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Research article

Examination of water uptake performance and mechanical properties of PLA/flax fiber biocomposites with the coupling agent



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ABSTRACT

This paper studies the effects of water sorption on the microstructural and flexural properties of the flax fiber/poly(lactic acid) (PLA) biocomposites, compared to the composites with maleic anhydride (MAH) as coupling agents and alkali treatment. Five different biocomposites were prepared: 15 wt% and 25 wt% flax/PLA as the control group, and 15 wt%, 25 wt%, and 35 wt% flax/PLA with 5 wt% MAH. Ten various soaking times were selected to understand the water absorption behavior of the biocomposites. To investigate the mechanical properties of the biocomposites, impact tests were applied on the dry sample as well as the composites with 750-h and 1850-h water sorption. A three-point bending test was performed on the dry and 1850-h water sorption biocomposites to determine their flexural strength. Short flax fiber-reinforced PLA matrix biocomposites were compounded using extrusion and manufactured by injection molding. The flax fiber surface was treated using sodium hydroxy solution to advance the interface interaction between the fiber matrix and surface performance of the fiber and matrix. According to the results, alkali treatment improved the water gain resistance of the composites due to the enhancement of the interfacial bonding. The alkali-treated composites with maleic anhydride showed better impact and flexural strength than the composites without alkali-treated after 1850-h water sorption.

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KEYWORDS

Biocomposites
Water uptake
Flexural performance



1. Introduction

Environmental reasons and government legislation around the world have promoted academic and industrial research to develop renewable, sustainable, and biodegradable composite materials which are called green composites. These biodegradable and eco-friendly materials can use as an alternative to synthetic fibers and petrol chemical polymers used in traditional fiber-reinforced polymer composites [1, 2]. Biocomposites suffer from lower strength and impact resistance due to

their lower water/moisture resistance despite academic and industrial attractiveness. Muñoz and García reported that water absorption led to reducing in the flexural modulus of flax/bio-epoxy composites [3]. Chilali et al. found that the tensile strength and modulus of both flax/epoxy and flax/acrylic composites decreased after hygrothermal aging. Studies have shown that after absorbing moisture from the environment composite samples exhibited poorer mechanical properties [4].

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Natural fibers which consist of mainly cellulose and hemicelluloses highly have an affinity for water uptake since their many easily available hydrophilic hydroxyl groups [3]. The hydrophilic character can lead to swelling by water uptake of the natural fibers; thus, degradation and micro-cracks can occur in the composite. Water uptake is a significant problem that to be solved. It is known that making a stronger interface between fiber and matrix could decrease the hygroscopicity of the natural fibers. For this reason, researchers [1, 2, 4–8] have used chemical treatments and suitable coupling agents to improve the mechanical properties of the natural fiber-reinforced composites. The most used chemical treatments of the natural fibers are alkali treatment, grafting UV, silane, acrylate, permanganate, acetylation, and peroxide. Alkali treatment is one of the economic and effective methods for the surface modification of natural fibers. It also increases the mechanical properties of the composites due to enhancing interfacial adhesion between matrix and fiber. Moreover, alkali treatment decreases the hydrogen bonding on the surface of natural fibers and boosts fibers' surface roughness. The rough surface promotes bonding reaction with the hydroxyl groups on fiber surfaces and mechanical interlocking and resulting in hydrogen or covalent bonding occurring.

Natural fibers take water or moisture from the environment, being in a higher moisture uptake as the relative humidity enhances. Increasing water and moisture into fiber leads to decreases in the mechanical properties of the biocomposites. Espert et al. [9] reinforced four different fibers which are cellulose, sisal, corn, and pumpkin, into a polypropylene matrix with two different chemical compositions. In the study, maleic anhydride was applied to improve the matrix and fiber interfacial bond. To determine the water sorption properties of the composite materials, water uptake tests were carried out at 23, 50, and 70 °C. It was observed that the tensile strength of the water-uptake composites decreased significantly compared to the dry samples. Abdelmouleh et al. [10] treated cellulose fibers with silane and reinforced them into an epoxy matrix. After the silane treatment, a significant decrease was achieved in the water sorption hydrophilic property of the fibers. Soleimani et al. [11] alkaline-treated flax fiber was reinforced into the polypropylene matrix and mixed with maleic anhydride by extrusion and hot molding method. In the study, maleic anhydride and alkali treatment increased the impact strength, tensile strength, and water gain percentages of the biocomposite enhanced. Zhu et al. [12] alkali, esterification, and silane treatment were applied to natural fibers and compared the results with each other. After the alkali treatment, it was seen that the flax fiber removed the pectin and lignin in the first cell wall. It was understood that strong cross-links were formed after the silane and esterification process. After the fiber treatment processes, there was a certain decrease in the tensile properties of the flax fiber. While Duigou et al. [13] investigated the seawater resistance of PLA/flax biocomposite as a yacht material. Water gain percentages of the flax fibers reinforced composite were around 12%. They reported that there is a linear relationship between water uptake and loss of mechanical properties. When the water sorption time raised mechanical properties (Young's modulus and tensile strength) were decreased. Elias and Njuguna [14] investigated PLA/flax fiber biocomposite's thermo-mechanical properties as a body part for the automotive industry. They found that moisture content significantly affects the physical properties of flax fibres. Georgiopoulos et al. [15] produced biocomposites by adding silane-treated flax fibers to the PLA matrix by the thin-film compression

method. In the study, both water uptake and bending strength of fiber-reinforced composites with 2% by weight silane improved significantly. Moudood et al. [16] studied the influence of moisture and water absorption on the properties of flax fiber and its composites. They reported that the presence of water and moisture in the composites is responsible for decreasing the mechanical properties of the composites. Dehouche et al. [17] microcrystalline olive husk flour were reinforced into poly (3-hydroxybutyrate-co-3-hydroxyhexanoate) (PHBHHx) matrix by melt in the controlled release of insecticides with different filler content ratios which are 10, 20, and 30 wt%. they reported that water gain values of the biocomposites increase with the filler content. Pivsa-Art et al. [18] manufactured bamboo fiber-reinforced into poly(butylene succinate) biodegradable polymer matrix. Alkali treated and untreated bamboo fibers were used as reinforcement material. They reported that alkali treatment promoted reducing hydrophobic components on the bamboo fiber surface. The alkali-treated composite exhibited higher tensile strength than untreated composites. They also found that alkali treatment was provided to decrease in water sorption capacity of the fiber. Kamaludin et al. [19] produced polylactic acid/chitosan composites by melt compounding and compression molding methods. They reported that chitosan reinforced into PLA four different ratios which are 2.5, 5, 7.5, and 10% parts of polymer. The water sorption of the composite increased with chitosan ration in the matrix. They found that the tensile test results of dry composites were increased in tensile strength and tensile modulus by about 2% and 14%, respectively. Cervantes et al. [20] manufactured polyhydroxyalkanoates/agave fiber biodegradable composites using compression molding. They reported that mechanical properties such as flexural strength and flexural modulus of the biocomposites improved about 170% and 18% compared to uncompatibilized biocomposites, respectively. The water absorption resistance increased in propionylated treated fibers reducing from 6.6% to 4.4% compared with biocomposites with untreated fibers. Tensile strength and impact strength of the biocomposites also improved using treated fiber. In the literature, the mechanical, flammability, and water resistance properties of biocomposites are lower than synthetic fiber composites. However, these properties can be improved by subjecting the fiber or matrix to chemical or physical modification processes. Physical and/or chemical modification of the natural fiber and/or polymer matrix increases the mechanical properties of the composite by increasing the interface interaction between matrix and fiber [21]. In this study, ecofriendly biocomposites with and without alkali treatment and coupling agents have been fabricated with 15 wt%, 25 wt%, and 35 wt% flax ratios. These composites were immersed into the water tank to understand water gain values and the effect of water sorption on the flexural properties and microstructure of the composite.

2. Experimental procedure

2.1. Materials

Poly(lactic acid (LX 175) with 1.24 g.cm⁻³ density and a residual monomer content of < 0.3% in a granular form was bought from Total Corbion Company. Thalline flax fiber was supplied from a local market. The flax fiber was treated using NaOH (sodium hydroxide) which was an inorganic compound, supplied from Merck Company. The coupling agent maleic anhydrite was provided by DuPont

Company (Fusabond E226) with a 1.75 g/10 min melt flow rate and, 102 °C melting temperature.

2.2. Fiber surface treatment

Flax fiber was immersed in a 5 wt% sodium hydroxide solution for 1 hour to advance the adhesion between PLA matrix and flax fiber. Acidic acid was added to the solution to obtain a neutral pH state. After that, the flax fibers were cleaned with water several times to remove the residual alkali solution. Washing fibers firstly dried under the sunshine and then they placed into an electric oven at 65 °C for 12 hours to disappear the residual moisture. Alkali-treated fibers were shorted in different dimensions between 3 and 6 cm by a guillotine. Finally, the fibers' length was recut with clippers for 1 to 3 cm by manual.

2.3. Composite processing

Biocomposites were compounded using a co-rotating twin-screw extruder (Microsan Instrument, Turkey) with a 25 mm screw diameter and 30 L/D ratio at the temperatures were between 165-170-180-190 °C with 75 rpm screw speed. PLA, flax fiber and coupling agent were dried in the oven at 60 °C which is lower than the glass transition temperature of PLA, for the 12 hours prior extrusion process. And then, the flax fibers, the matrix, and the coupling agent were weighted by digital balance scale with 1 g precision according to different formulations which are given in Table 1.

The obtained compound was crushed to make the pellet form by a crusher before being fed into an injection molding. The injection molding machine (Hastek Instrument, Turkey) temperatures were 165–170–175–180–190 °C, which were the lower than thermal degradation temperature of the flax fiber, from the first nozzle to the die molding respectively. The injection molding machine was set as; screw speed for melting set in at 75 rpm (revolution per minute), molding time was 30 s, and the molding pressure was 100 bar under 50 mm.s⁻¹ injection speed.

2.4. Scanning electron microscopy (SEM)

A scanning electron microscope (SEM) was carried out to investigate the macro and micromorphology of the specimens. The SEM photographs were obtained from the impact fracture surface of the composites. Phenom-World XL SEM device was used at 15 kV to observe the interface interaction between matrix and fiber interface. Before SEM observation the samples were gold-coated by the sputtering device (Quorum 150, Netherlands) to stay away from the electrostatic charging and to get a clear examination.

Table 1. Composition of the produced composites.

Sample	Composition, PLA/flax fiber/MAH	
	vol%	wt%
PFL15	87.2/12.8/0	85/15
PFL25	78.7/21.3/0	75/25
PFL15MAH	80.8/12.5/6.7	85/15/5
PFL25MAH	71.9/21.3/6.8	75/25/5
PFL35MAH	62.8/30.3/6.9	65/35/5

2.5. Mechanical testing

A three-point bending test was applied on the specimens using a tensile testing machine (Shimadzu AG-IS) equipped with a 50 KN load cell to decide the influences of water absorption on the properties such as flexural strength of the composites. Flexural tests were applied on the composites according to the ASTM D790-17 standard. A crosshead speed was calculated as 1.75 mm.min⁻¹ for 67 mm gauge distance from the ASTM D790-17.

Unnotched impact tests were carried out according to BS EN ISO 179-1:2010 standard using a Zwick Izod impact machine with a maximum pendulum ability of 6 J. The samples notch was prepared as 3 mm. The impact strength of specimens was found using the following Eq. 1:

$$A_{cu} = \frac{E_c}{bh} \times 10^3 \quad (1)$$

where E_c (joules) is the absorbed energy by breaking, b (mm) is the width, and h (mm) is the thickness of the sample. The results were obtained as the average of at least five specimens of each sample for the mechanic tests to increase the reliability.

2.6. Water absorption

Water absorption tests were carried out on the injected flexural test samples according to ASTM D-570 test standards. Priority samples were dried in an electrical oven at 60 °C for 5 h and then were cooled to room temperature. After that samples were immediately weighed (W_d = dry weight) using a scale with an accuracy of 4 decimal places. Due to PLAs' rigid structure, the samples did not show remarkable and measurable swelling. For this reason, dimension difference was ignored to get a better result. Biocomposite samples were immersed in a water bath at room temperature until the sample reached saturation. Water uptake studies were performed for ten different water absorption times which are 24, 48, 72, 175, 200, 300, 500, 750, 1150, and 1850 hours at room temperature. After each time cycle, the samples' surface water was immediately dried using a clean dry cloth and then the samples were weighed. The water gain percentages of the biocomposites were calculated by the following Eq. 2:

$$M_t \% = \frac{W_i - W_d}{W_d} \times 100 \quad (2)$$

where M_t is the percentage of weight gains, W_i is the weight of water uptake a material at a specific time, and W_d is the initial weight of the dry biocomposites.

3. Results and discussion

3.1. Water absorption behavior

Fig. 1 shows the water absorption behavior of the biocomposites. The graph shows weight gain values of the different content of fiber biocomposites as a function of time. The result shows that all composites exhibited the same trend of water uptake behavior which is a rapid water uptake is observed in the first 72 h. After 175 h soaking graph showed a stable tendency till 750 h.

Between 175 h to 750 h the samples showed the same water uptake tendency. The water uptake regularly increased for all samples until 750 h. After 750 h PFL15MAH showed a sharply increasing difference from the others. Except for PFL15MAH water uptake values of the samples started reducing after 1150 h. As a result, the highest water

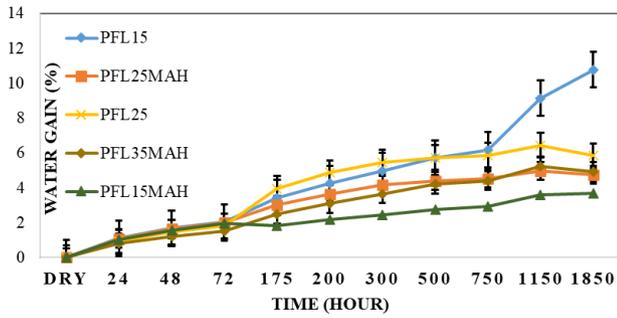


Fig. 1. Sorption behavior for biocomposites at room temperature.

uptake value which is about 11% was obtained in PFL15 samples due to it does not include alkali treatment and maleic anhydride. Untreated fibers surface includes hydroxyl groups that have a high affinity for water [22]. Therefore, percentages of water absorption of the untreated fibers are higher than alkali treated, which removes the hydroxyl groups surface of the fiber, composites. Due to, the addition of the coupling agent providing better interface interaction between fiber and matrix, the water cannot easily diffuse from surface to inside of the composites. In other words, without alkali treatment and coupling agent composite showed poor adhesion between the fiber and the matrix. As a result, water can easily diffuse from the surface to the inside of the weak interfacial composite structure [23]. As expected, the untreated composites showed a water absorption rate in direct proportion to the fiber ratio for up to 600 hours. After the 600th hour unexpectedly, although PFL15 has a lower fiber ratio than the PFL25 composite, it showed a higher water absorption tendency than PFL25. It can be explained by, there is a weak interfacial adhesion which can be seen the mechanical results and SEM pictures, between fiber and matrix the weak interfacial bond promotes water to easily transfer from the surface to the interior of the composite and causes the fibers to swell. It is most likely that the PFL15 was not mixed well during the manufacturing process, the interfacial bond adhesion to be even lower than PFL25. As a result, the fibers inside the PFL15 swelled more, breaking the fiber-matrix interfacial adhesion and opening up more

space for water uptake. Moreover, weak interfacial adhesion would be a trigger to increase the capillarity effect of the composite. As expected PFL15MAH composite with alkali treatment and coupling agent showed the lowest water uptake behavior because it had lower fiber content and coupling agent. It is explained by good adhesion fiber and matrix which results in water diffusion from surface to inside of the composite was slow down [24]. Normally the hydrophilic character of natural fibers is responsible for the water uptake in the composites, thus, a higher ratio on fibers causes a higher amount of water absorption.

3.2. Effect of water absorption on the flexural strength and impact properties

The effects of water absorption on flexural strain and strength were investigated by performing three-point bending tests of biocomposites after 1850 hours of water retention and in their initial state. In Fig. 2 and Fig. 3, the bending test results of the biocomposites at the initial state and after 1850 hours are given. After the bending test, it was found that the PFL15MAH and PFL35MAH samples had a bending strength value of approximately 50 MPa. The PFL 25MAH sample, on the other hand, has a flexural strength of approximately 38 MPa. The flexural strength of the composites without treatment (control composite) was found to be between 20 and 25 MPa. In the bending test performed after 1850 hours of water retention, the bending strength of the PFL15MAH sample decreased from 50 MPa to 30 MPa. The flexural strength value of PFL25MAH decreased from 38 MPa to around 12 MPa. A similar result was valid for the PFL35MAH sample, and the flexural strength was found to be around 15 MPa, with a sharp decrease from 50 MPa. Flexural strength PFL15MAH, PFL25MAH, and PFL35MAH showed higher flexural strength than virgin composites. As expected the addition 5 wt% MAH into PLA/flax and alkali treatment provide improving the flexural and impact strength of the biocomposite compared to virgin composites. PFL15MAH and PFL35MAH had higher flexural strength than PFL25MAH. It can be explained by several possibilities, flax fiber would be agglomeration in the composite structure. Due to the extrusion screw speed being automatically changed at the extrusion process, it might decrease the wettability distribution of biocomposites. Insufficient wettability of the

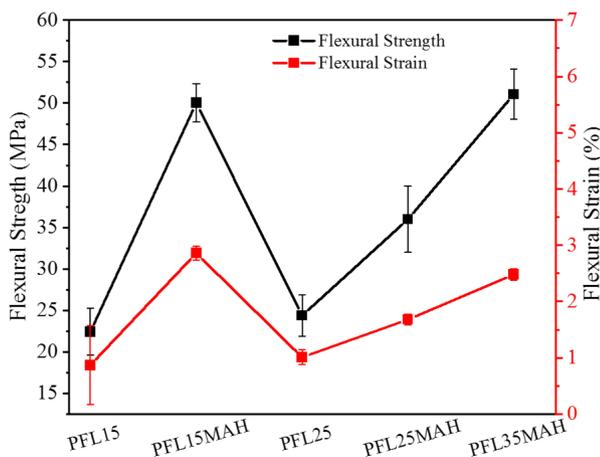


Fig. 2. Flexural properties for dry samples.

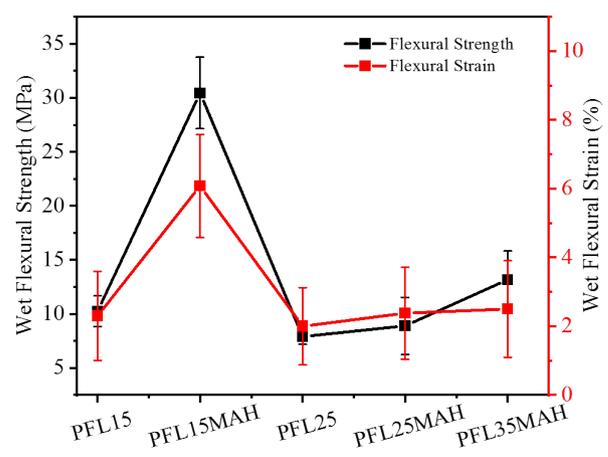


Fig. 3. Flexural properties for 1850 h water immersed samples.

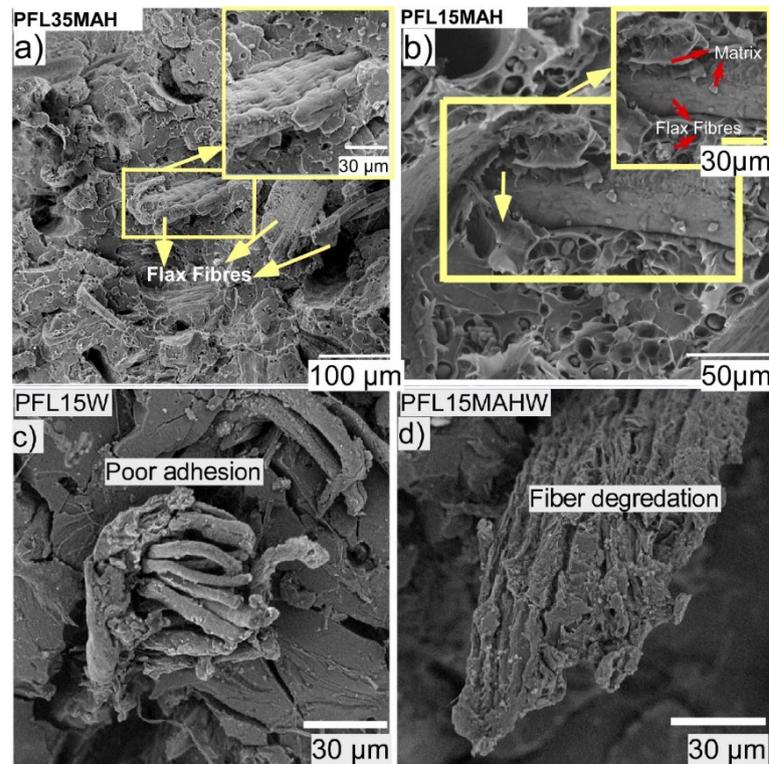


Fig. 4. SEM images of the biocomposites; a, b) injected biocomposite PFL35MAH and PFL15MAH, c) PFL15W after water uptake, and d) PFL15MAHW after water uptake.

fiber and matrix also negatively affects the biocomposite interfacial adhesion. Bax and Müssig [25] reported that decreasing would be occurring due to an inhomogeneous mixing during the biocomposite manufacturing process.

Despite the treatment processes, it is seen that the mechanical strength of the composites decreased to the same levels after being in the water for a certain time. The reason for this can be explained by the disappearance of the matrix-fiber interfacial bond, the degradation of the fibers, and the reduction of the fiber cross-section with the long-term ingress of water into the structure. While the flexural strain of the untreated samples was approximately 0.9%, the values were measured as 2% to 3% after the treatment. After the water immersion, the flexural strain of all composites increased to a certain extent. The best wet flexural strain of 6% was measured in the PFL15MAH sample. Muñoz et al. [3] reported that the flexural strain of the flax-reinforced bio epoxy matrix composite material increased after 768 hours of water retention. In the study, the authors stated the probable cause of this situation; they attributed those cellulosic fibers became more ductile after water absorption and that water molecules had a plasticizing effect on cellulose.

Fig. 4 shows SEM images of dry and water sorption samples. SEM observations were applied to observe the interface interaction between the matrix and the fiber for dry composites and the influence of the water sorption on the wet composite. Figs. 4a and 4b exhibit SEM images illustrating the influence of alkali treatment and coupling agent with perfect adhesion fiber and matrix. In Fig. 4a, the PLA is firmly hugged on the alkali-treated fibers since the coupling agent can provide the reaction between the polymeric matrix [26]. Also, alkali-treated fibers with a rougher surface can provide more reaction surface area.

The addition, the flax fibers break in line with the PLA surface and exhibit the proper load carry from the matrix to the fiber. Also, the fiber de-bonding and voids are observed in these figures due to their weak interface interaction between the fiber and the matrix. Figs. 4c and 4d show SEM images of the water sorption composite which include degradation, pull-out, and gaps. Water led to swelling of the flax fiber in the composite structure it is released to fill the gap between fiber and matrix. Thus, this effect helps to increase composite mechanical properties until a short water uptake time.

In Fig. 5, the impact strengths of biocomposites after 750 hours and

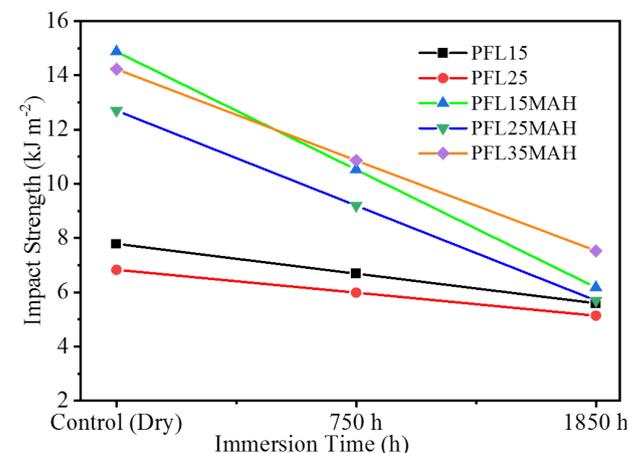


Fig. 5. Variation of impact properties due to immersion time.

1850 hours of soaking in water are compared with the impact strengths of control composites. In the test results, it is seen that the impact strengths of MAH added, and alkali-treated composites are approximately 13 to 15 kJ.m⁻². After 750 hours of water immersion, the impact strength of the control samples decreased by about 40% and was found to be 9 to 11 kJ.m⁻². While the impact strength of the samples PFL15 and PFL25, which have an impact strength of 7 to 8 kJ.m⁻², their impact strength decreased after 750 hours. In the impact tests performed after 1850 hours, the impact strengths of all samples were found to be approximately the same. The reduction in mechanical properties after water absorption can be explained by the capillary effect and the degradation of the swelling fibers. Moreover, it is known that cellulose and hemicellulose on the fiber surface accelerate the diffusion of water [27]. It is seen that the flax fibers in the composite structure swell and deteriorate (Fig. 4). SEM pictures Figs. 4c and d show that there are gaps between the fiber and the matrix and there are breaks in the fiber heads. All these factors cause a decrease in the mechanical properties by preventing the load transfer from the matrix. In general, PFL15MAH and PFL35MAH samples have the best impact strength values after water absorption.

4. Conclusions

This study investigates alkali treatment and coupling agent and water uptake on the flexural properties of the biocomposites. Produced biocomposites can be used in automotive interior parts, such as dashboard, door insulation, and children's goods such as spots-pans, plates, and dishes. The addition, these biocomposites also can be used in the constructions industry and toy car components. However, due to the decrease in mechanical properties of biocomposites after long-term water uptake, the long-term water absorption strength still needs to be improved.

- The highest water sorption value about 12% was got in the PFL15 composite which was untreated and without a coupling agent. The lowest water uptake value under 4% was obtained from PFL15MAH.
- Although alkali treatment and coupling agent increase the flexural properties up to 50% of the dry composite after a long-time water sorption alkali treatment and coupling agent did not positively affect the flexural properties.
- Using MAH decreased water gain percentages of the composite at about 65% as expected. After 750 h of soaking time, water uptake was slightly decreased for samples except for foPFL15MAH.
- SEM images showed the addition of MAH into pure PLA improved interface interaction between the polymeric matrix and the flax fiber. After long-term water sorption fiber degradation was observed.
- Flexural strain, flexural and impact strength of the composites reduced after the long-term water uptake test.

CRedit authorship contribution statement

Ali Avci: Writing – original draft, Conceptualization, Resources.

Aysegül Akdoğan Eker: Supervision, Writing – review & editing, Project administration.

Mustafa Bakkal: Supervision, Writing – review & editing, Methodology.

Mehmet Safa Bodur: Conceptualization, Data curation, Validation.

Data availability

The data underlying this article will be shared on reasonable request to the corresponding author. Data will be shared on request.

Declaration of competing interest

The authors declare no competing interests. There is no conflict of interests.

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