes, teores de microcelulose cristalina (MCC) (0,4%, 0,6% e 0,8%). Foi avaliado o impacto da microfibra de celulose e da microcelulose cristalina na resistência à compressão e tração na flexão, mineralogia e microestrutura de compósitos cimentícios. O aumento gradual nas adições combinadas de microfibra de celulose e microcelulose cristalina levou a uma redução nas propriedades mecânicas. Os perfis dos difratogramas com adição de celuloses FC-MCC foram similares aos dos compósitos de cimento Portland sem adições. As combinações de teores FC 0,5-MCC 0,4, FC 1,0-MCC 0,4 e FC 0,5-MCC 0,6 favoreceram um maior grau de hidratação, o que resultou em um desempenho mecânico superior em compressão, comparado aos compósitos cimentícios sem a adição desses materiais.

Palavras-chave: compósito cimentício, celulose microfibra, microcelulose cristalina.

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1 INTRODUCTION

Cementitious systems are widely employed in most applications of structural and non-structural building materials due to their high compressive strength. However, their use can be limited due to the formation of microcracks from the molding phase, potentially leading to cracks in cementitious systems [1]–[3]. In this context, the development of new cementitious materials with improved properties derived from renewable raw materials is considered highly advantageous.

Cellulose fiber is a natural material widely used in forest products, textiles, and pharmaceuticals, among other industries [4]. With the increasing demand for high-performance materials, a generation of cellulosic products has emerged, based on the extraction of cellulose fiber on a micro/nanometric scale [5].

The advantages of cellulose microfiber include its low density and wide availability [6]–[10]. The mechanism behind the improvement of cellulose microfibers in cementitious materials can be attributed to their high hydrophilic and hygroscopic characteristics, resulting in increased chemical reactivity and the formation of highly dense calcium-silicate-hydrate (C-S-H) gel [11]–[15]. However, the performance of cellulose microfibers is affected by the formulation of cement pastes, curing conditions, and the microfiber itself [9]–[16]. Table 1 shows the effect of percentages and lengths of cellulose fibers and microfibers on the properties of cementitious systems.

Table 1 – Summary of the percentages and lengths of cellulose fiber and microfiber used in cementitious materials

Authors	Percentage of the mass of cement	Cementitious material	Length	Effect on physical and mechanical properties
Bezerra et al. [17]	1% to 3%	Cementitious composite	0.47 mm	Percentages exceeding 2% did not show improvement in compressive strength
Palomar et al. [18]	0.8%	Rendering mortar	1 mm	Reduction of pores in the mortar
Kawashima and Shad [19]	1%	Cementitious composite	2.1 mm	13% reduction in autogenous shrinkage
Claramunt et al. [20]	2%	Cementitious composite	6 cm	Higher modulus of elasticity (22%)
Hwang et al. [21]	1%	Cementitious composite	17 mm	42.3% increase in the modulus of rupture
Xie et al. [22]	4.8%, 12% and 16%	Cementitious composite	2.5 mm	Increase in fibers promoted agglomeration of the composites
Sabarish et al. [23]	2%	Concrete	19 mm	Improvement in toughness
Gwon et al. [24]	0.3%	Cementitious composite	5 mm	4.3% increase in compressive strength
Gwon et al. [25]	0.3% and 0.6%	Cementitious composite	400 μm	85% increase in compressive strength
Pescarollo et al. [26]	0.5%, 0.3% and 0.1%	Rendering mortar	500-45 μm	Facilitated fluidity through the lubrication mechanism of cement grains

Cellulose particles can be obtained through different processes, potentially produced biotechnologically by bacteria (bacterial nano/microcelluloses), through mechanical processes (nano/microfibrils of cellulose), or via chemical hydrolysis treatments (nano/microcrystals of cellulose) [27]–[29]. Nano and microcellulose materials offer numerous advantages, including a high specific surface area about volume, which can increase packing density. This contributes to the refinement of the matrix pores, thereby improving the properties of cementitious systems [8], [24], [30], [31]. Table 2 presents the influence of crystalline microcellulose percentages on the properties of cementitious composites.

Table 2 – Summary of the percentages of crystalline microcellulose used in cementitious materials

Authors	Percentage about the mass of cement	Cementitious material	Effect on mechanical properties
Silva et al. [32]	0.4%	Cementitious composite	20.5% increase in flexural strength and 19.8% increase in compressive strength
Moraes et al. [33]	0.2%	Cementitious mortar	6% increase in flexural tensile strength
Lisboa et al. [34]	0.2%	Cementitious paste	8.06% increase in the modulus of rupture
Parveen et al. [35]	0.6%	Cementitious composite	Increase of 31% in flexural strength and 66% in compressive strength
	1%		Increase of 19.2% in flexural strength and 51.4% in compressive strength
Souza et al. [36]	0.75%	Cementitious composite	Fivefold increase in flexural strength compared to the reference
Alshaghel et al. [37]	0.5%	Cementitious composite	12.3% increase in flexural strength and 23.25% increase in compressive strength

Cellulose fibers and microfibers may have a negative effect on the hydration process of cement composites, which can lead to a delay in the setting and hardening time. This occurs due to the presence of water-soluble sugars that result in an alkaline hydrolysis of lignin and partial solubilization of hemicellulose contained in these fibers [38].

Based on research related to the addition of cellulose microfiber and crystalline microcellulose in cementitious systems, a systematic review was conducted using the *Methodi Ordinatio* [39] on the addressed topic. Through this tool, the points of innovation in this research were identified. The combinations of keywords are presented in Table 3.

Table 3 – Systematic review axes

Axes	Database		Total
	SCIENCE DIRECT	SCIELO	
	Total articles found		
“Microfiber cellulose” AND “Cellulose microcrystalline” AND “Cement paste”	0	0	0
“Fiber cellulose” AND “Cellulose microcrystalline” AND “Cement paste”	0	0	0
“Fiber” AND “Cellulose microcrystalline” AND “Cement paste”	6	1	7
“Microfiber cellulose” AND “Microcellulose” AND “Cement paste”	0	0	0
“Fiber cellulose” AND “Microcellulose” AND “Cement paste”	0	0	0
“Fiber” AND “Microcellulose” AND “Cement paste”		0	
“Fiber cellulose” AND “Nanocellulose” AND “Cement paste”	7	0	7
“Fiber” AND “Nanocellulose” AND “Cement paste”	93	0	93

The filtering procedure, as shown in Table 4, was carried out in order to harmonize the results obtained from the databases. This was done by individually applying the following filters: eliminating duplicate articles in the databases (easily done using the reference manager) and eliminating articles belonging to book chapters or conferences (they do not have an impact factor). Finally, the articles were analyzed to verify whether or not they were related to the topic.

Table 4 – Filtering procedure for the studied articles

Filtering procedures used	Selected articles	Excluded articles	(%)
Gross total of articles	107		100
Book chapter/conference articles		33	30,84
Article duplication		10	9,35
Number of articles excluded after reading		60	56,1

The survey was conducted on the Science Direct and Scielo databases. The final database consisted of 4 studies, as shown in Table 5.

Table 5 – Results of the articles obtained through the *Methodi Ordinatio*

Ranking	Authors	Citations	Year
1º	A. Balea, E. Fuente, A. Blanco, and C. Negro. Nanocelluloses: Natural-Based Materials for Fiber-Reinforced cement composites. A critical Review [40]	91	2019
2º	A. Souza Fo., S. Parveen, S. Rana, R. Vanderlei, and R. Fangueiro. Mechanical and micro-structural investigation of multi-scale cementitious composites developed using sisal fibers and microcrystalline cellulose [41]	15	2021
3º	F. Mohammadkazemi, R. Aguiar, and N. Cordeiro. Improvement of bagasse fiber-cement composites by addition of bacterial nanocellulose: an inverse gas chromatography study [42]	30	2017
4º	L. J. Capelin, K. K. Moraes, J. P. Zampieri, and R. D. Vanderlei. Evaluation of the effects of coconut fiber and microcrystalline on the properties of cementitious mortars [43]	9	2020

Based on the results of the systematic review, it was observed that the combined assessment of cellulose microfiber and crystalline microcellulose has not yet been studied in cementitious systems. However, articles combining cellulosic materials were identified [41]–[43].

Mohammadkazemi et al. [42] developed cementitious composites combining bacterial nanocelluloses with sugarcane bagasse fiber. It was observed that nanocellulose particles inhibit the penetration of alkaline hydration products into the fiber lumen, thereby preventing embrittlement and improving the durability of the composites.

Cellulose fibers have a high tendency to form hydrogen bonds, similar to cellulose microfibers. Furthermore, nano or microcelluloses particles adsorb to the fiber surface through hydrogen bonding, causing the fibers to be coated. Coating fibers with nano and microcelluloses increases the specific surface area and available reactive groups [42].

Souza et al. [41] evaluated multiscale cementitious composites reinforced with crystalline microcellulose and sisal fiber. Aqueous suspensions of MCC were added to the cement and sand mixture along with sisal fibers. They concluded that the mechanical strengths in compression and flexure improved by 24% and 18%, respectively, using 0.1% MCC and 0.5% sisal fiber compared to the reference.

Balea et al. [40], in their review work, suggest, although not experimentally proven, that low levels of micro or nanocellulose combined with fibers can lead to a superior quality product by improving compressive and flexural strength. They further explain that porosity negatively impacts the material's durability during wet/dry aging cycles by facilitating water ingress into the matrix. This leads to the dissolution of hydration products, particularly calcium hydroxide, which reprecipitates during the drying phase as the water evaporates. The repeated precipitation contributes to the mineralization of fibers, compromising the material's integrity.

According to Balea et al. [40], an alternative to improve the interaction between the brittle matrix and cellulose fibers, thus enhancing the fibers' or microfibers' capabilities, is to employ cellulose at the micro or nanoscale level. The incorporation of bacterial nanocelluloses can make the surface more reactive, increasing the dispersive component of the surface energy of cementitious systems with the addition of sugarcane bagasse fibers with a length of 1.13 mm [42].

The advantages of using micro and nanocelluloses include better adhesion to the fiber-matrix interface and higher mechanical properties, whereby the specific surface area and available reactive groups are increased [40]. The alkaline matrix hydrolyzes a portion of the cellulose, generating organic acids and non-acidic byproducts, which release energy to accelerate the hydration reaction. It was observed that while cellulose fibers tend to decrease the hydration rate, the incorporation of micro or nanocelluloses enhances the hydration reactions [42]–[45].

In the study by Capelin et al. [43], the influence of additions of coconut fiber and crystalline microcellulose in cementitious mortars was evaluated. They concluded that combinations of 0.2% coconut fiber with 0.3% crystalline microcellulose show a significant improvement in flexural tensile strength (an increase of 50.93%).

In this context, a promising solution for cementitious composites should be addressed through multi-scale addition [37]. Therefore, obtaining detailed information on how the interaction between cellulose microfiber and crystalline microcellulose affects properties over time in cementitious systems is crucial. This information is essential for formulating cementitious construction materials that achieve the desired performance.

2 MATERIALS

Two different cellulosic materials, cellulose microfiber, and crystalline microcellulose, were used as a combined addition in Portland cement composites (The CPV ARI type equivalent to type III according to ASTM C 150-07) [46]. The oxide content and chemical characterization of the cement used in the research are shown in Table 6.

Table 6 - Ordinary Portland cement oxide contents and chemical description

Components	Percent by mass (%)
SiO_2	19.12
Al_2O_3	4.51
Fe_2O_3	2.88
CaO	62.72
MgO	2.82
SO_3	2.62
Loss on Ignition	3.31
Free Lime	1.43
Insoluble Residue	0.67
Equivalent Alkali (NaEq%)	0.73
Blaine Fineness (cm^2/g)	4.302

The celluloses used in the research are comercial products, and the production process was provided by the manufacturer. The production of cellulose microfiber was carried out through the reprocessing of industrial paper waste. The material underwent an initial grinding stage, followed by separation using air separators and magnetic filtration to remove heavy particles. It was then moistened to form a pulp. After this stage, the pulp was washed, screened, and oven-dried. Subsequently, it went through another separation process and was ground until it reached a size of 400 micrometers. In the final stage of production, calcium carbonate was added to improve dispersion and chemical compatibility with Portland cement. Table 7 shows the average size and density of FC-MCC celluloses.

Table 7 - Density and average size of FC-MCC celluloses

Samples	Apparent density ($g/cm^{3)}$	Average length	Average diameter	Aspect ratio
Microfiber cellulose (FC)	1,1 – 1,3	400 µm	45 µm	8,88
Cellulose microcrystalline (MCC)	0,35 – 0,46	65 µm	15 µm	4,33

Crystalline microcellulose has a diameter in the micrometer range, being finer than the cement used in the study, and may play an active role in the hydration kinetics of the cementitious system. On the other hand, cellulose microfibers have an average diameter of 0.4 mm and may act as micro-reinforcement for the cementitious system. Figure 1 shows the X-ray diffractograms of FC-MCC celluloses.

The XRD analysis revealed that the samples exhibited peaks around $2\theta = 14.5°$, 22°, and 34°, which are associated with the cellulose I crystalline component. The microcrystalline cellulose displayed a typical cellulose I diffractogram, with peaks in the amorphous region ($18° \leq 2\theta \leq 19°$) and a prominent peak in the crystalline region ($22° \leq 2\theta \leq 23°$) [47]. The XRD results for the cellulose microfibers showed the three characteristic peaks of cellulose I crystalline ($2\theta = 15°$, 22°, and 35°). Additionally, compounds such as quartz, calcium carbonate, tricalcium aluminate, and tetracalcium ferroaluminate, found in the samples, were absent in native cellulose fibers. This corroborates the X-ray fluorescence analysis, which suggests that the cellulose microfibers were subjected to chemical treatment.

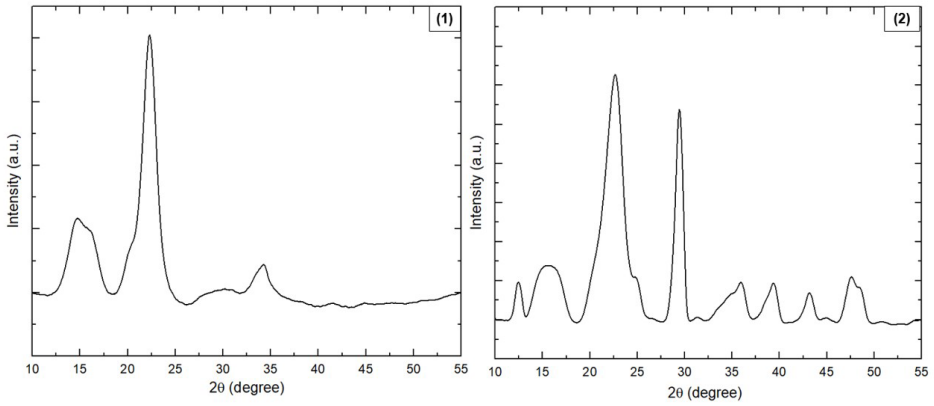

Figure 1 – XRD results of crystalline microcellulose (1) and cellulose microfiber (2) Legend: Fe = Brownmillerite; Ce = Cellulose; Ca = Calcite; H = C-S-H; Q = Quartz.

Cementitious composites incorporating crystalline microcellulose and cellulose microfibers were produced using various combined formulations. The composition of the studied composites is shown in Table 8.

Table 8. Mix design concerning the quantity of different components used for the preparation of cement paste

Samples	Cement Type CP V ARI (g)	Water/Cement	FC (g)	MCC (g)
Reference	200	0.45	-	-
FC (0.5)	200	0.45	1.0	-
FC (1.0)	200	0.45	2.0	-
FC (1.5)	200	0.45	3.0	-
MCC (0.4)	200	0.45	-	0.8
MCC (0.6)	200	0.45	-	1.2
MCC (0.8)	200	0.45	-	1.6
FC (0.5) + MCC (0.4)	200	0.45	1.0	0.8
FC (0.5) + MCC (0.6)	200	0.45	1.0	1.2
FC (0.5) + MCC (0.8)	200	0.45	1.0	1.6
FC (1.0) + MCC (0.4)	200	0.45	2.0	0.8
FC (1.0) + MCC (0.6)	200	0.45	2.0	1.2
FC (1.0) + MCC (0.8)	200	0.45	2.0	1.6
FC (1.5) + MCC (0.4)	200	0.45	3.0	0.8
FC (1.5) + MCC (0.6)	200	0.45	3.0	1.2
FC (1.5) + MCC (0.8)	200	0.45	3.0	1.6

This work focused on developing a simpler and less intensive technique to achieve homogeneous dispersion of MCC, as per the methodology adopted by Bilcati et al. [48]. The dispersion process of crystalline microcelluloses involved adding selected amounts of MCCs to water, followed by hydrating the microparticles for 24 hours. After this step, the solution was manually stirred for 5 minutes and then immediately added to the dry materials in the mortar mixer for the production of cementitious composites.

The preparation of the cementitious composites began with mixing the dry materials (cement and cellulose microfibers) in plastic bags until achieving adequate homogeneity. Once the dry materials were fully homogenized and the MCCs dispersion in water was complete, the dry materials were transferred to the mortar mixer. The aqueous solution was then gradually incorporated into the mixture and homogenized for 60 seconds at low speed. Finally, the equipment was turned off to allow scraping of the container with a spatula, and the mixing was continued for an additional 60 seconds at low speed.

3 METHODS

3.1 Fresh state density and air entrained

The physical indices analyzed included fresh-state density and theoretical air content (in accordance with ASTM C185-20 [49]). Theoretical parameters were determined by relating the material densities, excluding voids, to the fresh-state density in order to calculate the theoretical incorporated air content.

3.2 Mechanical tests

The compression strength and flexural tensile strength tests were performed according to ASTM C348-21 [50] and ASTM C349-18 [51], using 4x4x16 cm molds. The curing procedure was carried out by immersion in alkaline water, that is, water saturated with calcium hydroxide (2g $Ca(OH)_2$ per liter of water). After curing, the cement pastes were placed in an internal chamber at room temperature for cement maturation. The tests were conducted using a hydraulic test machine with a 1000 kN capacity.

To assess the influence of treatments, time, and their interaction, assuming the null hypothesis that the means among treatments, different times, and the interaction were all equal, analysis of variance (ANOVA) was applied. To evaluate the equality of means among treatments over time, repeated-measures ANOVA was employed. Marginal means were also estimated, and to assess the model fit quality, determination coefficients were calculated: R^2, adjusted R^2 (ANOVA), and conditional R^2 (repeated-measures ANOVA). Tukey's test was applied for multiple comparisons. The significance level adopted was 5%, and the software used was R Core Team 2021.

3.3 Microstructural and mineralogical analysis

The hydration of the cementitious composites was halted at the assessment ages (7, 28, 91, and 182 days). After the mechanical tests were conducted, selected fragments were immersed in isopropyl alcohol for 24 hours. They were then dried in an oven at 40°C for an additional 24 hours to halt the cement hydration process, preparing them for XRD and SEM analyses.

The scanning electron microscope (SEM) was used to evaluate the morphology and microstructure of the samples, as well as to characterize the hydration products formed. Micrographs were obtained using an Oxford analytical X-ray microscope, with magnifications ranging from 50x to 20,000x.

Diffraction data for all samples were collected using a diffractometer operating in transmission mode, employing CuKα1 radiation (λ = 1.54056 Å) in the range of 10 to 70 °, with step sizes of 0.015 ° and a counting time of 100 s for each 1.05°.

4. RESULTS AND DISCUSSIONS

4.1 Fresh state density and air entrained

Figure 2 highlights the results of fresh-state mass density and air content in cementitious composites with the addition of FC-MCC celluloses.

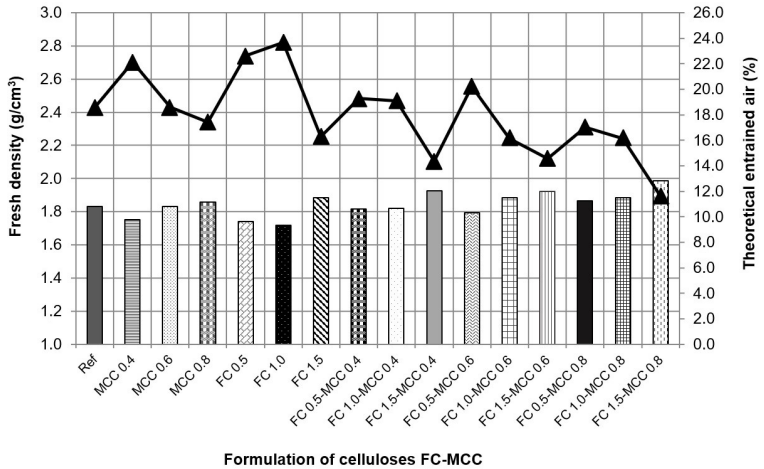

Figure 2 – Fresh Density (column) and Theoretical entrained air (line) of the cementitious composites

In this study, mass densities were found to range from 1.72 g/cm^3 to 1.99 g/cm^3, with air content varying between 11.64% and 23.65%. Based on the air content results, it was observed that the addition of cellulose microfibers and crystalline microcelluloses promoted air incorporation in the mixture, acting as air-entraining agents, which also led to a reduction in the mass density of the cementitious composites [52]. This may occur due to the contact of cement particles with the flat surface of microfibers and microcellulose particles, generating bubbles within the system and inducing the trapping of voids in the cementitious system [26]. The effect of microfibers and microcellulose particles on air incorporation results in a reduction in the density of the cementitious composites.

Regarding the combination of celluloses, it can be observed that high levels of cellulose microfiber (1.5%) increased the average mass density values and reduced the air content. The highest increase in mass density and the greatest reduction in air content occur with the FC 1.5-MCC 0.8 formulation, with 8.05% and 37.56%, respectively. This can be justified by the greater incorporation of fine materials in the cementitious system [53]. Concerning cellulose microfiber levels, it is evident that levels above 1.5% do not provide advantages when it is necessary to reduce the mass density of cementitious materials.

4.3 Mechanical strength

The results of compressive strength for cementitious composites with the addition of FC-MCC celluloses over hydration time (7, 28, 91, and 182 days) with an indication of the standard deviation of the measurements are schematically shown in Figure 3. The statistical test considering 95% confidence intervals is also presented.

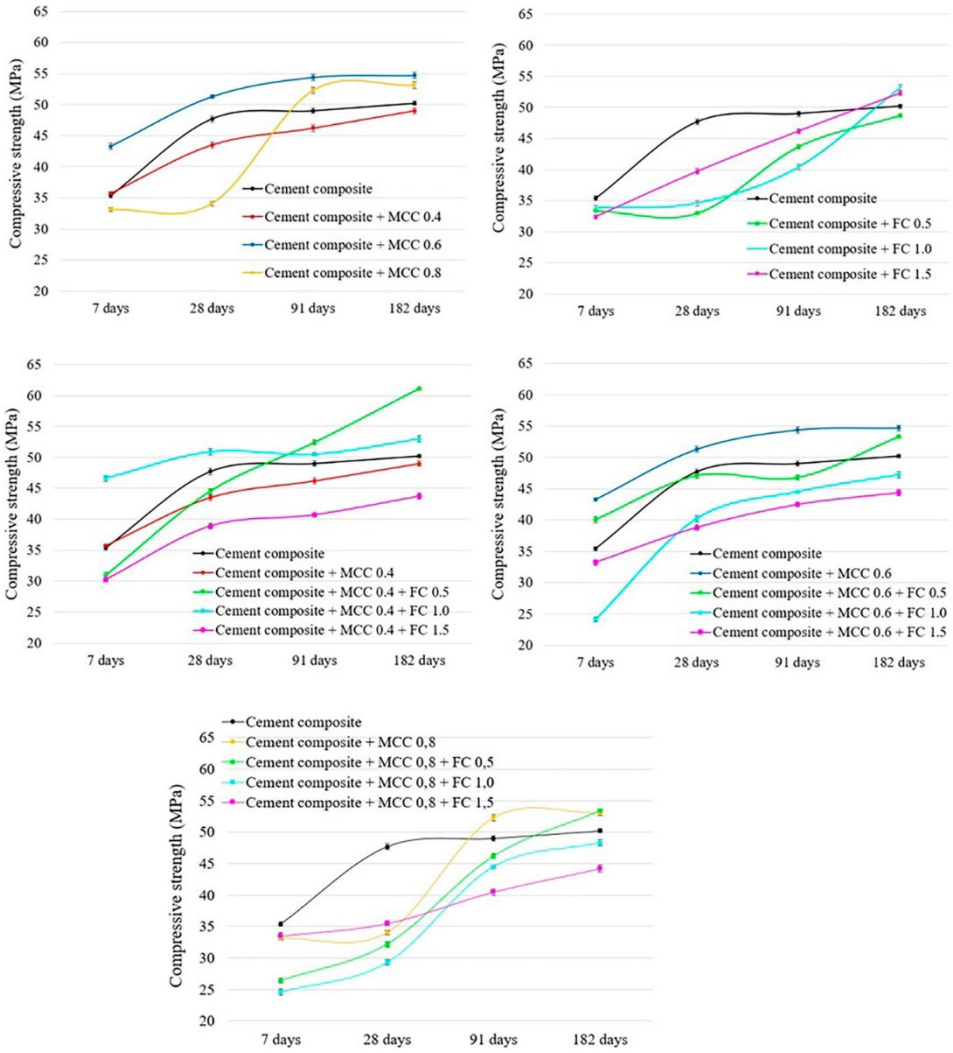

Figure 3 – Results of the compressive strength test for cementitious composites with the addition of FC-MCC celluloses

The results of cementitious composites with the addition of cellulose microfiber were similar to the reference at 7 days. At 28 days, the cementitious composites with cellulose microfibers (0.5%, 1.0%, and 1.5%) showed a significant percentage reduction of 30.81%, 27.46%, and 16.77%, respectively. It can be stated that cellulose microfibers did not positively contribute to compressive strength at the ages of 7, 28, and 91 days. The results obtained are consistent with the analysis of air content, where the addition of cellulose microfibers resulted in an increase in air incorporation in the cementitious composites. It can be observed that, despite the improvement in properties in the fresh state, the addition of cellulose microfibers also resulted in a decrease in compressive strength.

At 182 days of age, the incorporation of cellulose microfiber at 1.0% and 1.5% did not significantly influence compressive strength. Therefore, it can be concluded that the cellulose microfibers used in the research did not positively contribute to compressive mechanical performance in cementitious systems. The results found in this research are consistent with previous studies [30], [54]–[56], in which the authors justify the reduction in compressive strength by the water absorption phenomenon of microfibers and their subsequent release during the curing process, influencing the formation of voids and weakening the fiber/matrix interface. Additionally, cellulose microfibers promote air incorporation in cementitious systems.

Overall, the incorporation of crystalline microcellulose had a significant effect on compressive strength. The addition of 0.6% MCC contributed to a 22.31% increase at early ages (7 days) compared to the reference. However, cementitious composites with the addition of 0.4 and 0.8 MCC did not significantly influence compressive strength results at 7 days.

After 28 days, the incorporation of crystalline microcelluloses at 0.8% significantly decreased strength by 28.51% compared to the reference. On the other hand, cementitious composites with 0.6% MCC increased compressive strength by 7.54%. This suggests that at low doses, crystalline microcelluloses can effectively fill nanopores within the cementitious system, resulting in increased compressive strength [57].

The crystalline microcellulose levels at 0.6% and 0.8% increased by 11.02% and 6.73%, respectively, in percentage compared to the reference composite, enhancing compressive strength at 91 days. The incorporation of 0.8% MCC changed its behavior, as there was a decreasing trend at 7 and 28 days, where the reinforcing effect of MCC decreased as the MCC addition increased Oh et al. [57] and Nassiri et al. [58] also observed this phenomenon. It can be observed that there was a delay in the hydration process of the cementitious composites containing 0.8% MCC and the tendency for agglomeration promoted by the addition of 0.8% MCC, as observed in Figure 1c.

The addition of 0.4% MCC had no influence on compressive strength at any age. On the other hand, the addition of 0.6% and 0.8% increased by a percentage of 8.96% and 5.77% at 182 days, showing statistically significant increases.

For combinations of microfiber cellulose with crystalline microcellulose at 0.4%, the results indicate that the combination with a higher content of microfiber cellulose (FC 1.5-MCC 0.4) showed a negative effect at all studied ages. However, with the combinations of additions FC 0.5-MCC 0.4, there was a reduction by a percentage of 12.71% and 6.82% at ages 7 and 28 days, respectively, when compared to the reference samples, which was significant at a 95% level. Nevertheless, at more advanced ages, this formulation changed its behavior, increasing by 6.93% at 91 days and 21.71% at 182 days compared to the reference, and these were statistically significantly different, being the formulation that showed the highest increase in compressive strength at 182 days. The combination of formulations FC 1.0-MCC 0.4 showed an improvement in compressive strength at all studied ages.

The combinations of FC 1.0-MCC 0.6 and FC 1.5-MCC 0.6 significantly reduced compressive strength in cementitious composites at all ages studied. The combined treatment of FC 0.5-MCC 0.6 showed promising results with an increase of 13.27% at 7 days and 6.17% at 182 days compared to the reference, and these were statistically significant. At intermediate ages, the results were similar to the reference.

All combinations of microfibers of cellulose with crystalline microcellulose at 0.8% significantly reduced compressive strength in cementitious composites at all ages studied, except for the combined formulation of FC 0.5-MCC 0.8, where there was an increase of 6.17% only at 182 days.

The addition of crystalline microcelluloses at 0.6% increased compressive strength at all ages studied, potentially indicating that this formulation may have a nucleating effect, filling nanopores in cementitious systems. However, the high content of crystalline microcellulose (0.8%) did not show increased strength up to 182 days, suggesting a delay in the hydration degree of the cementitious pastes caused by the tendency of agglomeration of this formulation, however, further testing is needed.

Regarding the combination of FC-MCC in the cementitious matrix, it was possible to verify that high contents of microfiber cellulose (1.5%) and crystalline microcellulose (0.8%) gradually decreased the compressive strength. However, formulations MCC 0.4 + FC 0.5; MCC 0.4 + FC 0.5 and MCC 0.6 + FC 0.5 showed promising results for compressive mechanical performance compared to the reference.

The evolution of flexural tensile strength of cementitious composites with the addition of FC-MCC at different hydration ages (7, 28, 91, and 182 days) is schematically illustrated in Figure 4, along with the standard deviation of measurements and statistical testing considering 95% confidence intervals.

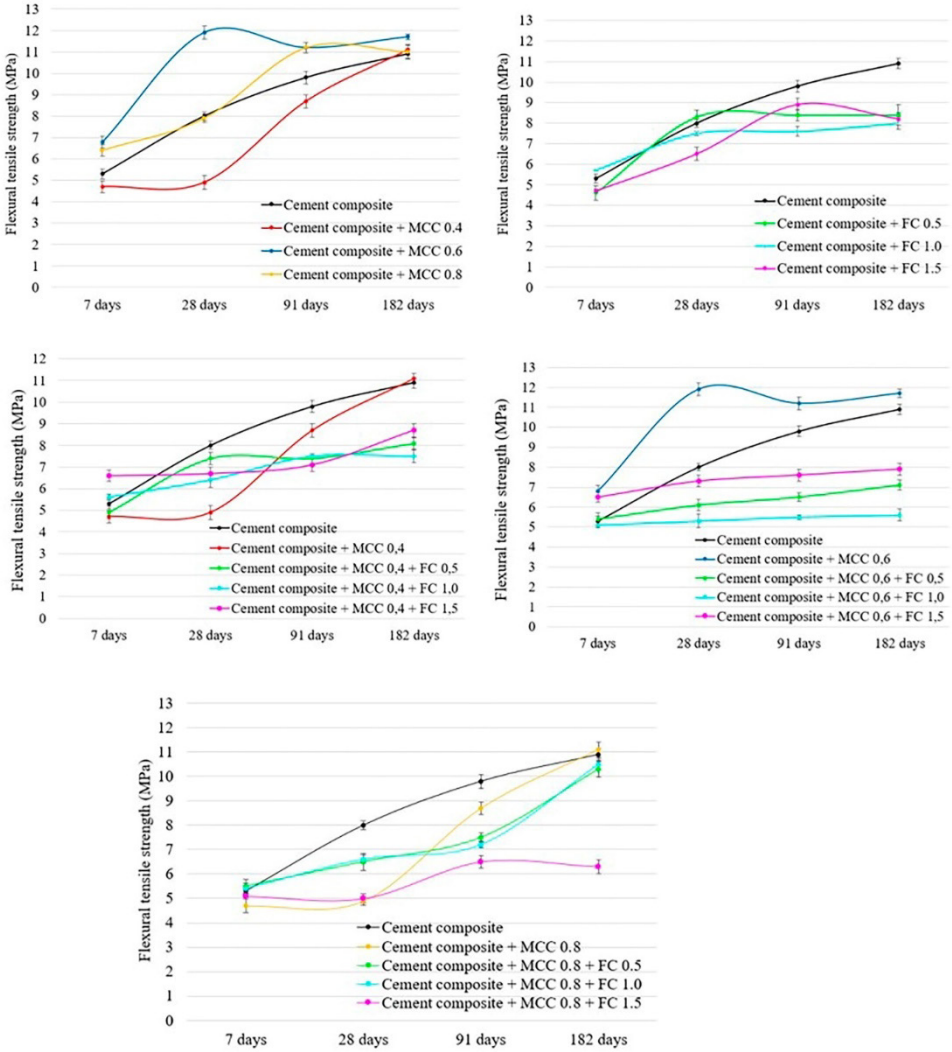

Figure 4 – Results of the flexural tensile strength test of cementitious composites with the addition of FC-MCC

Overall, the incorporation of cellulose microfiber did not contribute to the flexural tensile strength at the studied ages (7, 28, 91, and 182 days) of hydration in the cementitious composites. This result can be justified due to the micrometric scale of cellulose microfibers. Zhang et al. [1] evaluated cellulose fibers on a scale of μm and mm and concluded that only mm-scale cellulose fibers significantly improve flexural tensile strength, while μm-scale cellulose fibers do not contribute to the mechanical performance in compression and flexural tensile strength.

The additions of 0.5% FC and 1.5% FC showed a negative effect on the cementitious composites at 7 days. The addition of 1.0% FC resulted in an increase of 7.54% at this age, but this increase was not significant at a 95% confidence level. At 28 days, the incorporation of cellulose microfibers at 1.0% and 1.5% had a negative effect on the cementitious composites, and the addition of 0.5% had a result similar to the reference. At 91 and 182 days, the incorporation of all percentages of cellulose microfibers used in this research showed a negative effect on the cementitious system.

The incorporation of 0.6% MCC in cement pastes showed a percentage increase of 28.3% (7 days), 48.75% (28 days), 21.73% (91 days), and 7.33% (182 days) compared to the reference and was statistically significant, being the best formulation in mechanical performance, both in compressive strength and flexural tensile strength. These results are consistent with previous studies conducted by Ferreira [31], Silva et al. [32], Parveen et al. [35], Souza et al. [36],

Alshaghel et al. [37], which claim that crystalline microcellulose enhances the mechanical performance of cementitious systems, potentially correlating with the interaction between MCC and cement hydration products.

For the MCC 0.8 formulation, there was a percentage increase of 20.75%, which was statistically significant at the age of 7 days. However, at more advanced ages, the values were similar when compared to the reference. The MCC 0.4 formulations significantly reduced flexural tensile strength at 7, 28, and 91 days. At 182 days, the results were similar to the reference.

In general, the FC's-MCC 0.4 formulations did not show improvement in the flexural tensile strength of the cementitious composites, except for the FC 1.5-MCC 0.4 formulation at 7 days, which exhibited a percentage increase of 24.52% compared to the reference. At the ages of 91 and 182 days, reductions in flexural tensile strength were statistically significant.

The combined additions of FC's-MCC 0.6 decreased the flexural tensile strength of the cementitious composites, except for the FC 1.5-MCC 0.6 formulation at 7 days, which showed an improvement of 22.64% compared to the reference.

Similarly to the combinations FC's-MCC 0.4 and FC's-MCC 0.6, the multi-scale addition of FC's-MCC 0.8 showed a reduction in flexural tensile strength compared to the reference.

Based on the results obtained, it was possible to verify that the combinations of FC-MCC reduced the mechanical performance of flexural tensile strength, except for the isolated addition of crystalline microcellulose at 0.6%.

4.4 Effect of FC-MCC cellulose combinations on property estimation

The compression and flexural tensile strength tests were evaluated for variability through linear regression analysis. The factors assessed were the cellulose content (formulation factor) at sixteen levels (0.5%FC; 1.0%FC; 1.5%FC; 0.4%MCC; 0.6%MCC; 0.8%MCC; 0.5%FC+0.4%MCC; 1.0%FC+0.4%MCC; 1.5%FC+0.4%MCC; 0.5%FC+0.6%MCC; 1.0%FC+0.6%MCC; 1.5%FC+0.6%MCC; 0.5%FC+0.8%MCC; 1.0%FC+0.8%MCC; 1.5%FC+0.8MCC), the hydration evolution (age factor) at four levels (7, 28, 91, and 182 days), and the interaction factor between formulation and age. Table 9 shows the results of the linear model for the flexural tensile and compression strength tests.

Table 9 - The statistical analysis of mechanical performance

	dF	SS	MQ	F value	p-value	R^2	R^2 adj	R^2 c
Flexural tensile strength						0.985	0.980	
Formulation	15	329.57	21.97	302.88	<0.001			
Age	3	392.29	130.76	1,802.63	<0.001			
Formulation vs Age	45	196.84	4.37	60.30	<0.001			
Residual/Error	190	13.78	0.07		<0.001			
Treatment (Repeated Mesasure)	15	335.95	22.40	24.99	<0.001			0.788
Compression Strength						0.998	0.997	
Formulation	15	4,312.20	287.50	1,384.27	<0.001			
Age	3	9,918.10	3,306	15,919.26	<0.001			
Formulation vs Age	45	2,549.00	56.60	272.75	<0.001			
Residual/Error	174	36.10	0.20		<0.001			
Treatment (Repeated Mesasure)	15	4,406.00	293.73	24.88	<0.001			0.863

Legend: df – degree of freedom (n-1); SQ – Sum of Squares; MS – Mean Square; F value – F-statistic. R^2 – Coefficient of determination. R^2 adj – Adjusted coefficient of determination. R^2 c – Conditional coefficient of determination.

Overall, the adopted linear model shows very high coefficients of determination (R^2), adjusted determination coefficient (R^2adj), and conditional determination coefficient (R^2 c) (Table 6) for both compressive and flexural strength tests, indicating that the model terms are significant at a significance level of $\alpha = 0.05$. Thus, based on the analysis of variance, the employed linear model is significant, meaning that both compressive and flexural strengths are affected by formulations and hydration evolution.

Based on the characterizations of the cementitious composites, it was possible to conclude that the gradual increase in the addition of FC-MCC celluloses in the cementitious system leads to a similarly gradual reduction in compressive strength properties. Overall, FC-MCC celluloses reduced flexural tensile strength, showing average values lower than the reference.

The combined additions of FC-MCC celluloses that showed an improvement in compressive strength in the cementitious paste system were: FC 0.5-MCC 0.4; FC 1.0-MCC 0.4, and FC 0.5-MCC 0.6.

4.5 X-ray diffraction (XRD)

Comparing the diffractograms produced here with those found in the literature [45], [56], [59]–[62], it was possible to identify the crystalline phases resulting from the hydration of the reference sample (without FC-MCC celluloses) and the different cement pastes combined with FC-MCC celluloses produced. Figure 5 shows the X-ray diffraction patterns of the cementitious composites with the addition of FC-MCC celluloses at 28 days.

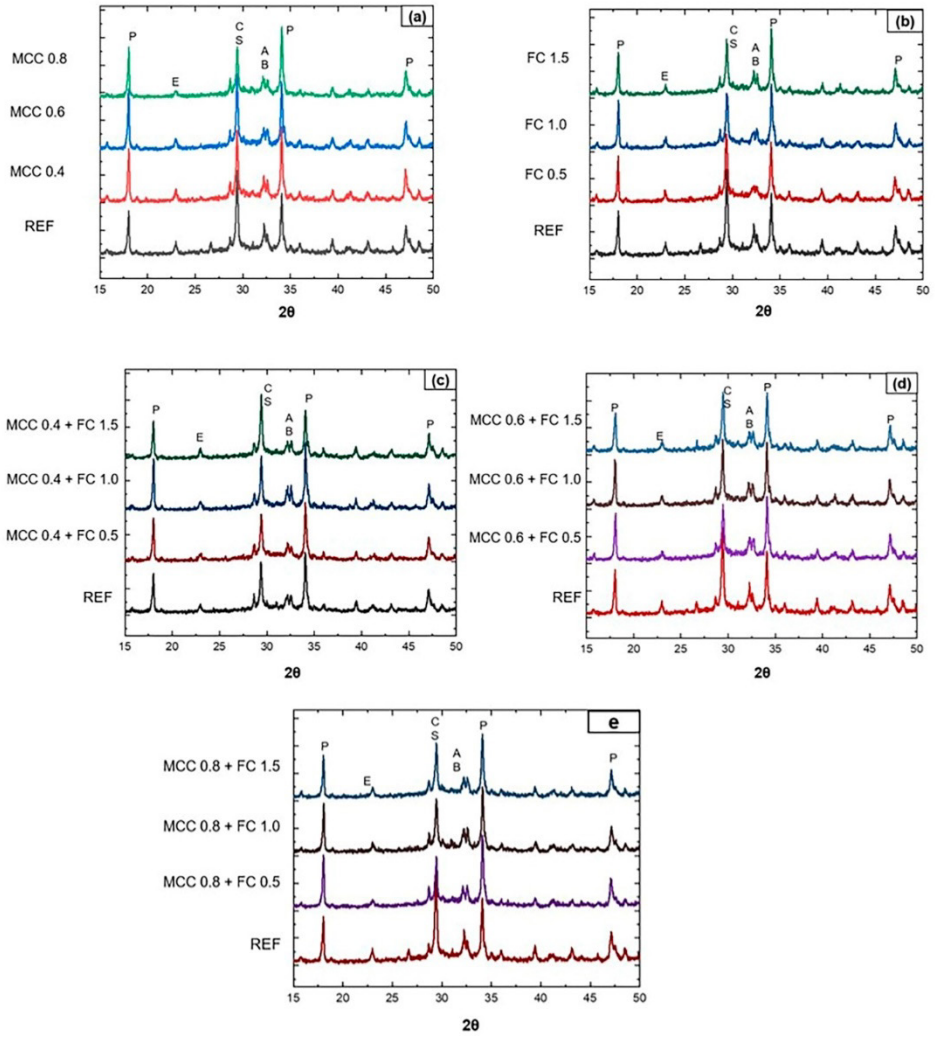

Figure 5 – X-ray Diffraction (XRD) test results of cementitious composites with crystalline microcellulose (a), cellulose microfiber (b), FC's-MCC 0.4 (c), FC's-MCC 0.6 (d), FC's-MCC 0.8 (e)Legend: P = Portlandite; E = Ettringite; C = Calcite; S = Hydrated calcium silicate; A = Alite; B = Belite; FC = Cellulose microfiber; MCC = Crystalline microcelulose

Through the XRD patterns, it was possible to verify that the crystalline phases of cementitious composites with the addition of crystalline microcellulose and cellulose microfiber were similar to the reference, indicating that the evaluated FC-MCC formulations did not lead to the emergence of a new mineral phase. They can be used without altering the crystalline conformation of the mineral phases in the cementitious matrix.

The X-ray diffraction patterns of samples with and without the addition of crystalline microcellulose and cellulose microfibers, as well as the combinations of FC-MCC, showed the expected hydration products, including the presence of Portlandite or calcium hydroxide (P) (2θ = 18.2°; 34.5°; 47.8°, 51°), Calcite or calcium carbonate (C), and Hydrated calcium silicate (S) (2θ = 29.0°), as well as peaks related to the anhydrous compounds of cement: Alite (A) (C3S) and Belite (C2S) (B) (2θ = 32.12°; 32.8°).

In the diffractograms of cementitious composites with the addition of FC-MCC celluloses, the presence of the main cementitious phases, such as Alite and Belite, as well as phases resulting from Portland cement hydration, such as Portlandite, Ettringite, Calcite, and hydrated Calcium Silicate compounds, are observed. The diffractogram profiles of

cementitious composites with the addition of FC-MCC celluloses, in general, are similar to those of Portland cement composites without additives.

4.5 Scanning electron microscopy

The morphology of cementitious composites with cellulose microfiber (0.5%; 1.0%; 1.5%) and crystalline microcellulose (0.4%; 0.6%; 0.8%) were examined. This was done through scanning electron microscopy (SEM) images of the fractured surface, highlighting the differences based on the type of addition, formulation, and hydration evolution. Figure 6 illustrates the influence of cellulose microfibers on the cementitious matrix.

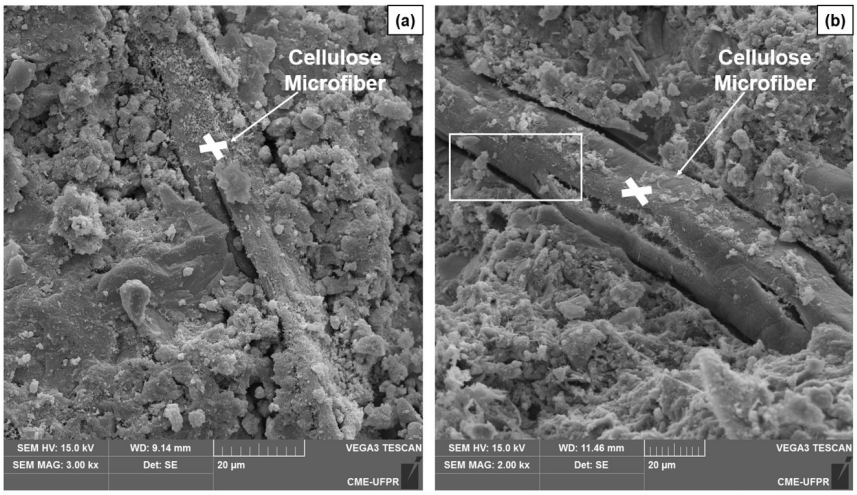

Figure 6 – Scanning Electron Microscopy (SEM) micrographs of the combined samples of FC 1.0-MCC 0.4 (a) and FC 1.5-MCC 0.8 (b) at 7 days

At 7 days, the interface between FC-MCC/hydrated cement is shown in Figure 6a, where it was observed that for the combined formulation FC 1.0-MCC 0.4, the microfibers are well incorporated into the cement matrix, demonstrating good fiber/cement adhesion. However, with a high content of microfiber cellulose (1.5%) and microcrystalline cellulose (0.8%) (Figure 6b), a weak adhesion with the cement matrix was observed, evidenced by the voids found between the microfiber cellulose and the cement matrix.

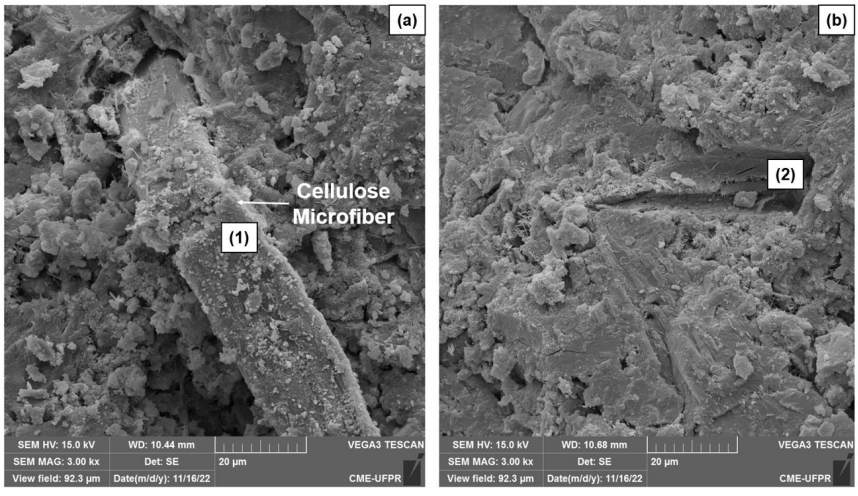

Figure 7 – Scanning Electron Microscopy (SEM) micrographs of combined samples FC 1.0-MCC 0.4 (a) and with the addition of FC 1.5-MCC 0.8 (b) at 182 days.

In Figure 7a, it was possible to observe the rupture of the cellulose microfiber, as indicated by number 1, in contrast to the fracture of the cementitious composites. On the other hand, in Figure 7b, the cellulose microfibers were pulled out, as indicated by number 2, demonstrating weak adhesion between the matrix and the reinforcement [63]. According to Soroushian et al. [10], microfibers that exhibit strong adhesion to the cement matrix tend to break rather than be pulled out from the fracture surface of the cementitious system. Therefore, it can be concluded that high contents of cellulose fibers (1.5%) and microcrystalline cellulose (0.8%) promoted weak adhesion to the cement matrix. The interfacial region of composite materials controls the stress transfer between the microfiber and the matrix, and performance mainly depends on the level of interfacial adhesion [64].

4 CONCLUSIONS

This study aimed to investigate the influence of additions of cellulose microfiber and microcrystalline cellulose on the compressive and flexural strength, as well as the microstructure of Portland cement composites at different ages. Therefore, the following conclusions can be highlighted:

- Regarding the mechanical performance of the cementitious composites, the cellulose microfibers used in the study did not contribute to the mechanical performance in terms of compression and flexural strength in the cementitious composites at any studied age.
- Crystalline microcellulose at 0.6% showed a significant increase in both compression and flexural strength at all studied ages, indicating that crystalline microcellulose can effectively fill nanopores within the cementitious system, resulting in improvements in mechanical performance. However, the addition of 0.8% did not show a significant impact, as the reinforcing effect of MCCs decreased as the MCC addition increased.
- Regarding the combined additions of FC-MCC celluloses in the mechanical performance of cementitious composites, the formulations FC 0.5-MCC 0.4, FC 1.0-MCC 0.4, and FC 0.5-MCC 0.6 showed an increase in compressive strength that was significant at a 95% confidence level. It was possible to conclude that the increase in the content of microfiber cellulose and crystalline microcellulose resulted in a reduction in mechanical properties. Additionally, it was observed that the decreasing relationship between compressive and flexural strength and the FC-MCC cellulose contents studied could be explained by a linear regression model, with regression coefficients (R^2) exceeding 0.788.
- The combined formulations that showed improvements in the mechanical strength of the cementitious pastes were FC 0.5-MCC 0.4, FC 1.0-MCC 0.4, and FC 0.5-MCC 0.6, indicating promising potential for application in construction materials.

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