Effect of silanized-chitosan on flammability, mechanical, water absorption and biodegradability properties of pseudo-stem banana fiber and montmorillonite filled waste polypropylene biocomposite

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Abstract. Growing consciousness for an eco-friendly environment has revived the interest to develop composite fibers from biobased products. In this study, flammability, mechanical, water absorption and biodegradability properties of chitosan filled biocomposite waste polypropylene (wPP) reinforced with pseudo-stem banana fiber (PBF) and montmorillonite (MMt) biocomposites has been conducted investigate. It was successfully processed in solution method. Chitosan was chemically treated with glycidyloxypropyltrimethoxysilane (GPTMS) to improve interfacial adhesion between chitosan and wPP. The chitosan treated with GPTMS content in the biocomposites were varied from 0 to 7% (dry wt. basis). Flammability, tensile strength and water absorption index of biocomposites were measured according to ASTM D635, ASTM D638, and ASTM D570 respectively. To study the nature of its biodegradability, the biocomposites were technically buried in garbage dump land. The results show that the addition of treated chitosan 3-GPTMS has improved thermal properties such as Time to Ignition (TTi), Burning Rate (BR), and Heat release (HR) of treated biocomposites compared with neat PP and untreated biocomposite with treated chitosan. The treated biocomposites exhibit higher tensile strength and Young's modulus, but lower elongation at break compared with neat PP and untreated biocomposites with treated chitosan. The biocomposites show a reduction in the rate of water uptake with higher loading of CH.

1. Introduction

Nowadays, plastics especially polypropylene (PP) has been widely used in many fields such as mineral water packaging, electronic and electric industry, automobile and housing due to its good mechanical properties, excellent electrical resistance, low toxicity, low density and good chemical resistance[1-2]. However, PP is not a biodegradable polymer and makes potential hazard to the environment [3]. The degradation of waste PP (wPP) requires a long time, and most of them end up overburdening on landfill. This can make industrial waste creating disposal problems over the world [4]. Together with the other industrial wastes, the effect on the ecological equilibrium of these wastes increases every day and may reach levels to the point of threatening natural life. Therefore, the recycling of the industrial wastes is not only an attempt to decrease environmental pollution but also an effort to increase economical effectiveness. Waste polypropylene (wPP) can be transformed into another useful product such as biocomposite by adding natural fiber, which has higher economic value and saving the environment

from pollution [5]. Because of these environmental issues, a variety of biomaterial such as biocomposites has been developed. But the main drawbacks of PP are its inherent flammability and its poor melt strength limits which restrict its application in many fields for safety consideration. The PP also lacks functional groups necessary for good adhesion to synthetic or natural fillers. Consequently, the improvement of flame retardancy of PP is not only necessary but also urgent [6-9].

In the recent years, significant attention has been given to the use of natural fibers and polymer as reinforcing composite materials [10]. Both of natural fiber and chitosan can be obtained from agricultural residues that represented a disposal problem because they can lead to environmental pollution and biomass spoilage. Compared with synthetic fibers, natural fibers, and polymer are low in cost, low density, have highly specific properties and low abrasive nature. Other advantages they are environment-friendly, biodegradable, easily available [11-17]. There are many natural fibers have been used as filler or to reinforce polymer composites, such as cotton, coir, sisal, jute, hemp, sisal, bamboo, kenaf banana and chitosan [18-19]. Pseudo stem banana fiber (PBF) is one of potential candidate natural fiber for biocomposites. Al-Qureshi [20] using banana fiber mixed with epoxy resin and hardener to develop a truck model called Manaca. Corbiere-Nicollier et al. [21] have investigated physically and mechanically of banana fibercement biocomposites. It was reported that kraft pulped banana fiber biocomposite has good flexural strength. Maleque et al (2007) [22] also used pseudo-stem banana woven fabric to reinforce epoxy composite. The results show that the mechanical properties of biocomposites were increases compared to the virgin epoxy. Also, Geethama et al [23] reported that the tenacity and elastic modulus of banana fiber, and are in the range of 529-759 MPa, and 820 GPa, respectively. Due to low density, high tensile strength, high tensile modulus, and low elongation at break of banana fibers, biocomposites based on these fibers have very good potential use in the various sectors, such as automotive industries, construction materials, machinery, household applications, and various plastic products for future industrial applications [24-25]. In the other hand, natural fibers and natural polymer also have some drawbacks, such as large variations in mechanical properties, moisture absorption, and sensitivity to UV radiation, and low resistance to impact between the hydrophobic polymeric matrix and the hydrophilic fibres. However, the main loss of these natural fibre/polymer biocomposites seems to be the incompatibility between the hydrophobic PP matrix and hydrophilic natural fibres [26]. Consequently, the use of compatibilizers or coupling agents is necessary to improve the adhesion between fibre and matrix which yield biocomposites with poor properties [26-29].

Chitosan has very kind properties as a biomaterial. It is the naturally abundant polysaccharide and renewable polymers which have excellent properties such as, biodegradability, bio-compatibility, non-toxicity, and adsorption [30-32]. Due to its superior properties, chitosan has been used as reinforcement in composite materials. It also can be modified by several chemicals to make biocomposites with enhanced properties [33]. The addition of coupling agents or compatibilizers have been used to improve dispersion, adhesion, and compatibility between hydrophilic filler and hydrophobic polymer [34]. The coupling agent provides a stable bond between two otherwise poorly bonding surfaces. A coupling agent is a chemical that functions at the interface to create a chemical bridge between the reinforcement and matrix. Silanization is a convenient method that can be used to cover the biomaterials surface through the covalent binding of organofunctional alkoxysilane molecules and surface hydroxyl groups [35-36]. Glycidoxypropyltrimethoxysilane (GPTMS) and aminopropyltriethoxysilane (APTES) are among of the coupling agents and currently they are most commonly used as coupling agents. Salmah et al [36] have reported that the use of coupling agent like APTMS was able to improve the mechanical properties of polypropylene-chitosan composite. Liu et al 2005 reported that the hydrophilicity organic-inorganic chitosan membrane was able reduced by adding GPTMS [37].

Due to the lack of information about the thermal, physical and mechanical properties of some biocomposites materials, in the present study, the feasibility of using GPTMS to silanize chitosan was evaluated. The effect of the chemical modification of chitosan with GPTMS on the flammability, mechanical properties, water absorption and biodegradability properties of PP/SPP/MMt/chitosan biocomposites were measured. Characterization was analyzed by Fourier transform infrared (FTIR). Examinations with Universal testing machine (UTM), and flammability test were conducted to

investigate the physical and thermal properties of PBS/CF biocomposites. Biodegradability was conducted by burial test method.

2. Experimental

2.1. Materials
Raw material waste polypropylene (wPP) was obtained from waste drinking water cup. It was prepared by cleaning the waste drinking cups with ethanol and chopped in 2x2 cm^2. The PBF used was obtained from Banana agricultural field in Salatiga, Central of Java, Indonesia. It was pre-treated by delignification with NaOH 4% (w/v). Then, it was cleaned with ethanol, dried in oven at 40 °C, and grounded into 100 mesh particle size. Chitosan was obtained from PT. Biotech, Indonesia with an average size of 80 mm with the degree of deacetylation (DD) of 88%. The silane coupling agent, γ-glycidyloxypropyltrimethoxysilane (GPTMS), Maleic Anhydride and montmorillonite were obtained from Sigma-Aldrich, Singapore. Acetic acid, Natrium Hydroxide, and ethanol were purchased from Merck. The distilled water was supplied from Chemistry Laboratory of Sebelas Maret University. The formulation of untreated and treated PP/PBF/MMt/chitosan biocomposites with GPTMS in this study is shown in Table 1.

Table 1. The formulation of untreated and treated PP/PBF/MMt/chitosan biocomposites

Ingredients (php)	Biocomposites					
	W1	**W2**	**W3**	**W4**	**W5**	**W6**
wPP	60	60	60	60	60	60
PP-g-MAH	-	10	10	10	10	10
PBF	-	20	17	17	15	13
CH	-	-	3	-	-	-
CH-GPTMS	-	-	-	3	5	7
MMt	-	10	10	10	10	10

2.2. Chitosan modification
The coupling agent GPTMS was diluted in ethanol to make 20% solution. The amount of GPTMS used was 3% by weight of filler. The filler was charged into a mixer and solution was added slowly to ensure uniform distribution of GPTMS. After completion of the GPTMS addition, the filler was continuously mixed for another 30 min. The treated filler was then dried at 100 °C for about 5 h to remove residual acetic acid.

2.3. Preparation of pp/chitosan biocomposites
The synthesis of wPP/wPP-g-MAH/PBF/MMt/CH and wPP/wPP-g-MAH/PBF/MMt/CH-GPTMS biocomposites were reactively processed using benzoyl peroxide (BPO) in solution method using xylene as solvent. The formulation of biocomposites synthesis can be seen in Table 1.

2.4. Characterization and testing of biocomposites

2.4.1. FTIR analyze. Fourier transform infrared (FTIR) spectroscopy analysis of untreated and treated chitosan were carried out with a FTIR (Fourier Transform Infra Red) IRPrestige-21 SHIMADZU Spectroscopy to determine functional group. The biocomposites sample was dispersed in dry KBr powder and was ground to obtain fine particles. The KBr technique was applied. The scanned range was 400-4000 cm^{-1}.

2.4.2. Flammability properties. The flammability of the wPP and biocomposites were tested according to ASTM D635 to determine their time for ignition (TTi), burning rate (BR), and heat release (HR).
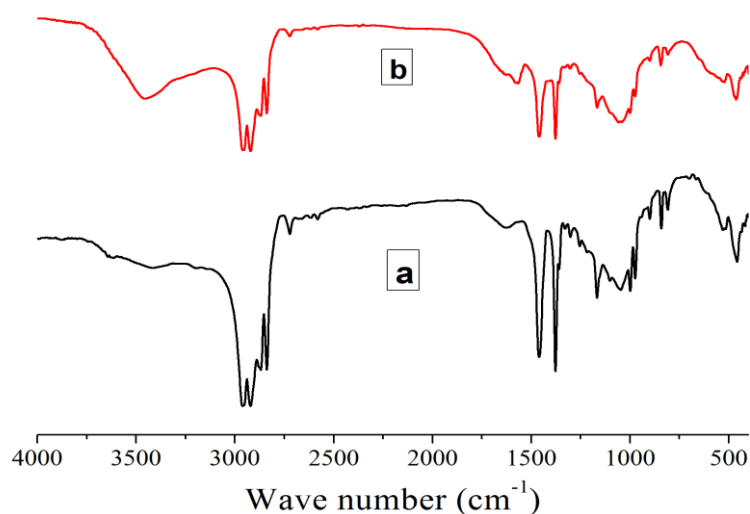
2.4.3. Mechanical properties. Tensile strength (TS), tensile modulus (TM), and elongation at break (Eb) of the films were measured by using Universal Testing Machine (model H5KT, with a 1 kN load cell, Tinius-Olsen, Horsham, USA). The samples were cut using ASTM procedure D 638-99. Tensile tests were carried out according to ASTM D-638 type IV.

2.4.4. Water absorption and biodegradability properties. Water absorption studies were performed following the ASTM D 570-95 standard.The biocomposites films were soaked in aquadest for 24 h. The samples were taken out periodically, wiped out the water on the surface of the sample, and weighed immediately to find out the content of water absorbed. Biodegradability was determined by burial test method with cellulolytic bacteria enriched with garbage soil for two months. Soil used in the test was 1:1 mixture of black soil and leaf mold for gardening. Then it was measured the weight loss (LW) of the biocomposites after biodegradation process.

3. Result and discussion

3.1. Material characterization
Chemical analysis of biocomposites and its modification were conducted using FTIR. This technique is very important to analyze the functional group of each material. Moreover, it was also used by the researcher for proving the chemical interaction in compounding.
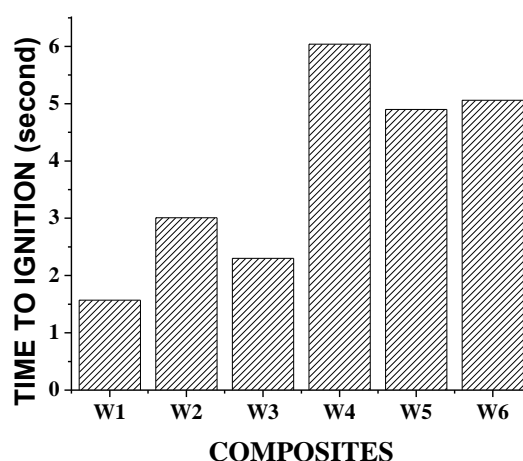

Figure 1. Infra-red spectrum of (a) wPP/wPP-g-MAH/PBF/MMt,
and (b) wPP/wPP-g-MAH/PBF/MMt/CH-GPTMS

Figures 1. (a) and (b) exhibits the FTIR spectrum for wPP/wPP-g-MAH/PBF/MMt and wPP/wPP-g-MAH/PBF/MMt/CH-GPTMS biocomposites respectively. It can be seen that wPP/wPP-g-MAH/PBF/MMt (figure 1a) biocomposites exhibit the main characteristic peaks at 3405 cm^{-1} that is corresponded to stretching vibration of hydroxyl group, 2864 cm assigns to C–H stretch vibration, 1633 cm^{-1} associated to C=O stretching vibration of carbonyl groups, 2723 cm^{-1} typicals of PP; 2843, 1446, 1369 and 996 cm^{-1} from methine, methylene and methyl groups [38]. In IR spectrum of wPP/wPP-g-MAH/PBF/MMt/CH-GPTMS biocomposites (figure 10 b) shows the FTIR spectra of treated chitosan with GPTMS. The appearance of a new absorption peak at lower frequency 1627 cm^{-1} corresponds to the –NH$_2$ bending vibration. The peak intensity at 3418 cm^{-1} increases to 3450 cm^{-1}, due to the hydrogen bonding interaction between hydrolyzed amino silane and the –OH group in chitosan. This is further supported by the decreased absorbance for –CH$_2$ bending vibration 1466 to 1459 cm^{-1} which is due to
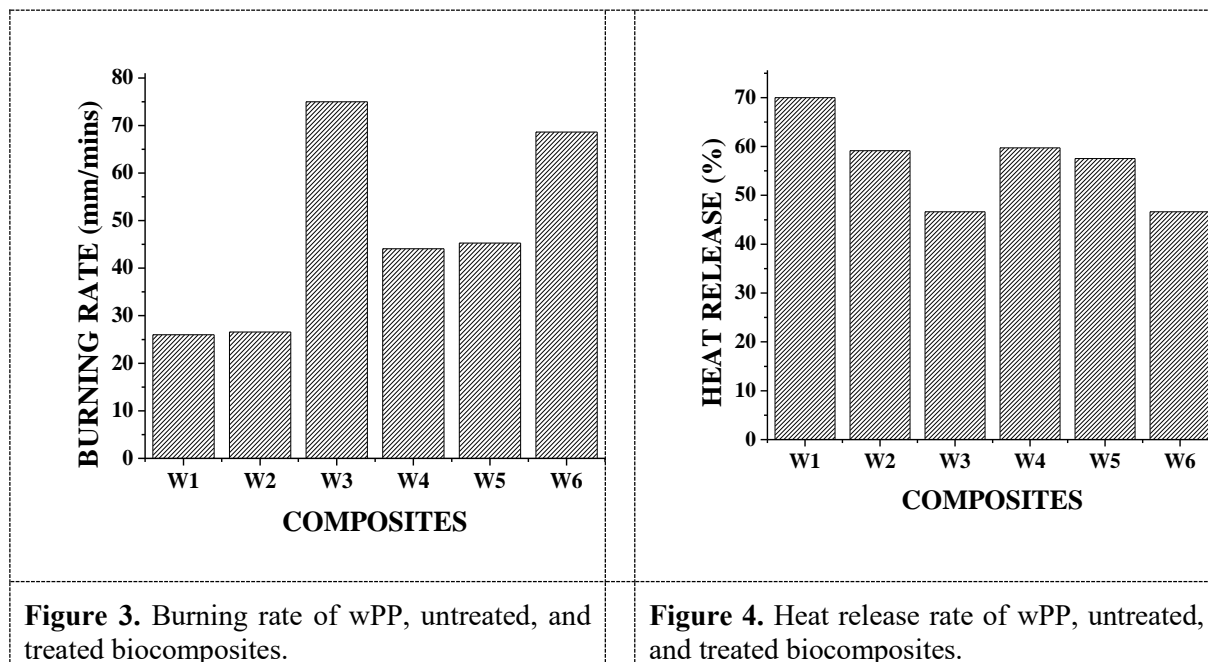
interaction between the amine group of chitosan and OH group of GPTMS. The increasing at peak 1098 to 1163 cm^{-1} indicates the GPTMS has bonded to the chitosan.

3.2. Flammability properties
Figures 2, 3, and 4 exhibit the flammability properties of wPP, wPP/wPP-g-MAH/PBF/MMt composites without chitosan, wPP/wPP-g-MAH/PBF/MMt/CH without the coupling agent, and the wPP/wPP-g-MAH/PBF/MMt/CH composites with coupling agent. Results in figure 2 indicate that of the fillers studied, the addition of 3 php treated CH with GPTMS was the most successful in inhibiting combustion delaying the time to ignition from 1.57 to 6.1 sec and also reducing the HR (figure. 4) from 70 to 59 % for W1 and W4 respectively. The highest TTI, the lowest BR, and HR indicate that the material composites have poor flammability [39]. Both of biocomposites without (W2) and with untreated chitosan (W3) have a higher flammability than biocomposite with treated chitosan (W4, W5, W6). It indicates that the addition of 3 php CH to the wPP increased the flammability about 188%. But, the addition of the 3 php treated CH with coupling agent, GPTMS, affected the flammability when compared to the biocomposites with and without the coupling agents. It could imply that the coupling agents affected the dispersion of the fillers in the polymer matrix. The optimum formula reached at loading of 3% chitosan.

Figure 2. Time to ignition of wPP, untreated, and treated biocomposites.
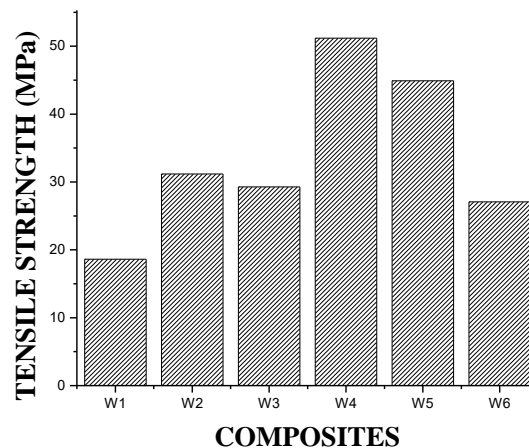
Figure 3. Burning rate of wPP, untreated, and treated biocomposites.

Figure 4. Heat release rate of wPP, untreated, and treated biocomposites.

3.3. Mechanical properties

The main aspect that affects the mechanical properties of the fiber reinforced material is the fiber-polymer matrix interfacial adhesion. Figure 5 exhibits the effect of filler loading on the tensile strength of untreated and treated wPP/wPP-g-MAH/PBF/MMt/CH composites. Compared with wPP/wPP-g-MAH/PBF/MMt composite (W2), the formula W3 has lower TS. The reduced value of tensile strength is due to poor interfacial bonding between hydrophilic filler and hydrophobic polymer matrix [36]. But, in the same composition at 3 php CH (treated with GPTMS), the tensile strength of composite W4 was increased until 75 %. These results exhibit that with the increasing of filler content the tensile strength of composites decreased. This might be due to poor interfacial bonding between filler and matrix polymer at higher filler content. Chemical modification of chitosan by silanization with GPTMS was carried out to reduce the weak bonding between filler and polymer matrix. It also can improve tensile strength of the composites. This is allowed, because that the addition of GPTMS can form a good siloxane network, so it has a high tensile strength due to strong chemical interaction between the matrix of chitosan by the presence of a cross-linked network of the silane groups [37]. The chemical treatment of chitosan with GPTMS was proven effective in enhancing the dispersion, adhesion, and compatibility of a system consisting of hydrophilic chitosan and hydrophobic matrix. It can be seen that the tensile strength of treated composite is higher compared to untreated composites. However, the increasing filler loading of chitosan decreased the TS of W5 and W6 biocomposites. It is because of the poor interfacial bonding between the hydrophilic filler and the hydrophobic matrix polymer.
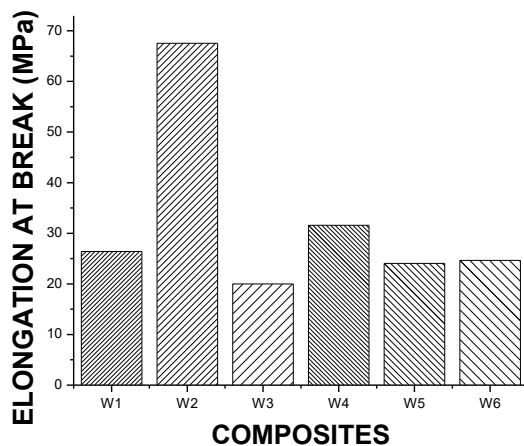
The results of elongation at break of wPP and biocomposites at different filler loading are depicted in Figure 6. It can be seen that chitosan addition (W3) decreased of % E until 238 %. This is because the addition of chitosan reduced the mobility and increased the brittleness of the composites. But in the same composition, the addition of 3 php treated chitosan (W4) with GPTMS can lead into increased of % E until 58 % when compared with biocomposite without coupling agent (W3). The modification with GPTMS has increased the tensile strength of biocomposites with the enhancement in rigidity and decrement of the ductility of composites, which consequently lowered the elongation at break of the composites [36].

Figure 7 exhibits the effect of filler content on Young's modulus of wPP, wPP, wPP/wPP-g-MAH/PBF/MMt composites without chitosan, wPP/wPP-g-MAH/PBF/MMt/CH without the coupling agent, and the wPP/wPP-g-MAH/PBF/MMt/CH composites with coupling agent. It can be seen that biocomposite without chitosan (W2) has lower MY than wPP. But with the presence of chitosan treated
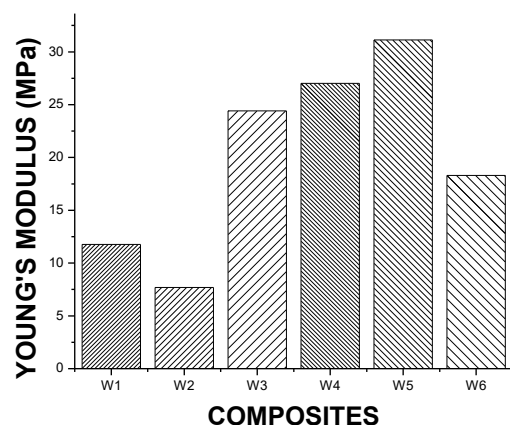
with GPTMS can lead into increasing of MY. This is because increasing the filler content significantly increases the stiffness of composites [36]. The Young's modulus of treated wPP/wPP-g-MAH/PBF/MMt/CH composites with GPTMS is higher than untreated composites. This indicates that the presence of GPTMS has enhanced the compatibility between chitosan-polypropylene.

Figure 5. The tensile strength of wPP, untreated, and treated biocomposites.

Figure 6. Elongation at Break of wPP, untreated, and treated biocomposites.
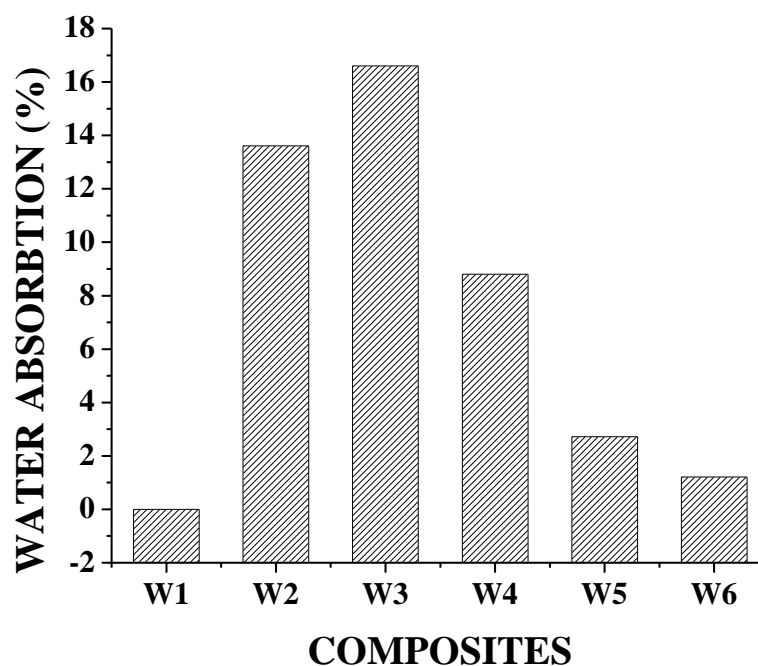

Figure 7. Young's Modulus of wPP, untreated, and treated biocomposites.

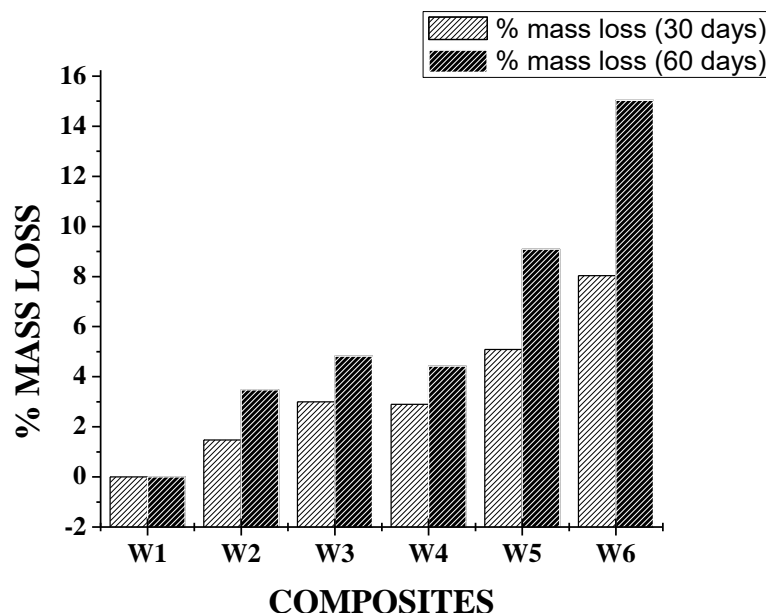*3.4. Water absorption and biodegradability*

Poor resistance of the fibres to water absorption can have undesirable effects on the mechanical properties and the dimensional stability of the biocomposites. Water not only interacts with the matrix physically, chemically, or regarding plasticization, but also attacks the fiber/matrix interface, where it can cause de-bonding [41]. It is very important to study in detail about the water absorption behaviour to estimate not only the consequences that the water absorbed may have but also how this water uptake can be minimized in some ways. The amount of water absorbed in the biocomposites was calculated by the weight difference between the samples exposed to water and the dried samples. Figure 8 shows the percentage of water absorbed for all the samples. The matrix had little effect on the amount of water absorbed as wPP demonstrated around the same water content, still 0.0 % after 24 h of exposure.

However, both biocomposites with (W3) and without (W2) chitosan have higher water absorbed, 16 and 13 % respectively. It indicates that the chitosan addition increased the hydrophilic character on biocomposites. The hydrophilic character of natural fibers is responsible for the water absorption in the biocomposites. Therefore, a higher content on fibers leads to a higher amount of water absorbed. But, the presence of treated chitosan with GPTMS decreased higher amount of water absorbed. It has better resistant to water both in long-term immersion in water. This is because of the coupling agent like GPTMS can build chemical bonds (silanol bonds) and hydrogen bonds, which reduce the moisture-caused fiber-matrix de-bonding [42].

In general, degradation is defined as a detrimental change in its appearance, mechanical, physical properties, and chemical structure of a molecule [43]. The results of biodegradability test can be seen in Figure 9. The presence of PBF and CH can improve the degradation properties. It is indicated that PBF and CH improved the degradation of the biocomposites. As compared to the biocomposites with (W4) and without (W3) silane treatments, the untreated biocomposite showed a slightly higher degradation rate than the treated biocomposite. The improved interfacial adhesion between PBS matrix and CF caused by the silane coupling agent reduces the possibility of water and microorganisms or enzymes (i.e., cellulase and lipase) to penetrate the samples [44], thus resulted in the lower degradation rate.

Figure 8. % Water absorption of wPP, untreated, and treated biocomposites.

Figure 9. % Mass loss of wPP, untreated, and treated biocomposites.

4. Conclusion

The application of y-glycidoxypropyltrimethoxy-silane as coupling agent enhances the fiber-matrix adhesion because silanol bonds and hydrogen bonds are provided. This leads to increase the interfacial interaction between filler particles and resin matrix which in turn improves the physical and mechanical properties. The results of the present study showed that the presence of GPTMS on biocomposites can reduce the interfacial energy and homogenize the polar chitosan with the non-polar synthetic polymer phase. Chemical modification of chitosan with GPTMS has improved the thermal stability properties both of wPP/wPP-g-MAH/PBF/MMt/CH biocomposites and wPP. It can also improve the tensile strength and Young's modulus, but reduce the elongation at break of biocomposites. The thermal properties show that the presence of GPTMS has increased the thermal stability of wPP/wPP-g-MAH/PBF/MMt/CH biocomposites. The effect of chemical modification of chitosan with GPTMS was obvious from FTIR analysis. The hydrophilic character of natural fiber is also reduced by adding GPTMS. Additionally, the additions of GPTMS on chitosan is also able to reduce the hydrophilic character of chitosan enhancing its adhesion with a hydrophobic wPP matrix and led to improved mechanical and thermal properties of the wPP/wPP-g-MAH/PBF/MMt/CH biocomposites.

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