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The influence of cellulose pulp and cellulose microfibers on the flexural performance of green-engineered cementitious composites

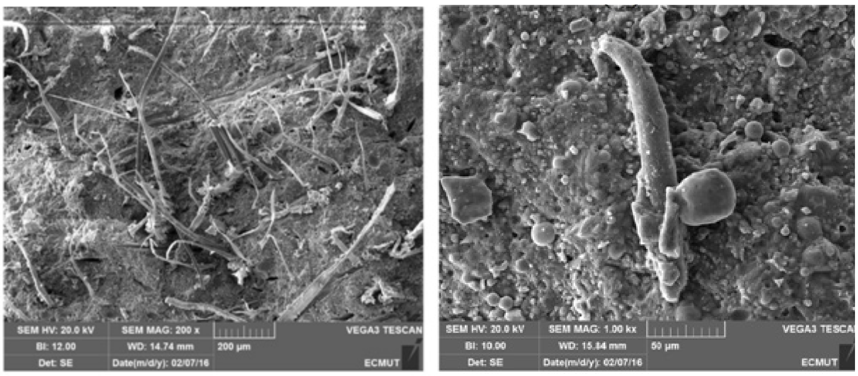
Fatemeh Masoudzadeh, Mohammad Fasihi, Masoud Jamshidi*

School of Chemical, Petroleum and Gas Engineering, Iran University of Science and Technology, Tehran, Iran

HIGHLIGHTS

- Mechanical and chemical treatment of Kraf paper to produce cellulose pulp (CP) and cellulose microfibers (CMF)
- Investigation of flexural properties of cementitious composites reinforced by CP and CMF
- Comparison of flexural behavior cement composites reinforced by CP, CMF and PVA fiber
- Preparation and study of hybrid engineered cementitious composites (ECC) by using a mixture of CMF and PVA fiber

GRAPHICAL ABSTRACT



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ABSTRACT

The aim of this study was to investigate the flexural behavior of engineered cementitious composites (ECCs) reinforced by cellulose pulp (CP) and cellulose microfibers (CMF). The reinforcements were obtained from chemical-mechanical treatments of Kraft paper and used in ECC mix design. Results showed that cement reinforced by CP exhibited a strain-hardening behavior in the three-point bending test, while CMF led to a brittle behavior in cement composites. Moreover, different hybrid combinations of polyvinyl alcohol (PVA) and CMF achieved quite a high strength while maintaining a high level of flexural toughness. A combination of 0.5 vol% CMF and 1.5 vol% PVA resulted in a significant increase in flexural toughness and a slight improvement in flexural strength. The properties of this hybrid composite were comparable with one containing 2 vol% of PVA fiber.

* *Corresponding author: Tel.: +9821-77240496 ; Fax: +9821-77240495 ; E-mail address: mjamshidi@iust.ac.ir*
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1. Introduction

In the past few decades, many studies have been conducted to modify the brittle behavior of cement materials to a more ductile one, mainly by incorporating various fibers. For this purpose, different types of man-made fibers and natural fibers were used in cement composites [1]. Environmental issues have created a great incentive to do extensive research on eco-friendly materials such as fibers obtained from natural resources. Natural fibers have many advantages over synthetic fibers as they are accessible, sustainable, non-hazardous and have low cost. Therefore, natural fibers were investigated as a potential alternative to synthetic fibers in many researches [2].

Natural fibers are obtained from plants, animals, and minerals. Plant fibers are individually comprised of four main chemical components namely cellulose, hemicelluloses, lignin and pectin. The mechanical properties of plant fibers strongly depend on their cellulose content [3,4]. In recent decades, some research has been performed on using natural fibers in cement composites. In a comparative study of plant fibers and PVA in cement-based composite, Juarez et al. [5] concluded that natural fibers can be an alternative candidate for PVA fibers to reduce the crack formation caused by plastic shrinkage. Khorami et al. [6] examined the effect of three types of agricultural wastes including bagasse, wheat and eucalyptus fibers on the properties of cement composites. Their results showed that natural fibers improved the energy absorption and flexural strength of the cement composites. They also added Kraft cardboard wastes into the cement matrix and observed strain-softening behavior in the prepared cementitious composites [7]. In another study sisal fibers were incorporated into cement mortar and led to strain-hardening behavior [8]. This illustrated the capability of these fibers in structural applications. In addition, a cement composite with a strain-hardening behavior was obtained by adding bleached pine fiber to the cement [9]. Singh et al. [10] used a combination of polypropylene fiber and steel fiber to improve the toughness of cementitious composites.

Today, efforts to achieve sustainable consumption patterns, especially in construction and building materials, are growing. Engineered cementitious composites (ECCs), normally fine-grained concrete reinforced with PVA fibers with a strain-hardening behavior, are not an exception. Over the past few decades, many studies have been performed to develop a green engineered cementitious composite for infrastructure systems by replacing its ingredients (i.e. cement and fibers) with industrial wastes and/or natural fibers [11]. The first study on green ECC was performed by Lee [12,13]. Some studies focused on replacement of cement with industrial wastes or mineral fillers to produce green ECC [12,14]. Moreover, the performance of various synthetic fibers such as polyvinyl alcohol (PVA) [15], polyethylene (PE) [16], polypropylene (PP) [15,17,18], acrylic and nylon fibers [18-20] used in ECCs have been studied. However, there are limited reports on the application of natural fibers as reinforcement in cement composites.

In this study, the flexural behavior of ECCs reinforced by cellulose pulp and cellulose microfibers were studied and compared with that prepared by PVA fibers. In addition, the properties of hybrid ECC reinforced by a mixture of cellulose microfiber and PVA fiber was investigated.

2. Experimental

2.1.Materials

Ordinary Portland cement type I was supplied by Tehran Cement Co., Iran. The chemical composition of the cement is seen in Table 1. Brown Kraft paper was provided from Roxcel GmbH, India. Fly ash was supplied by Dirk G., India. Sand was provided from local companies. Polycarboxylate ether Paya 210 from Payazhic Co. (Iran) was used as super plasticizer. The particle size distribution of the sand is shown in Figure 1. PVA with the characteristics reported in Table 2 was used as reinforcement.

Table 1. Chemical composition of the cement (% wt)

SiO_2	Al_2O_3	Fe_2O_3	CaO	Na_2O	MgO	SO_2	Other
21.4	5.3	3.2	63.4	0.3	3	1	1.4

2.2. Preparation of cellulose pulp and cellulose microfibers

In order to produce cellulose pulp, the Kraft paper was cut into small pieces and soaked in water for 24 h. Then, it was stirred for 1.5 hr at 500 rpm by a mechanical mixer to obtain a homogeneous pulp suspension. This

Table 2. Physical and mechanical properties of PVA fiber

Fiber type	Fiber diameter (μm)	Length (mm)	Tensile strength (MPa)	Elongation (%)	Modulus of elasticity (CN/dtex)
PVA	20	6	1600	6.92	300

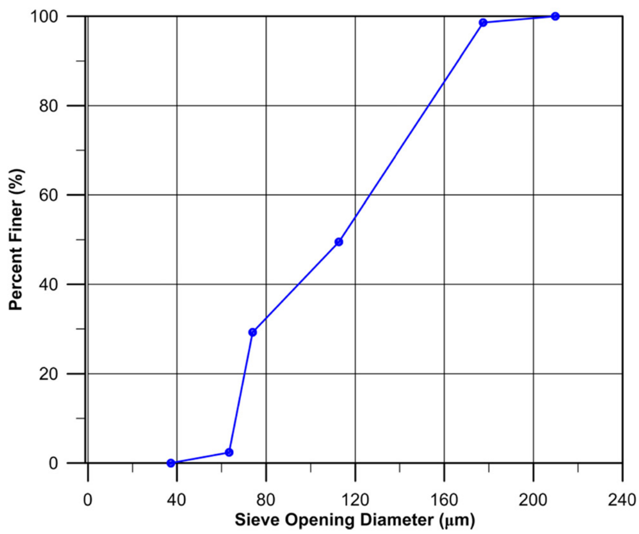


Fig.1. Cumulative particle size distribution of the sand

paste was used as cellulose pulp in the ECC mix design seen in Table 3. Cellulose microfiber was prepared in a mechanical-chemical treatment using the acid hydrolysis method [21,22]. To do this, first the cellulose pulp was immersed in sodium hydroxide solution (10 wt%) for 2 hr at 80°C under stirring. Then, the fibers were neutralized several times by washing with distilled water. The fibers were subjected to, acidic hydrolysis by sulfuric acid solution (30 wt%) for 3 hr at 80°C. The fibers were neutralization again after the hydrolysis process. Finally, the fiber suspension was sonicated for 15 min with a sonicator (UHP-400 from Ultrasonic Technology Development Co., Iran). Figure 2 presents the SEM images of the fibers before and after mechanical-chemical treatments.

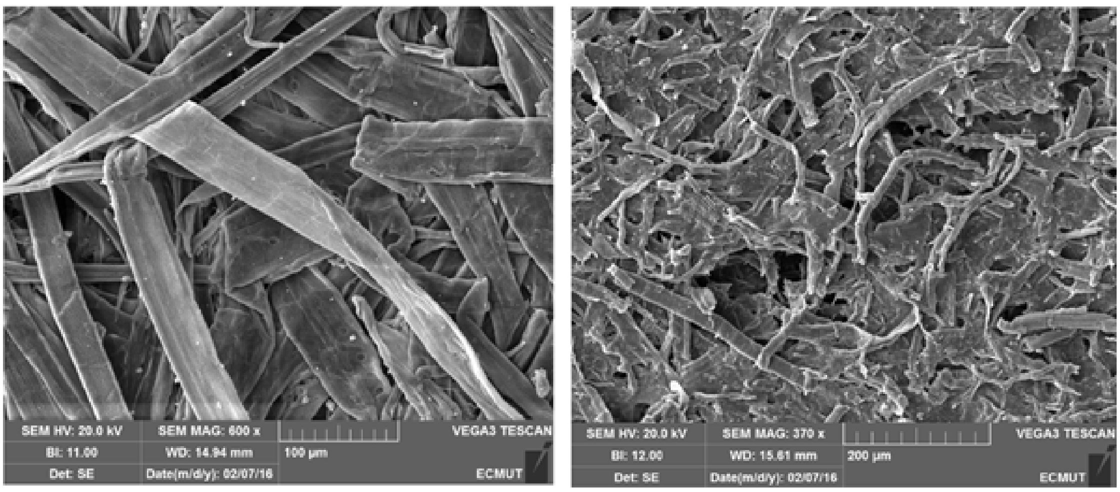
2.3. Cementitious composite preparation

A standard mix design, ECC-M45, was used to prepare the ECC samples [23]. The composition of ECC is presented in Table 3. The fibers used in this mix design were either PVA, CP or CMF.

A control sample without fiber was also prepared for comparison. Moreover, some hybrid fiber reinforced ECCs were prepared using different combinations of PVA and CMF. The total fiber content in the hybrid composites was 2 vol%. For each formulation three plate specimens with dimensions of 9×100×210 mm were cast using a suitable mold. Figure 3 presents an image of the applied mold. The samples were removed

Table 3. ECC mix design proportions

Cement (g)	Flay ash (g)	Sand (g)	Water (g)	Fiber (% vol)	Super plasticizer (g)
1	1.2	0.8	0.57	0-2	0.01-0.02



(a) (b)

Fig.2. SEM images of (a) Kraft and (b) cellulose microfibers

from the mold after two days rest and kept at ambient temperature and 95±5% relative humidity for four weeks before any tests.

2.4. Testing method

A three-point bending test was conducted using a SANTAM-STM150 universal machine according to EN-12467 standard test method. The flexural stress (σ) and strain (ε) were obtained by the following equations:

$$\sigma = \frac{3FL}{2bh^2} \tag{1}$$

$$\varepsilon = \frac{6Dh}{L^2} \tag{2}$$

where, F is the bending force, L is the bearing distance, b and h are the width and thickness of the ECC sheet, respectively, and D is the displacement of the loading bar. Toughness energy was calculated from the area under the stress-strain curve. The result of mechanical properties was obtained from averaging over five measurements.

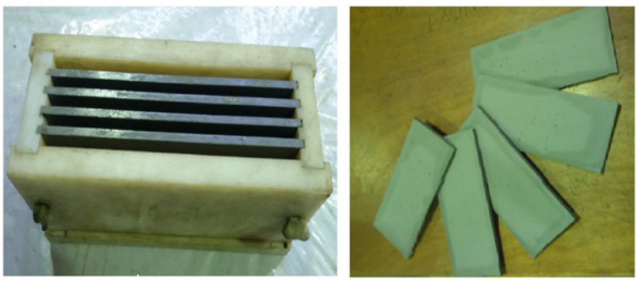
The microstructure of samples were observed by using scanning electron microscopy (SEM, Vega 3 Tescan, Czech).

3. Results and Discussions

3.1. Cement composites reinforced by CP and CMF

Figure 4 presents the bending test results of composites with different volume fractions of CP. As can be clearly seen, the ductility of the composites containing cellulose pulp increased compared to the control sample. The increment of ductility of the composite was proportional to the fiber content.

Claramuntet et al. [24] stated that the toughness and ductility of cement composites increased by adding



(a) **(b)**

Fig. 3. The mold used for preparing composite samples

softwood pulp. In this study, at 0.5% CP content, the composites displayed a reduction in flexural strength while their ductility increased. At higher concentrations of CP, the strength remained almost constant, but ductility obviously increased.

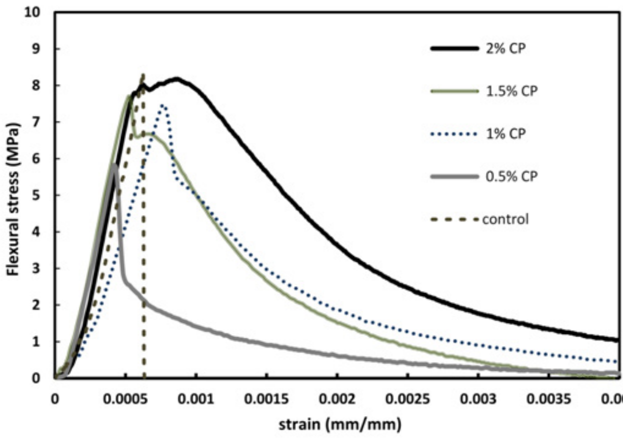


Fig. 4. Stress-strain curve of cement composites containing cellulose pulp

The flexural stress-strain curves of CMF reinforced composites are shown in Figure 5. The addition of microfiber to the cement not only decreased the flexural strength, but also lowered its strain to break and ductility. CP, which included long fibers, enhanced the ductility of the composites, while CMF failed to reinforce the cement because of its short fiber length. The aspect ratio and length of fibers in CMF decreased due to the fracturing of fiber during the mechanical-chemical treatment.

Figure 6 presents SEM images of the cross-section of specimens containing CP and CMF after the bending test. The image presents the pull-out mechanism for the composite containing CP, indicating non-strong

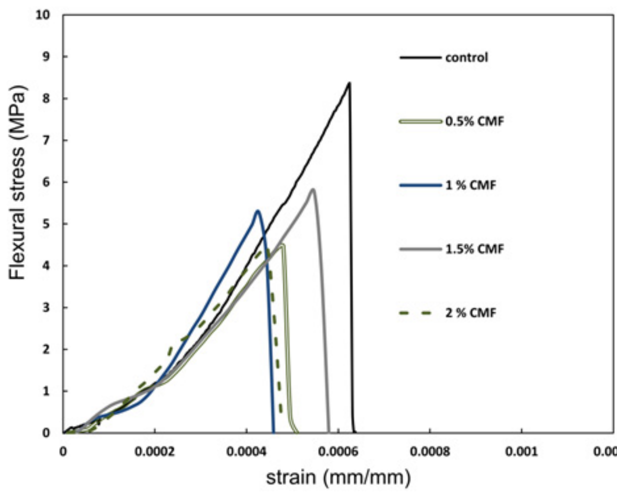
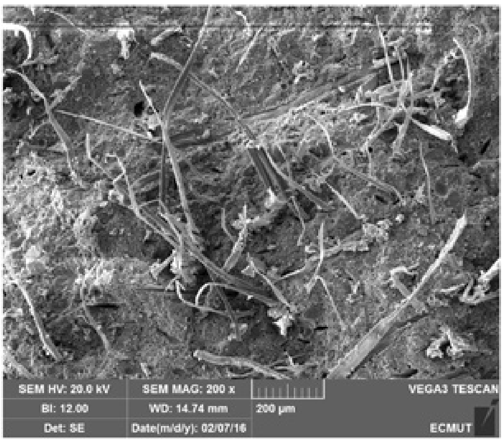
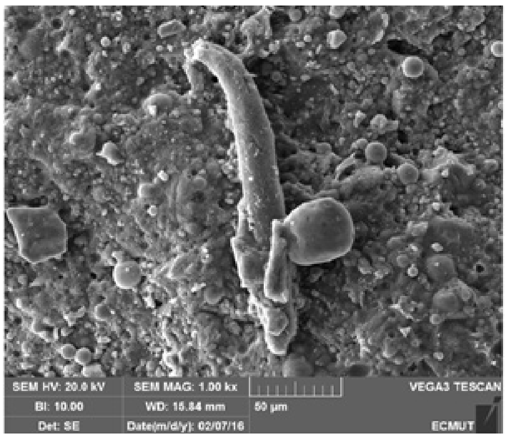


Fig. 5. Stress-strain curve of cementitious composites containing cellulose microfiber

adhesion between the untreated cellulose and cement matrix. The long fibers in CP can bridge a crack owing to their length and so improve the ductility of the composite. On the other hand, in the CMF reinforced cements, no pull-out of fiber from the cement was observed, indicating good adhesion between the treated fibers and cement matrix. However, CMF could not resist against crack growth by crack bridging due to their short length.



(a)



(b)

Fig. 6 SEM image of cement composites containing (a) CP and (b) CMF.

3.2. Cellulose vs. PVA fibers in the cement composites

PVA is the main polymer often used to modify cement. In order to study the performance of cellulose reinforced composites, cement composites including PVA fiber were also prepared and their properties were then compared with the cellulose-reinforced composites. Figure 7 presents the results of the flexural behavior of ECC prepared with up to 2% PVA fiber. As observed in this figure, strain-softening behavior was observed in the composite containing 0.5% PVA. By increasing the amount of fiber up to 2% both strength and toughness were increased and a strain-hardening behavior was observed.

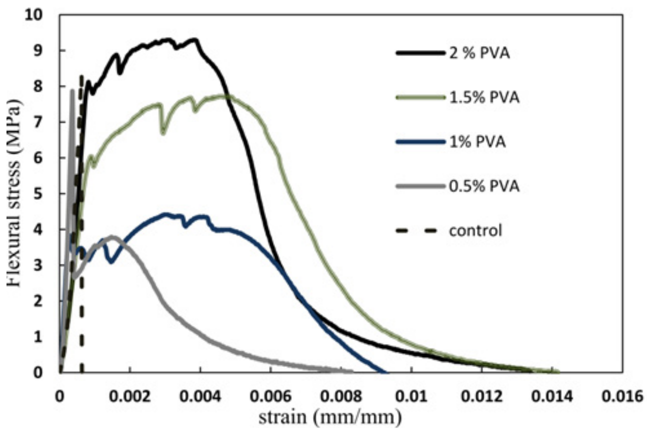


Fig. 7. Stress-strain curve of ECC containing PVA fiber

A comparison between the strength and toughness of the ECC containing PVA fiber and the composites reinforced by CP are depicted in Fig. 8. First-crack strength refers to the point at which the stress-strain curve becomes non-linear. Post-crack (second peak) strength is the maximum strength after the first-crack peak. The specimens produced by 0.5% and 1% CP only showed the first-crack peak as a result of semi strain-softening behavior. By increasing the fiber content to 1.5% and 2%, strain-hardening behavior and a post-crack peak appeared due to the higher surface contact between fibers and matrix. Also, in the ECC including PVA, increasing fiber volume fraction led to a reduction of the first-crack and an increase in the post-crack strength.

Figure 9 represents a comparison between the properties of CP, CMF, and PVA fiber composites at 2% of fiber volume fraction. The flexural strength of the composite containing CP was comparable with that of the PVA reinforced composite. While, the flexural strength of the CMF composite was about 40% lower. However, the toughness energy of the CP and CMF composites were much smaller than the PVA composite.

3.3. Composites prepared by hybrid fibers

Although CP made more improvement than CMF in the mechanical properties of cement, in hybrid form it showed a weak processability due to large bundles and aggregates, which did not facilitate its uniform dispersion in the cement mixture. On this basis, CP was not use in hybrid composites. Composites containing hybrid fibers were prepared by using CMF along with

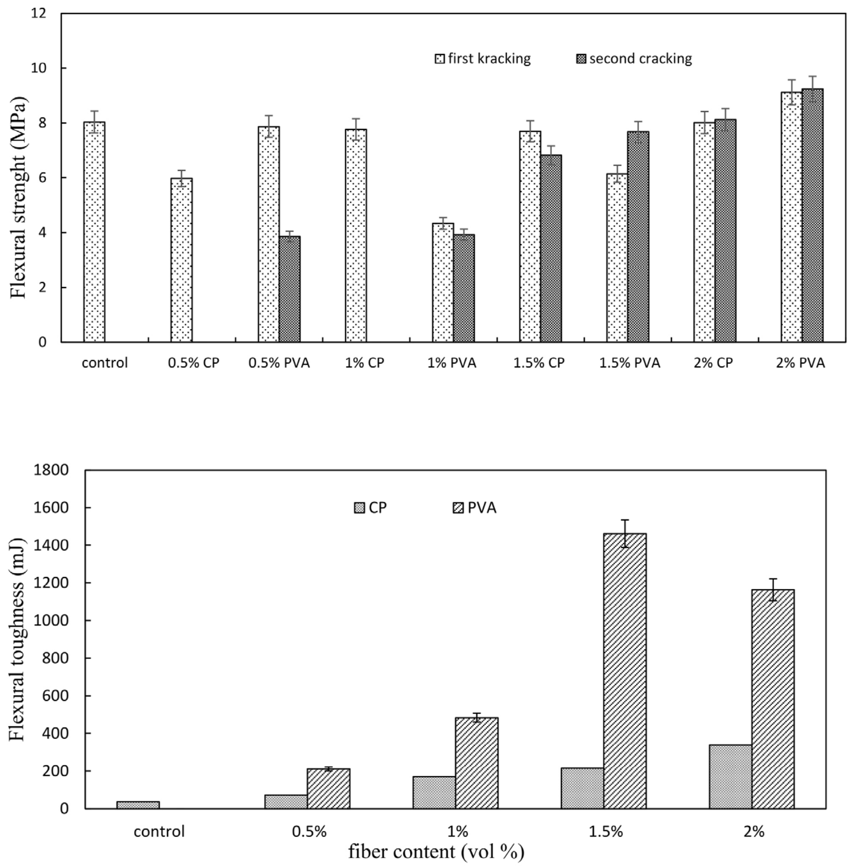


Fig. 8. Flexural properties of composites containing different content of PVA and CP

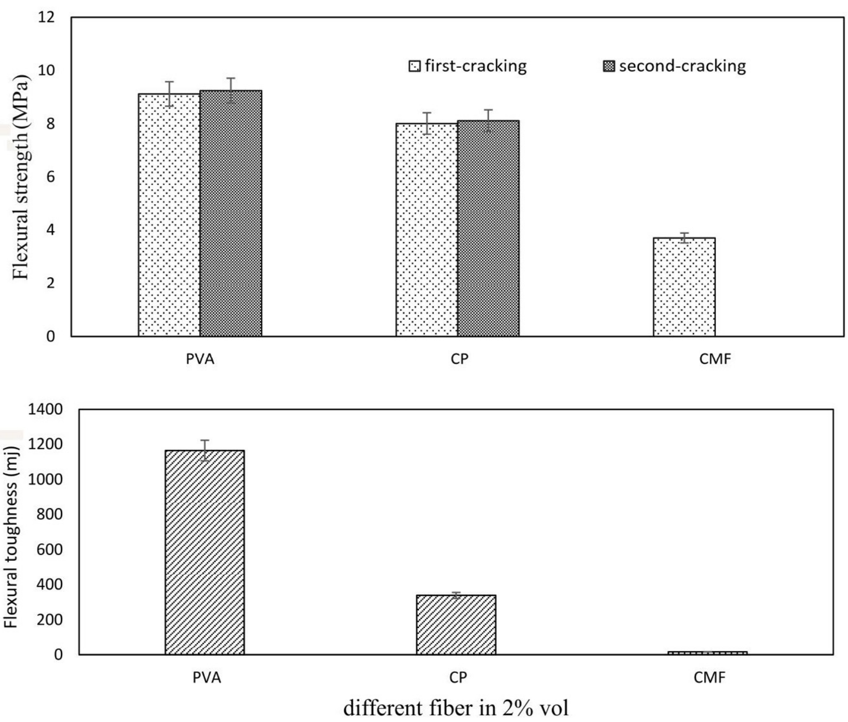


Fig. 9. Flexural properties of different cement composites containing 2 vol% fiber.

a total fiber concentration of 2 vol%. Figure10, illustrates the flexural properties of the hybrid composites. The flexural strength of the hybrid composites decreased as the CMF content increased. The results of flexural first- and post-crack strength and flexural toughness of the hybrid composites are displayed in Figure 11. As shown, the samples containing 1.5/0.5 and 1/1 volume percent of PVA/CMF represented a strain-hardening behavior similar to PVA-ECC. The hybrid composite including 0.5% CMF showed just a little lower flexural strength compared with 2% PVA ECC. In addition, the toughness energy of this hybrid fiber's ECC was on the level of pure PVA ECC. So, PVA fiber could be partially replaced by CMF in ECC mix design.

4. Conclusions

In this study, green cement composites were prepared by CP, CMF, and a hybrid of PVA fiber and CMF. By adding CP to the cement, the flexural strength did not change considerably, but toughness energies were significantly increased. The composite containing 2% CP presented up to nine times more toughness energy compared with the control sample. Although, CMF did not improve the mechanical properties of cement, the mixture of PVA and CMF in an ECC mix design created comparable properties with PVA-ECC at 2 vol% fiber. This indicated that in ECCs, PVA fiber could be replaced by CMF to some extent.

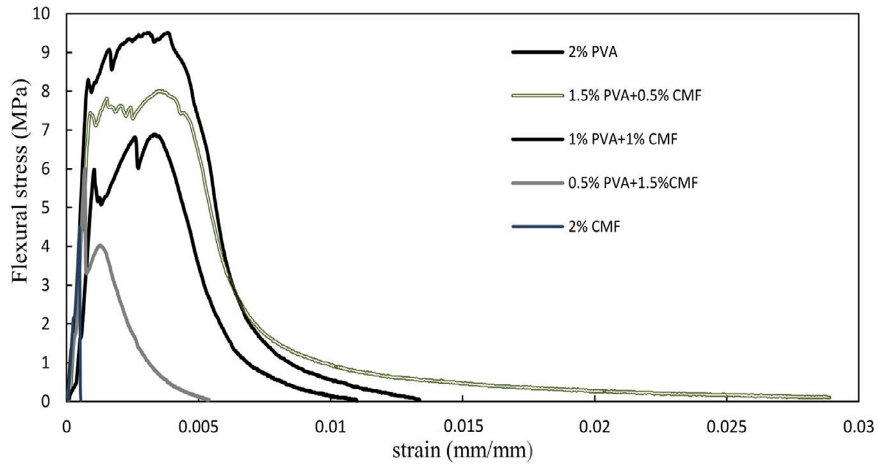


Fig. 10. Stress-strain curve of hybrid ECCs.

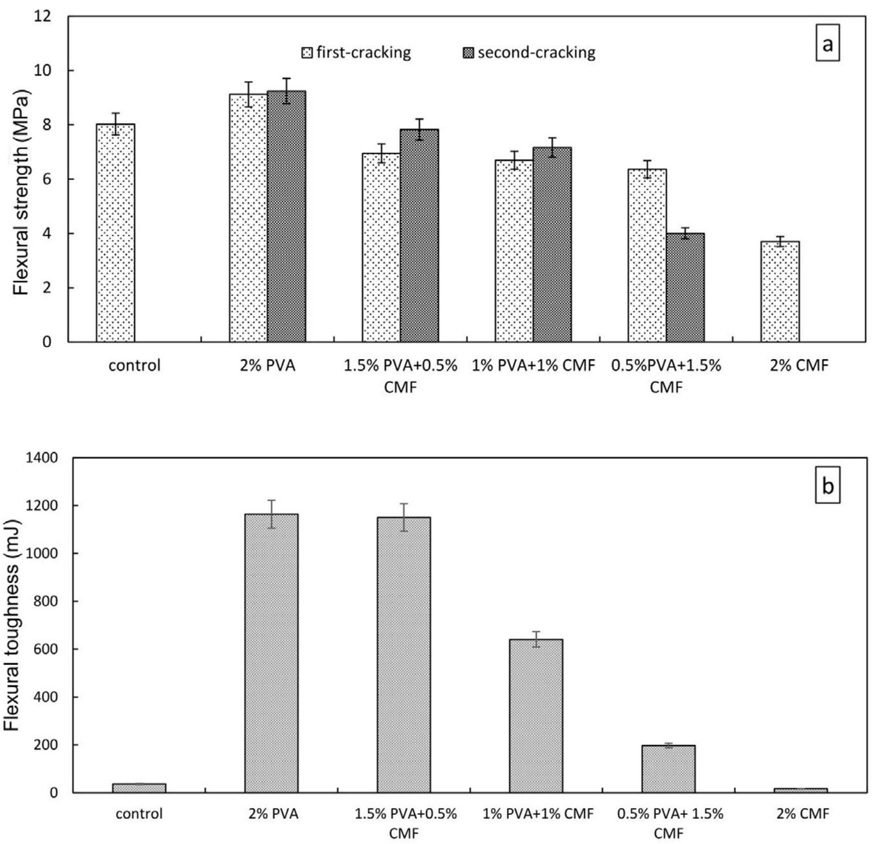


Fig. 11. The flexural properties of hybrid composites.

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