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CHARACTERIZATION OF FIBER-CEMENT COMPOSITES REINFORCED WITH ALTERNATE NATURAL CELLULOSIC FIBERS

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ABSTRACT

The eucalyptus and araucaria fibers were used as alternatives to virgin cellulose common in fiber-cement production. Three different types of these virgin-cellulose replacements were utilized as main raw materials while silica sand was the aggregate. Chemical and physical analyses were performed on the raw materials. In the experiments, the effect of various fiber types on the mechanical strength, water absorption and the density of fiber-cement mixture was studied for 8 wt % additions of these three fiber types. The excess water in homogeneous mixtures of the raw materials was first removed and then samples were pressed and baked in the autoclave. Physical and mechanical tests on these samples indicated that the fiber-cements produced with eucalyptus and araucaria cellulose were mechanically superior than those produced with virgin cellulose.

Keywords: Cellulosic fibers, Fiber-cement, Cement-based composites, Mechanical properties, Composite processing

1. INTRODUCTION

Composite Fiber Cement Board (FCB), used in building materials for many years are produced by mixing cement with various organic or mineral fibers at specified ratios. FCB can be produced in flat or corrugated shapes and FCB is a material which is usually used for external facades as well as is used for inner walls and floors [1].

Once the asbestos was established as a hazardous material to human health and its subsequent usage ban, finding alternative fiber materials has drawn the attention of many researchers [1] and natural fibers have begun to be used because of their advantages such as biodegradability, enhanced energy recovery, reduction of greenhouse gas emissions and lower pollutant emissions [2]. Environmentally, friendly and widely available materials such as jute, flax, sisal, kenaf, cotton, and hemp have been brought into consideration. Besides the mentioned privileges, the potential alternatives can also be extracted from low cost plant leaves [3].

The main ingredients of plant fibers are lignin, cellulose and hemicellulose [4]. In the development process of fiber reinforced cement composites without asbestos, lignocellulosic fibers have attracted appreciable consideration in past few years [5-9]. Wood fibers have great advantages, such as availability, non-hazardous nature, biodegradability, renewability and recyclability, lower cost and simple production processes when compared to asbestos fibers [9-12]. Cellulose fibers may have very different physical and mechanical properties due to their diameter, length, may density and different methods of processing and treatment [1].

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As asbestos was replaced with cellulosic fibers, manufacturing costs were lowered without sacrificing the mechanical properties. These fibers enhanced the composites toughness, flexural and tensile strength. The improvement of these properties depends on the fiber properties [12-13].

Wood fibers are separated into two categories: softwood and hardwood. It's very well known fact that these fibers are sourced from wide varieties of trees, the supply across the world is plentiful. There are two important distinctions between the two categories. Softwood fibers tend to be longer, and their amounts in a gram of pulp is distinctively higher from those of hardwood pulp. Otherwise, industrially processed plant based cellulose fibers have unique properties such as bond strength and alkali resistance. Furthermore, controlled production of such fibers minimizes differences in the dimensional and mechanical properties of the untreated cellulose [14].

Because of the advantageous properties of wood fibers such as biodegradability, non-hazardous nature, relatively lower cost, availability, renewability and recyclability, ease of processing for producing cementitious composites and numerous researches have been devoted to the development of this fiber [1].

In 2010 Tonelli et al., evaluated the advantages of using eucalyptus cellulose as an alternative to pinus and polymer fibers in cement based composites. In the meantime, characteristically, hardwood eucalyptus has short fiber length, while softwood pinus has long fiber length. Cellulose fiber lengths effects, mechanical performance of FC composites were investigated beside microstructure before and after aging cycles. In the study, it was determined that eucalyptus cellulose, having hardwood fibers, is better dispersed in the cement matrix than pinus, having softwood fibers, and containing more cellulose fibers per unit volume [1, 15].

In the current study, the eucalyptus and araucaria fibers were investigated to determine if they provide inexpensive or mechanically superior alternatives to virgin cellulose in fiber-cement production.

2. EXPERIMENTAL

2.1. Materials

Cement: Type I, Ordinary Portland cement was used in the experiment. Cement was supplied from CİMSA (Turkey).

Fibers: Three different types of American (Brazilian, Canadian) and European (Spaniard, Portuguese, Swedish, French) fibers were selected and virgin cellulose supplied by Cellulose Lerox International Company, eucalyptus and araucaria fibers by Kombassan Company (Turkey). Fibers included: cropped virgin cellulose (VC), blenched eucalyptus cellulose (EC) and blenched araucaria cellulose (AC). The fibers were refined and pulped to increase their mechanical properties. All types of fibers were fibrillated in the water for 15 hours.

Silica sand: It (silica content > 99%) was used in the experiments and was supplied by Ergen Mining Company in Sile region which has the highest silica ratio found in Turkey.

Water: Recycling water (pH = 13) was used to prepare the mixture. Since it is necessary to use recycled water in the production line, the recycling water was also used in laboratory work to make sure that results are representative of mass production results.

2.2. Characterization of the Raw Materials

The freeness degree of fibers was measured according to Canadian standard of ISO 5257/2-1 and micro structure of the specimen was investigated by Philips XL 30 SFEG brand scanning electron microscope.

Particle size, XRD, XRF and SEM analyses were performed for materials in the form of powder. For particle size distribution Mastersizer 2000 instrument was employed Rigaku D/Max 2200 system and Rigaku ZSX Primus were utilized for XRD and XRF analysis of samples, respectively.

2.3. Characterization of the Fibers

The measurement of the drainage rate of wood paste pulp in the suspension is defined as the degree of freedom (3 g of pulp in 1 L of water). Freedom or drainage rate is one of the main characteristics of the fibers within cement matrix. This analysis depends on the welling of the fibers, fine particle content, the morphology, fiber type, the degree of fibrillation, and the elasticity of the fibers [16]. According to the measurement results, the stock preparation depends upon the temperature, the water quality and surface properties. The Freeness test, discovered by the Canadian Pulp and Paper Research Institute, is commonly referred to as the Canadian Standard Freeness (CSF) [16-17].

The physical properties were determined (Table 1), SEM images (Figure 1) were taken and freeness tests were performed of the fibers.

Table 1. Physical properties of fibers

Property	**VC**	**EC**	**AC**
Average Diameter (µm)	30.34	9.79	43.55
Freeness Degree:	32	73	68
Drying rate %	80+/-2	94.1	93.52
Humidity %	3.98	5.9	6.48

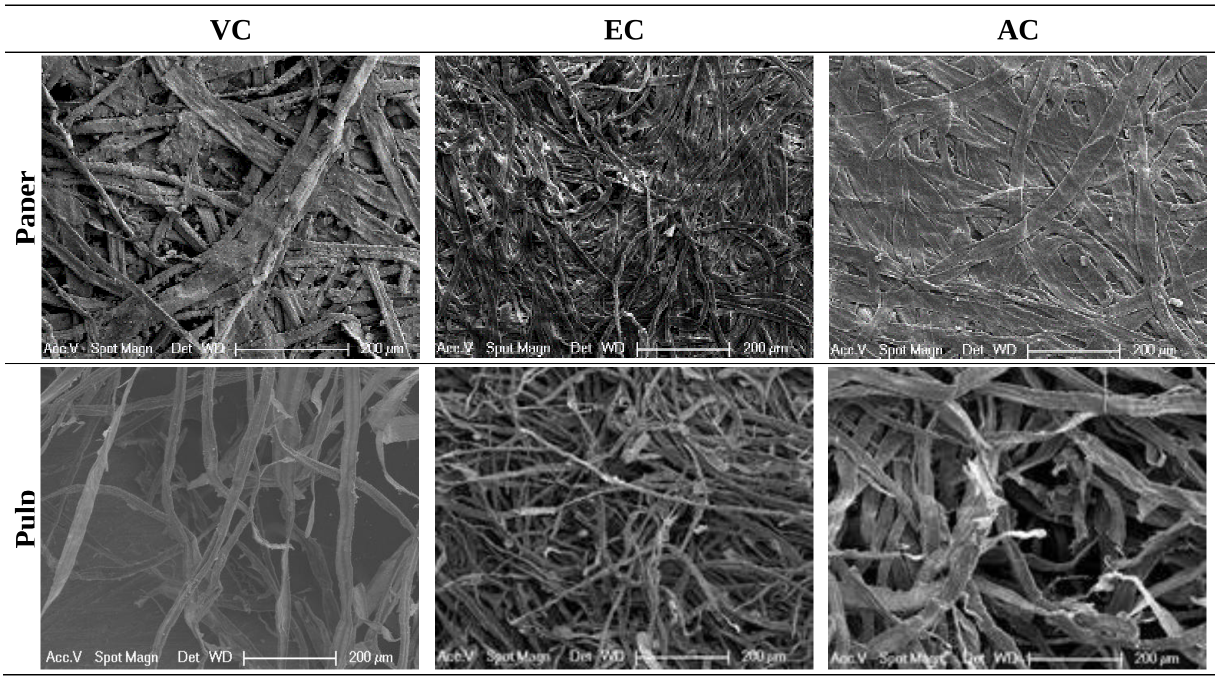

Figure 1. SEM pictures of cellulose samples (100X)

2.4. Characterization of silica sand and cement

The XRD results are shown in Figure 2, the particle size distribution is given below in Figure 3. The XRF analysis of cement and silica sand are supplied in Table 2.

X-ray diffraction patterns confirm that quartz is the main phase in the silica sand, other phases cannot be detected because of their low percentage. XRF results give better understanding for the content of the silica sand.

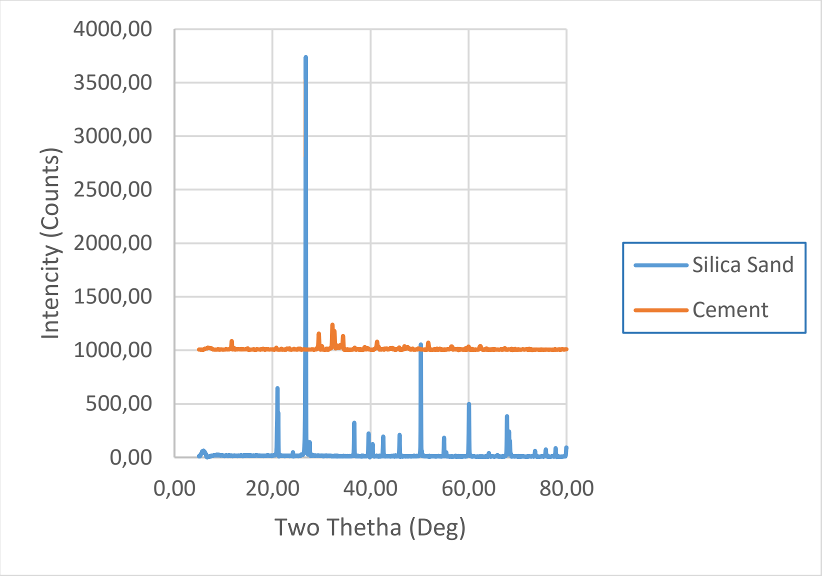

Figure 2. XRD analysis of cement and silica sand

Figure 2 presents the XRD patterns of the cement and silica sand. The most obvious peak is the quartz with a hexagonal structure coming from silica sand [18].

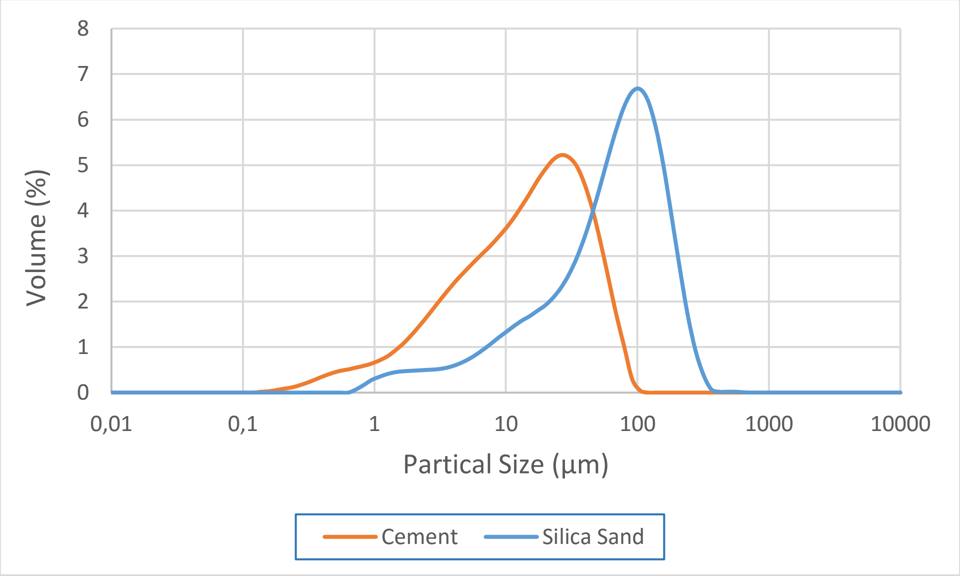

Figure 3. Particle size distribution of cement and silica sand

Figure 3 shows that the median size (d_{50}) of the cement was measured as 16.08 µm. The median size of the silica powder ground for two hours was 73.53 µm. The specific surface area of the cement was 1.23 $m^2.g^{-1}$ and the specific surface area of the silica sand was 0.83 $m^2.g^{-1}$.

Table 2. The chemical characterization of cement and silica sand

No.	Component	Result of Cement	Result of Silica Sand
1	Na_2O	0.3766	-
2	MgO	1.8747	-
3	Al_2O_3	4.7368	0.9310
4	SiO_2	19.3488	97.2451
5	P_2O_5	0.0766	-
6	SO_3	2.7137	-
7	K_2O	0.5299	0.3101
8	CaO	63.4967	0.2449
9	TiO_2	0.3630	0.2543
10	Cr_2O_3	0.1071	-
11	MnO	0.0765	-
12	Fe_2O_3	3.2886	0.6805
13	LOI*	3.0110	0.3340

LOI*: Loss in ignition.

The most important mineral components of Ordinary Portland cement are: $2CaO.SiO_2$ (dicalcium silicate), $3CaO.SiO_2$ (tricalcium silicate), $3CaO. Al_2O_3$ (tricalcium aluminate), $4CaO.Al_2O_3.Fe_2O_3$ (tetracalcium iron aluminate) and $CaO. SO_3.2H_2O$ (dehydrated calcium sulfate) [19]. As shown in Table 2, the silica sand has SiO_2 content of 97.3 wt%, CaO content of 0.24 wt%, Fe_2O_3 content of 0.68 wt%. The cement has SiO_2 content of 19 wt%, CaO content of 63.49 wt% and Fe_2O_3 content of 3.29 wt%. Figure 4 depicts the microstructure of the cement. It has been observed that the cement granules have heterogeneous grain size. Properties of the cement is good for general use according to TS EN 197-1: 2012 standard.

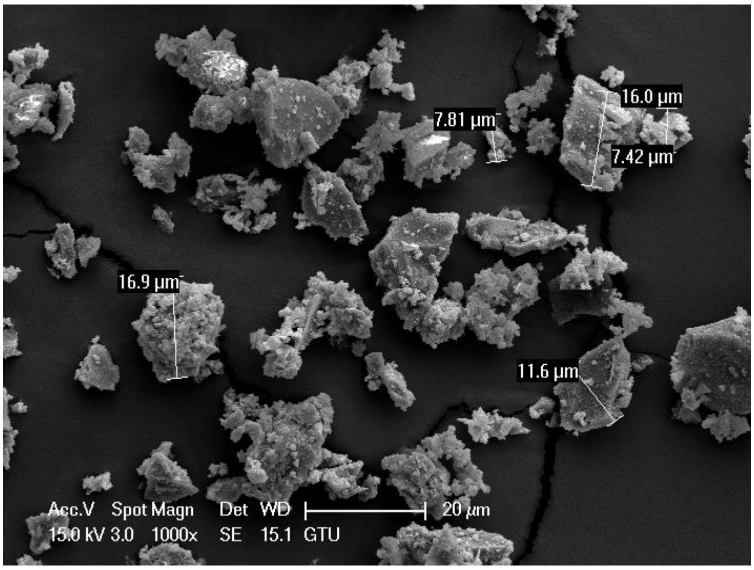

Figure 4. SEM picture of the cement

3. SPECIMEN PREPARATION

The flow chart of the production is exhibited in Figure 5. First step of the production is the preparation of the fiber-cement sludge which was obtained by adding cement and silica sand to the cellulose mixed for 15 hours. The addition process must be slow to avoid agglomeration. A small amount of flocculent water is added to the mixture for ensuring the adhesion of cement grains in the sludge. The water/cement ratio was targeted to be 30% by weight. The dry material content of the sludge are inhibited in Table 3.

Approximately 820 ml of the mixture is taken into a vacuum box at a pressure level of 400 mbar. The excess water in the slurry was discharged out with the help of vacuum pump. Following the vacuuming process, the fiber-cement samples were placed in the press machine and compressed at 7 MPa for 120 seconds. This process was repeated fifteen times for each fiber type. The produced samples were placed in the oven for pre-heating (30 ºC for 6 hours). After the pre-curing process was completed, the samples were cured in an autoclave for 11 hours at 180 ° C, 60 ºC/h interval was used for heating. Resulting fiber-cement samples are given in Figure 6.

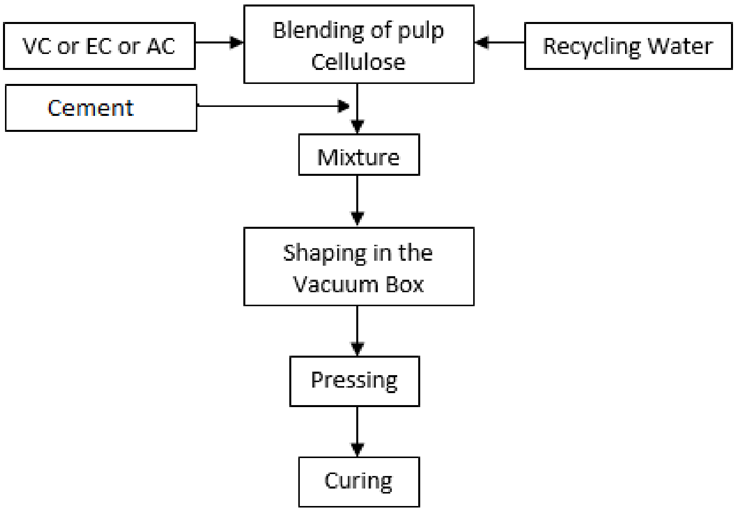

Figure 5. Flow chart of the production

Table 3. Mixing ratios of raw materials (weight without moisture)

	Cement (%)	Silica Sand (%)	VC (%)	EC (%)	AC (%)
A1	37	51	8	-	-
A2	37	51	-	8	
A3	37	51	-	-	8

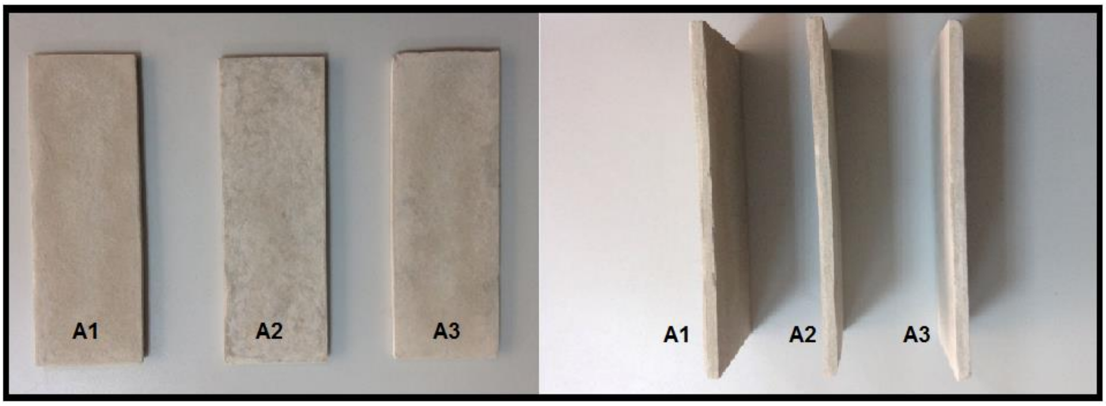

Figure 6. Produced samples

4. TESTS AND ANALYSIS

First, mechanical strength tests were performed on the produced samples to determine MOR (modulus of rupture) and MOE (modulus of elasticity) values. After that, Archimedes test was applied to the samples to determine the water absorption and the bulk density. SEM images were followingly taken.

5. EXPERIMENTAL RESULTS AND DISCUSSION

Mechanical tests were performed using the Intelli Jack 6 kN strength testing machine. A three point bending configuration was employed in the determination of MOR and MOE values.

$$\text{MOR (MPa)} = \left(\frac{L_{Max}}{b.h^2}\right) - (S_{down} - S_{up}) \tag{1}$$

$$\text{MOE (GPa)} = tg\alpha.\left(\frac{L_{Max}}{\delta}\right).\frac{(S_{down}-S_{up})^3}{b.h^3}\, x\, 10^6 \tag{2}$$

Where L_{Max} is the maximum load, $(S_{down} - S_{up})$ is the major span, b and h are the width and thickness of specimens respectively, $tg\alpha$ is the initial slope of the stress-strain curve and δ is the deflection of the fiber-cement. Stress-strain diagram can be seen from Figure 7.

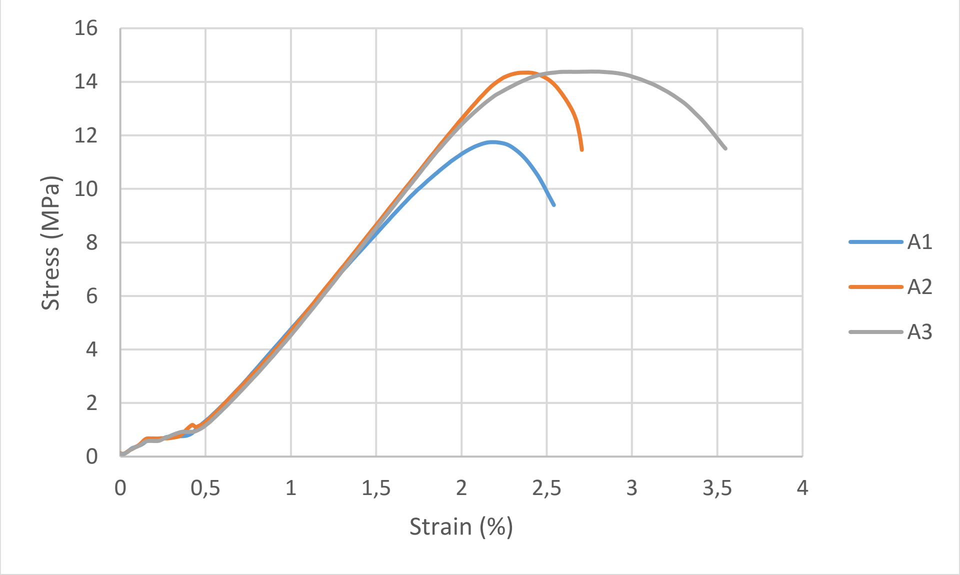

Figure 7. Stress-strain diagram of samples

The mechanical strength results of the produced specimens are listed in Table 4. It was observed that MOR value of the sample A1 was 12.30 MPa with a MOE value of 7.62 GPa, while the MOR value of sample A2 was measured higher 16.41 MPa with a MOE value of 8.08 Gpa. MOR value of sample A3 was measured as 14.67 MPa, and MOE value as 7.70 GPa. Figure 8 shows a graphical representation of the MOR and MOE values. Although all fiber-cement samples are sintered under the same conditions, the reasons of different mechanical properties are directly related to their fiber properties.

Table 4. Mechanical strength results of produced specimens

	A1	**A2**	**A3**
MOR (MPa)	12.30	16.41	14.67
MOE (GPa)	7.62	8.08	7.70

As shown in Figure 8, the higher MOE and MOR values of A2 are due to the shorter fibers of eucalyptus than those of virgin cellulose and araucaria cellulose. The main reason behind of this behavior was shorter microfibers having better adhesion to the matrix which was also supported with the micrographs given in Figure 9. In the composite material manufacturing process, it is possible to wet the fibers surface more efficiently when the fibers are short size, not to allow the formation of pores in the structure, and thus to provide a perfect hold at the matrix-fiber interface. If the sizes of the fibers are long, depending on the composite material preparation conditions, the pores in the structure can't be completely eliminated and thus a perfect adhesion at the fiber-matrix interface is not achieved. However, the addition of AC microfibers resulted in a dramatic increase in the ductility of A3 because the greater length of AC microfibers. This claim is also supported the larger values of the specific energy and the strain at the ultimate strength [20].

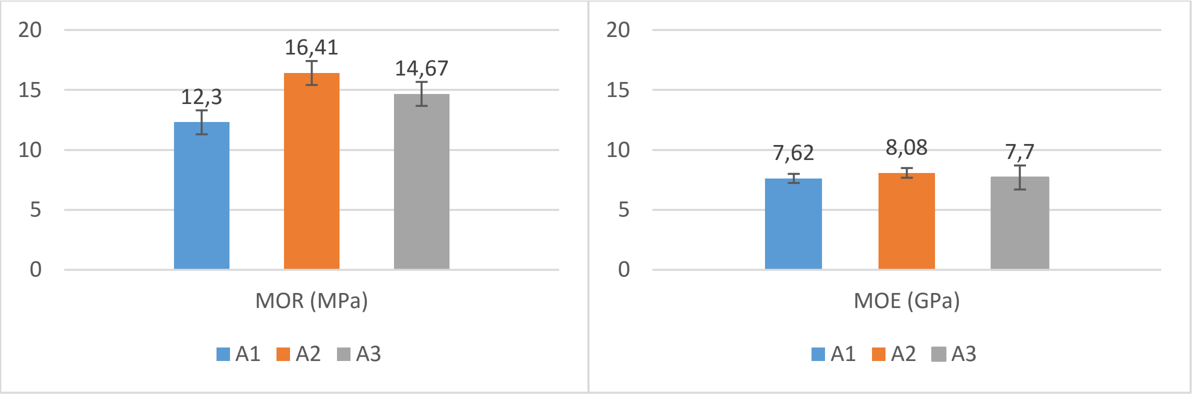

Figure 8. Graphical demonstration of MOR and MOE values

Archimedes method was performed in the evaluation of test samples. The values of bulk density, and water absorption were measured. The samples were dried in the oven to remove moist, then scaled and denoted as W1. Hereafter, the dried samples were soaked in water for 48 hours so that the samples could be saturated by water and then samples get scaled in the water with Archimedes scale, the suspended weight were denoted as W2. The samples were taken out from the water to determine their saturated weights which were denoted as W3. The bulk density and water absorption values of the samples were determined by the following equations:

$$\text{Bulk Density} = \frac{W1}{(W1-W2)} x100 \tag{3}$$

$$\text{Water Absorption } \% = \frac{(W3-W1)}{W1} x100 \tag{4}$$

The results of bulk density and water absorption of the samples are given in Table 5. The bulk density of the A1 sample was measured as 1,388 g/cm^3, the water absorption as 26.6 %. The sample A2 has shown the lowest bulk density with a value of 1.332 g/cm^3, and the highest water absorption value with 30.8 %. The bulk density of sample A3 was 1.468 g/cm^3, while the water absorption was 19.0 %.

Water absorption has negative effect on the mechanical properties and integrity of the fiber cement composites. Especially, the long-term stability of a fiber-cement composite is critically related to the conditions of micro fibers. Fiber-cement composites are sensitive to changes in length or diameter of the fiber, cracks can be formed within the material, as the cellulose fibers expand due to water absorption. Therefore, low water absorption is desired for these kinds of materials. Sample A3 has shown the best water absorption behavior, therefore long-term stability of A3 expected to be better than the others.

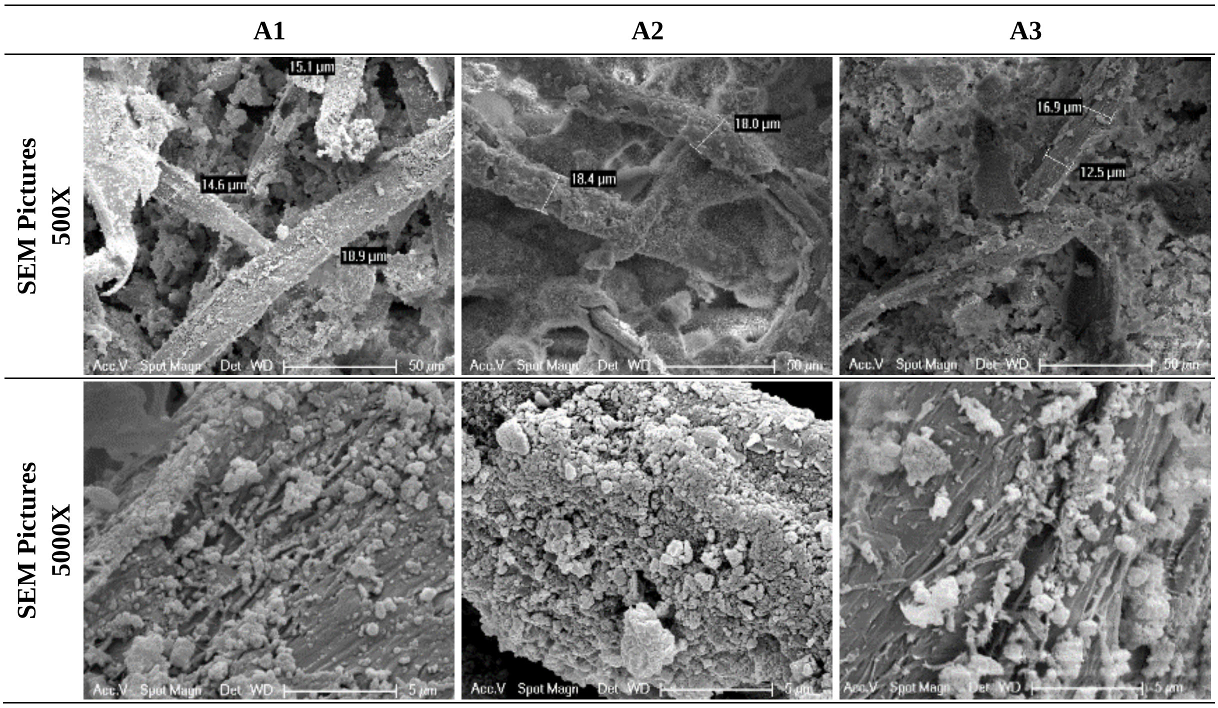

Figure 9. SEM images of samples

Table 5. Bulk density and water absorption results

	A1	A2	A3
Bulk Density (g/cm^3)	1.388	1.332	1.468
Water absorption (%)	26.6	30.8	19.0

SEM images were taken to analyze the microstructures of the produced samples and are supplied in Figure 9. The images were taken at 500X and 5000X and are showing the interaction of concrete and fibers. It seems that the fibers are finer in size, more homogeneous, multidirectional and entangled for A2 more than fibers in A1 which may suggest better and improved MOR values for eucalyptus cellulose reinforced cement. In A3, more of a cement-sand dominant structure emerges with pulled out larger fibers from the composite. The bulk density for A3 was measured to be higher than the other fiber-cement types. The microstructure for A3 seems to confirm that finding.

5.1. Economic Assessment of Cellulose Types

The primary problem with this technology is high material and manufacturing costs [21]. In Turkey, cellulose is one of the biggest problem of industry, which comprises about 40% of total raw materials cost. Table 6 shows the tonnage costs of cellulose types in Turkey. However, the price of cellulose may vary according to the country where they are produced or the energy costs and logistic expenses.

Table 6. The tonnage cost of cellulose types

	Virgin Cellulose	Eucalyptus Cellulose	Araucaria Cellulose
Costs ($/t)	570	550	600

6. CONCLUSION

According to the results of the present study, the mechanical and physical properties of the fiber-cement are directly related to the size and variety of fibers used. It has been found that eucalyptus cellulose offers advantages over virgin cellulose in the production of fiber-cement composite. This study has shown that, the eucalyptus and araucaria cellulose can be alternative to virgin cellulose in the production of cellulose fiber-cement composite.

The mechanical properties of fiber-cement composite with eucalyptus cellulose are higher than those produced with virgin cellulose and araucaria cellulose. The fiber-cement with eucalyptus cellulose is relatively inexpensive compared to others. However, long-term stability of eucalyptus cellulose is questionable because of its high water absorption. Still, the eucalyptus cellulose can be an alternative to fiber-cement composite producers in terms of cost. The mechanical properties of Araucaria cellulose are better than those of virgin cellulose. Additionally, water absorption value of this kind of cellulose is the lowest. Thus, Araucaria cellulose is a promising alternative. It can offer quality and life time advantage over virgin cellulose in fiber-cement production.

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