

## Dynamic Mechanical Properties of Natural Fiber/Polymer Biocomposites: The Effect of Fiber Treatment with Electron Beam

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**Abstract:** Environmentally friendly biocomposites were made using plant-based natural fibers, such as henequen and kenaf. The natural fiber reinforced polypropylene (PP) and unsaturated polyester (UP) biocomposites were examined in terms of the reinforcing effect of natural fibers on thermoplastic and thermosetting polymers. Kenaf (KE) and henequen (HQ) fibers were treated with an electron beam (EB) of 10 and 200 kGy doses, respectively, or with a 5 wt% NaOH solution. Four types of biocomposites (KE/PP, HQ/PP, KE/UP and HQ/UP) were fabricated by compression molding and each biocomposite was characterized by dynamic mechanical analysis and thermogravimetric analysis. The kenaf fiber had the larger reinforcing effect on the dynamic mechanical properties of both PP and UP biocomposites than the henequen fiber. The highest storage modulus was obtained from the biocomposite with the combination of UP matrix and 200 kGy EB treated kenaf fibers.

**Keywords:** natural fiber, biocomposites, kenaf, henequen, electron beam treatment, dynamic mechanical properties.

### Introduction

Nowadays, natural fibers such as hemp, kenaf, flax, and henequen have increasingly been utilized as reinforcements with polymers. Compared with the conventional reinforcement such as glass fiber or carbon fiber, these natural fibers of the composites have benefits such as low density, low price, biodegradability, etc. The mechanical properties of these biocomposites can be comparable to those of the polymer composites reinforced with glass fibers.<sup>1,2</sup> On the contrary, a main disadvantage of natural fibers is poor interfacial bonding between hydrophilic natural fibers and hydrophobic polymer matrix.<sup>3</sup> Thus, fiber surface modification methods have been developed to improve the interfacial bond strength between the natural fibers and matrix of the biocomposites.

The electron beam irradiation (EBI) has been applied for various polymers and fibers.<sup>4,5</sup> The EBI is fast and clean process and also induces cold modification and changes of

the mechanical properties of fibers.<sup>6,7</sup> Recently, we have reported the EBI effect on dynamic mechanical properties of kenaf (KE) reinforced polypropylene (PP) or henequen (HQ) reinforced unsaturated polyester (UP) biocomposites.<sup>8,9</sup> The result revealed that the EB treatment done on the natural fiber surfaces with certain doses significantly contributes to improving the interfacial and mechanical properties and the thermal stability of natural fiber/polymer biocomposites.

Several researchers have also reported the improvement of the mechanical properties of cellulose fibers when alkalinized at different NaOH concentrations.<sup>10,11</sup> Most of impurities existing on the fiber surfaces and part of the amorphous cellulose component could be removed by alkali treatment. It was reported that the alkali treatment changed the chemical composition and rearranged the crystallinity of natural fibers.<sup>12,13</sup>

Polypropylene as one of the most popular thermoplastic polymers provides many advantages like its low cost, recyclability, and high thermal stability.<sup>14,15</sup> Unsaturated polyester has been a popular thermosetting polymer resin utilized

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in conventional glass fiber reinforced polymer composites. It has been widely used due to its excellent processability and fast crosslinking reaction as well as good mechanical and chemical properties when fully cured.<sup>16</sup>

In this study, four different types of biocomposites (KE/PP, HQ/PP, KE/UP and HQ/UP) were fabricated using natural fibers treated at different electron beam and alkali conditions showing the optimum performance with each treatment (EB 10, 200 kGy, and NaOH 5%), which resulted from our previous research. Most of the work was focused on investigating the dynamic mechanical properties of these biocomposites in order to find out the best combination between polymer and natural fibers based on the dynamic mechanical response of biocomposites depending on the fiber surface treatment. In addition, the effect of the EBI and alkali treatments of natural fibers on the thermal stability of biocomposites was examined to support the dynamic mechanical result.

## Experimental

**Materials.** Kenaf (*Hibiscus Canabinus L.*) fibers in the 60–70 mm filament form were supplied from Bangladesh. Henequen (*Agave fourcroydes*) fibers were obtained from Cordmex, S.A. of Merida, Yucatan, Mexico. Unsaturated polyester (UP-GR235) resin was supplied from Sewon Chem. Co., Korea.<sup>17</sup> It has the styrene content of 35 wt%. Methyl ethyl ketone peroxide (MEKP) was used as catalyst. Polypropylene fibers were supplied from Kolon Glotech Co., Ltd.<sup>18</sup> The specific gravity and the melting point of PP fibers were 0.91 and 160–165 °C, respectively.

**Fiber Treatment.** Natural fibers were irradiated with EB dosages of 10 and 200 kGy, respectively. The EB processing was successfully conducted at EB-Tech Co., Korea.<sup>19</sup> For alkali treatment, the natural fibers were washed and soaked in NaOH solution of 5 wt% for 1 h. Then, the treated fibers were washed with distilled water, neutralized with 2 wt% acetic acid, washed with distilled water again, and then finally dried at room temperature for 3 days. The raw fibers without any treatment were also used for comparison.

**Fabrication of the Natural Fiber/PP Biocomposites.** The fiber contents and the chopped fiber lengths in natural fiber/PP biocomposites were 30 wt% and 1 cm in average, respectively. Prior to biocomposite fabrication by compression molding, polypropylene fibers were uniformly mixed with the chopped natural fibers. Then the mixture was placed in a stainless steel mold and melted with a heating rate of 4 °C/min for 40 min, holding at the pre-determined temperature (175 °C for kenaf and 165 °C for henequen) for 20 min, applying 1,000 psi for 10 min. The mold containing the biocomposite plaque was cooled down to 40 °C by circulating cold water around the mold. A pressure of the mold was applied about 1,000 psi during cooling. After biocomposites were ejected from the mold, and cooled down to

ambient temperature, finally, the dimensions of KE/PP and HQ/PP biocomposites were 50 mm × 50 mm × 1.8 mm.

**Fabrication of the Natural Fiber/UP Biocomposites.** The fiber contents and chopped fiber lengths were 30 wt% and 1 cm, respectively in natural fiber/UP biocomposites. The KE/UP and the HQ/UP biocomposites were fabricated by compression molding technique using a hot-press (Carver 2518). The molding compounds were heated with a heating rate of 2 °C/min. A pressure of 1,000 psi was applied throughout the molding. The compound at 40 °C was primarily cured and then maintained at 70 °C for 1 h. The mold was cooled down to room temperature. Finally the biocomposites were fully cured at 70 °C for 24 h in a vacuum oven.

**FT-IR Analysis.** Fourier transform Infrared spectra were acquired using a FT-IR-610 spectrometer (Jasco Co.). The natural fibers were mounted on an ATR accessory and the ATR-FTIR spectra were obtained with an accumulation of 128 scans and with a resolution of 4 cm<sup>-1</sup>.

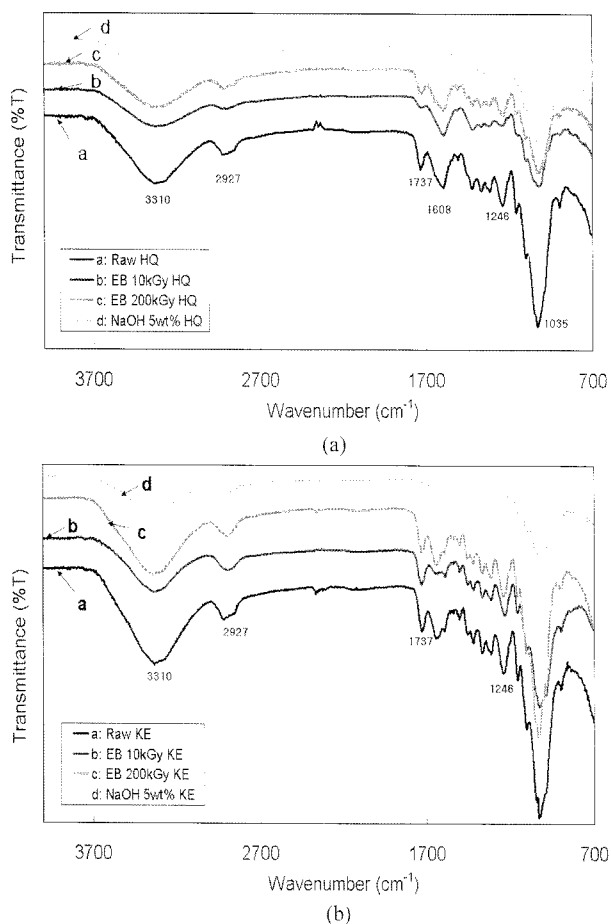
**X-ray Diffraction Analysis (XRD).** Crystalline structure of the natural fiber was analyzed by X-ray diffractometer (Rigaku-D/MAX 2000, Ultima<sup>+</sup>, Japan) with 5°/min scan speed and Ni-filtered Cu K $\alpha$  radiation ( $\lambda = 1.54 \text{ \AA}$ ) at 30 kV and 20 mA.

**Dynamic Mechanical Analysis (DMA).** Dynamic mechanical analysis of biocomposites was performed with DMA (Q800, TA Instruments) with a single cantilever mode at a fixed frequency of 1 Hz purging liquid nitrogen. The oscillating amplitude used was 0.2 mm. The heating rate was 5 °C/min. Before each measurement, the DMA instrument was calibrated to have the correct clamp position and clamp compliance. The specimen dimensions were 35 mm × 12 mm × 1.8 mm. The DMA experiments were carried out in the temperature range from -30 to 100 °C for PP biocomposites and from 30 to 200 °C for UP biocomposites, respectively.

**Thermogravimetric Analysis.** The thermogravimetric analysis of biocomposite was examined with a thermogravimetric analyzer (TGA Q500, TA instruments). The thermal stability of the biocomposites was analyzed in the range of 25–500 °C under the nitrogen atmosphere. The heating rate was 5 °C/min.

## Results and Discussion

**FT-IR Analysis.** Figure 1 shows FT-IR spectra of raw, EB and alkali treated natural fibers. The assignment of FT-IR spectra of cellulose is summarized in Table I. No significant change was observed in the spectra between raw and EB 10, 200 kGy treated natural fibers. On the contrary, the natural fiber undergoes very different chemical changes by treating with 5 wt% NaOH solution. The intensity of the absorption peaks of 3200–3400 cm<sup>-1</sup> from O-H stretching bond and 1373 cm<sup>-1</sup> from O-H in-plane bending was reduced because of the formation of glycoside bonding.

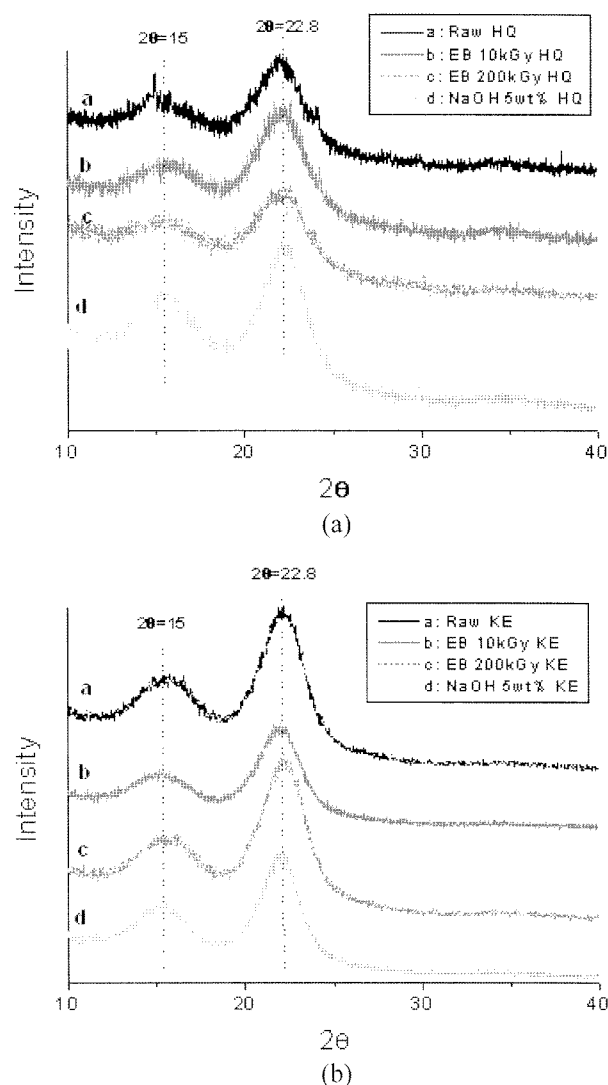


**Figure 1.** FT-IR curves of raw and treated natural fibers: (a) HQ fibers and (b) KE fibers.

**Table I. Assignment of FT-IR Spectra of Cellulose**

Wave Number (cm <sup>-1</sup> )	Assignment
3200-3400	O-H Stretching Bond
2700-3000	Typical CH <sub>2</sub> & CH
1740	Acid Carbonyl Adsorption
1735	Carbonyl Group(-O-C=O-) Stretching
1550-1650	COO' Stretching
1426-1430	CH <sub>2</sub> Bending
1365	CH Bending
1355	O-H in Plane Bending
1315	CH <sub>2</sub> Wagging
1245	C-H
1000-1500	Aromatic Region

Also, the peak of 1735 cm<sup>-1</sup> from carbonyl group stretching was reduced due to hemicellulose removal. The decrease of 1608 cm<sup>-1</sup> band can be explained by the removal of OH bending in the absorbed water molecules by alkalization.<sup>20</sup>



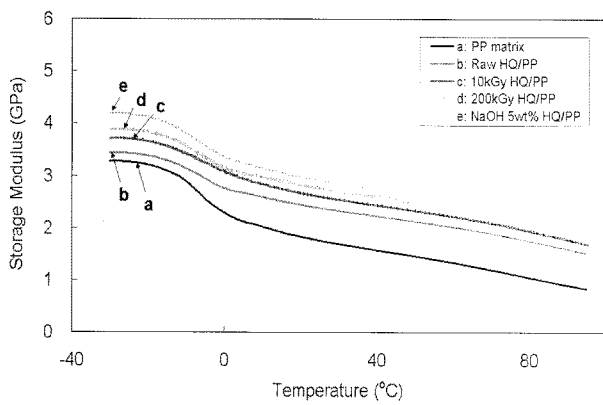
**Figure 2.** XRD curves of raw and treated natural fibers: (a) HQ fibers and (b) KE fibers.

**X-ray Diffraction Analysis.** Figure 2 compares the X-ray diffractograms of the natural fiber, EB and NaOH treated fibers. The raw natural fiber shows two distinct peaks with maximum value at 15 and 22.8 degree. The X-ray spectra of the EB and NaOH treated natural fibers was observed at the same peak position of raw natural fiber.<sup>21</sup> It means that the EB and alkali treatment cannot significantly affect on the crystalline properties of cellulose.

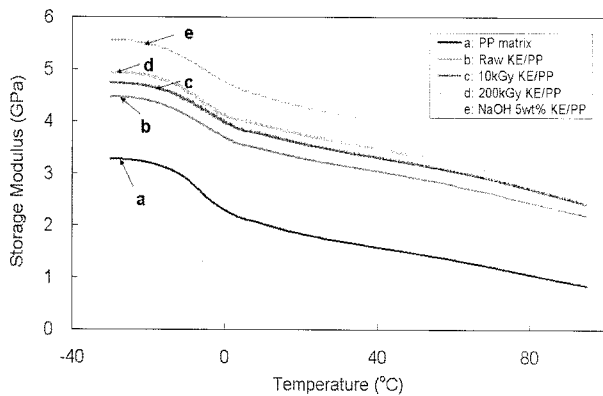
**Dynamic Mechanical Properties.** Dynamic mechanical analysis has been a useful tool for understanding the interfacial characteristics of polymer composites.<sup>22</sup> DMA measurements conducted over a wide range of temperature may inform the viscoelastic behavior of melted polymer systems, in particular on the glass transition phenomenon in fiber-reinforced polymer composites. The temperature-dependent dynamic mechanical characteristics such as storage modulus  $E'$ , loss modulus  $E''$  and energy dissipation factor  $\tan \delta$

**Table II. Comparison of Storage Modulus of Natural Fiber/PP Biocomposites**

	PP Matrix	HQ/PP Biocomposites			
		Raw (HQ/PP)	10 kGy (HQ_EB10/PP)	200 kGy (HQ_EB200/PP)	NaOH 5% (HQ_AK5/PP)
Storage Modulus (GPa)	3.27	3.42	3.71	3.88	4.15
Increase (%)	0	5	13	19	27
	PP Matrix	KE/PP Biocomposites			
		Raw (KE/PP)	10 kGy (KE_EB10/PP)	200 kGy (KE_EB200/PP)	NaOH 5% (KE_AK5/PP)
Storage Modulus (GPa)	3.27	4.47	4.74	4.94	5.59
Increase (%)	0	37	45	51	71



(a)



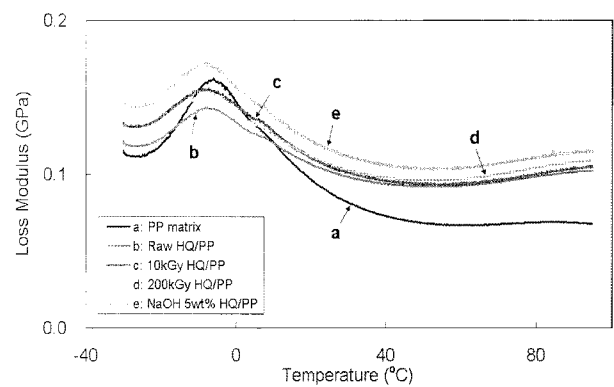
(b)

**Figure 3.** Storage modulus of natural fiber/PP biocomposites: (a) HQ/PP and (b) KE/PP.

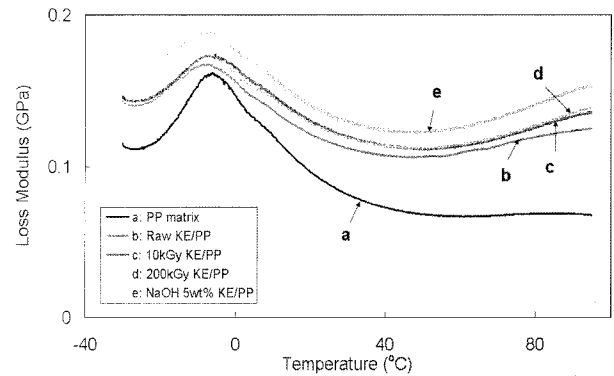
provide an insight into the level of interactions between the polymer matrix and the fiber reinforcement.

#### Natural Fiber/PP Biocomposites.

**Storage Modulus:** Table II (at -30 °C) and Figure 3 show the storage moduli of the PP matrix (PP), the biocomposites reinforced with raw henequen (HQ/PP) or kenaf fiber (KE/PP), in which the biocomposites were reinforced with 10 kGy (HQ\_EB10/PP, KE\_EB10/PP) or 200 kGy (HQ\_EB200/PP, KE\_EB200/PP) treated henequen or kenaf fibers and the biocomposites (HQ\_AK5/PP, KE\_AK5/PP) were reinforced with 5 wt% NaOH treated fiber. The storage moduli of the



(a)

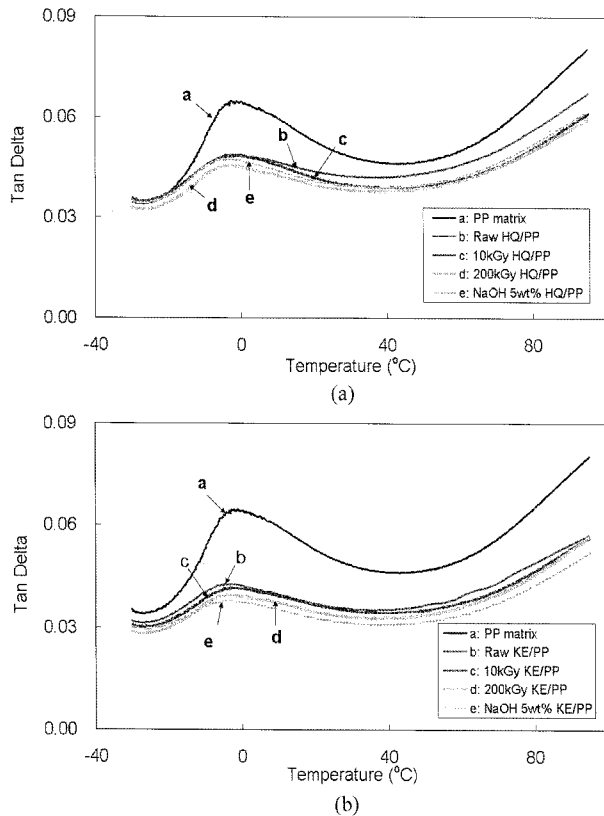


(b)

**Figure 4.** Loss modulus of natural fiber/PP biocomposites: (a) HQ/PP and (b) KE/PP.

HQ/PP biocomposites increased with treatment of the EB (HQ\_EB10/PP: 13%, HQ\_EB200/PP: 19%) and 5 wt% NaOH (HQ\_AK5/PP: 27%), respectively compare to the PP matrix (3.2 GPa) at -30 °C. Also, the storage moduli of the KE/PP biocomposites (KE\_EB10/PP: 45%, KE\_EB200/PP: 51% and 5 wt% NaOH (KE\_AK5/PP): 71%) increased with these treatment, and the amount of modulus increase in the KE/PP biocomposites was higher than that of the HQ/PP biocomposites. The highest storage modulus of biocomposite was obtained with the combination of PP and 5 wt% NaOH treated kenaf fibers.

**Loss Modulus:** The loss modulus was also increased with



**Figure 5.** Tan delta of natural fiber/PP biocomposites: (a) HQ/PP and (b) KE/PP.

the incorporation of natural fibers, as shown in Figure 4. When a composite is, in general, subjected to external stress, the energy may be dissipated by friction between fiber-fiber and fiber-polymer interactions. Additionally, similarly to the storage modulus result, when increasing temperature, the curves tend to converge and exhibit the intrinsic properties of PP matrix. The  $\alpha$  transition peak shown in  $E''$  around  $-5^\circ\text{C}$  is thought to be related to the glass transition of PP molecules.<sup>22</sup> The biocomposites reinforced with natural fibers showed a broader peak than PP matrix. The broadening of the loss modulus peak with in the biocomposites may be due to the increase in the energy absorption by the natu-

ral fibers therein. Thus, the molecular motion due to PP in the biocomposite was more restricted than that due to without the fibers.<sup>23,24</sup>

**Tan  $\delta$ :** The variation of  $\tan \delta$  as a function of temperature for the PP and the biocomposites is shown in Figure 5. The  $\tan \delta$ , which is the ratio of the loss modulus to the storage modulus  $E''/E'$ , was observed to be high for the PP matrix due to the gradual reduction of the storage modulus with increasing temperature. The incorporation of the reinforcing natural fibers restricted the mobility of PP molecules, increased the storage modulus and reduced the viscoelastic lag between the stress and the strain. As a result, the  $\tan \delta$  intensity was decreased in the biocomposites, as similarly found from other reports.<sup>25-27</sup> The  $\tan \delta$  values were lowered in the biocomposites compared to the PP matrix because there was less matrix by volume to dissipate the vibrational energy.<sup>28</sup>

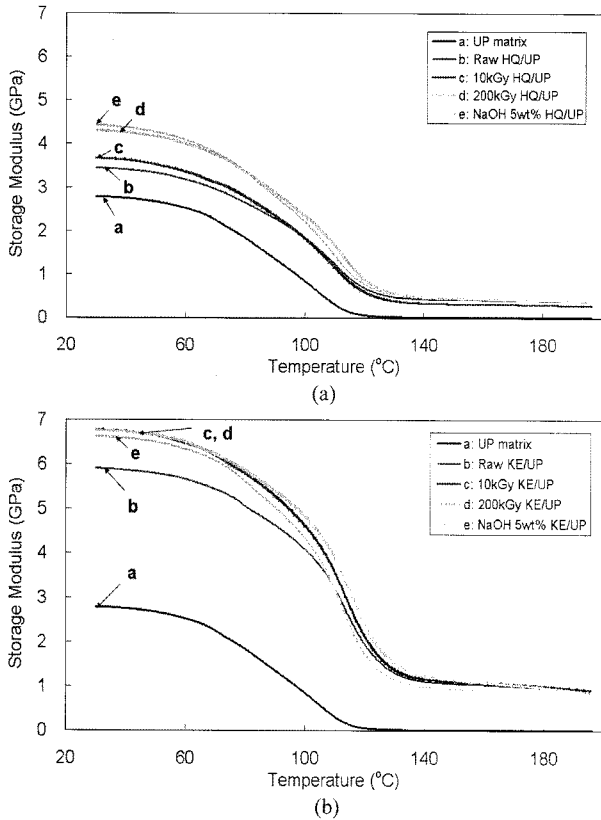
#### Natural Fiber/UP Biocomposites.

**Storage Modulus:** Table III and Figure 6 show the storage moduli of the UP and reinforced UP biocomposites of the raw fiber, 10 kGy (HQ\_EB10/UP, KE\_EB10/UP), 200 kGy (HQ\_EB200/UP, KE\_EB200/UP), and 5 wt% NaOH treated fibers (HQ\_AK5/UP, KE\_AK5/UP) at  $30^\circ\text{C}$ . The storage moduli of the HQ/UP biocomposites were increased by treatment of the EB (HQ\_EB10/UP: 32%, HQ\_EB200/UP: 55%) and 5 wt% NaOH (HQ\_AK5/UP: 59%), compare to that of the UP matrix at  $30^\circ\text{C}$ . Also, the storage moduli of the KE/UP biocomposites (KE\_EB10/UP: 143%, KE\_EB200/UP: 143% and 5 wt% NaOH (KE\_AK5/UP): 135%) increased with these treatment, and the amount of modulus increase in the KE/UP biocomposites was larger than that of the HQ/UP biocomposites. The highest storage modulus of biocomposite was obtained with the combination of UP and 10 or 200 kGy EBI treated kenaf fibers.

**Loss Modulus:** The loss moduli of the UP and the biocomposites as a function of temperature are shown in Figure 7. UP resin showed the loss modulus peak at  $90^\circ\text{C}$ , which was attributed to the mobility of the resin molecules.<sup>29</sup> This peak has also been considered as the glass transition temperature ( $T_g$ ) of the resin.<sup>25,27</sup> In the composites made with the natural fibers, the transition peak ( $T_g$ ) shifted to the higher tempera-

**Table III. Comparison of Storage Modulus of Natural Fiber/UP Biocomposites**

	UP Matrix	HQ/UP Biocomposites			
		Raw (HQ /UP)	10 kGy (HQ_EB10/UP)	200 kGy (HQ_EB200/UP)	NaOH 5% (HQ_AK5/UP)
Storage modulus (GPa)	2.78	3.44	3.67	4.30	4.43
Increase (%)	0	23	32	55	59
	UP Matrix	KE/UP Biocomposites			
		Raw (KE /UP)	10 kGy (KE_EB10/UP)	200 kGy (KE_EB200/UP)	NaOH 5% (KE_AK5/UP)
Storage modulus (GPa)	2.78	5.90	6.76	6.76	6.54
Increase (%)	0	112	143	143	135



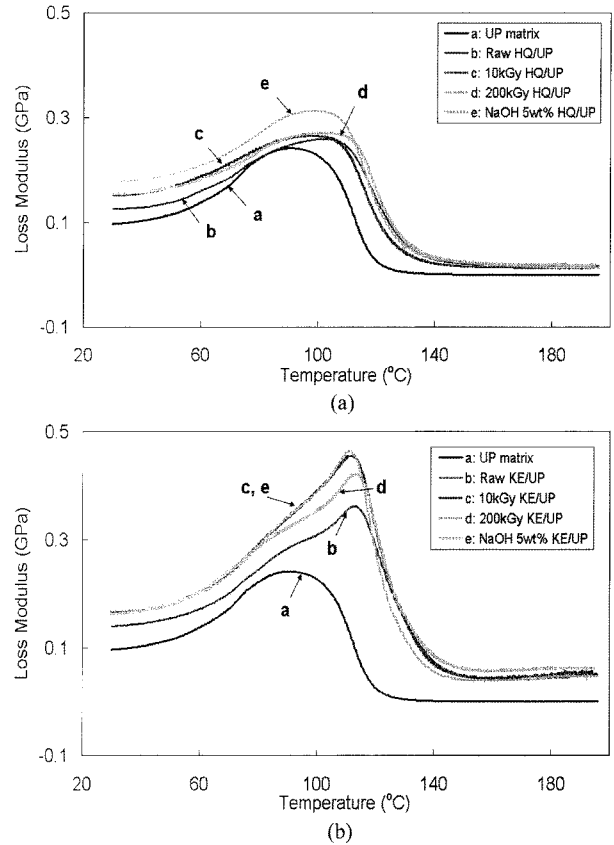
**Figure 6.** Storage modulus of natural fiber/UP biocomposites: (a) HQ/UP and (b) KE/UP.

ture. This may be due to the immobilization of the polymer molecules near the surface of the natural fibers due to various molecular interactions, which increased the  $T_g$  (HQ: 110 °C, KE: 115 °C) of the biocomposites. A similar increase in the  $T_g$  and an increase in the loss modulus due to the incorporation of the reinforcing fibers were also reported by some authors.<sup>25,27</sup> The broadening of the loss modulus peaks with the fibers in the biocomposites could be due to the increase in the energy absorption caused by the natural fibers.

**Tan  $\delta$ :** The variation of the tan  $\delta$  of the UP and the biocomposites as a function of temperature is shown in Figure 8. The tan  $\delta$  was very high for the UP matrix due to the high reduction of the storage modulus values on increasing the temperature. As similar to thermoplastic biocomposites the incorporation of the reinforcing natural fibers restricted the mobility of the UP molecules resulting in the raised storage modulus values and the reduced viscoelastic lag between the stress and the strain. This induced the decrease of the tan  $\delta$  values of the biocomposites.<sup>25-27</sup> The tan  $\delta$  values were lowered in the biocomposites compared to the UP matrix resulting from less matrix by volume to dissipate the vibrational energy.<sup>28</sup>

**Thermal Stability.**

**Natural Fiber/PP Biocomposites:** Table IV and Figure 9 show TGA curves of PP, HQ/PP and KE/PP biocomposites.

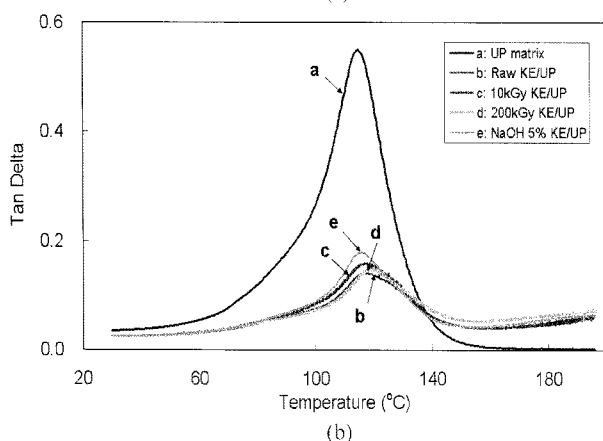
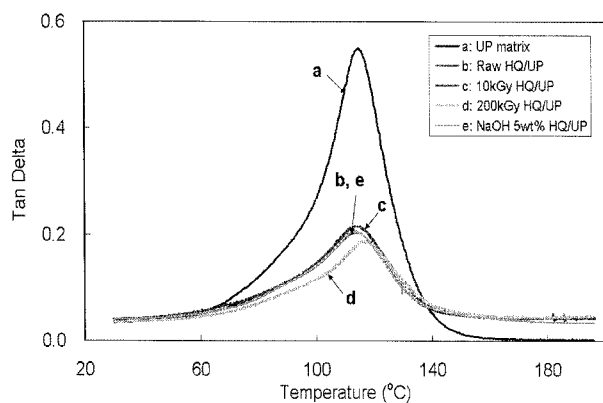


**Figure 7.** Loss modulus of natural fiber/UP biocomposites: (a) HQ/UP and (b) KE/UP.

**Table IV. Decomposition Temperature of Natural Fiber/PP Biocomposites**

	Hemicellulose	Cellulose	PP Matrix
Natural Fibers	280-320 °C	360 °C	-
PP Matrix	-	-	455-456 °C
<b>Biocomposites</b>			
EB Treated HQ	280-320 °C	346-359 °C	455-456 °C
NaOH Treated HQ	Removed	358-360 °C	455-456 °C
EB Treated KE	280-320 °C	344-360 °C	455-456 °C
NaOH Treated KE	Removed	360 °C	454-456 °C

The decomposition peaks were found in the range of 280-320, 340-365 and 430-470 °C, which was due to hemicellulose, cellulose, and PP, respectively. The natural fibers were incorporated into the PP matrix of the biocomposite. Thus, the maximum peak of PP shifted from 450 to 455 °C. The cellulose peaks in the raw natural fiber/PP biocomposite were gradually decreased from 360 to 344 °C with increasing the EBI dose. The peak for hemicellulose in NaOH 5 wt% treated natural fibers/PP disappeared in the range of 280-320 °C.<sup>30</sup>

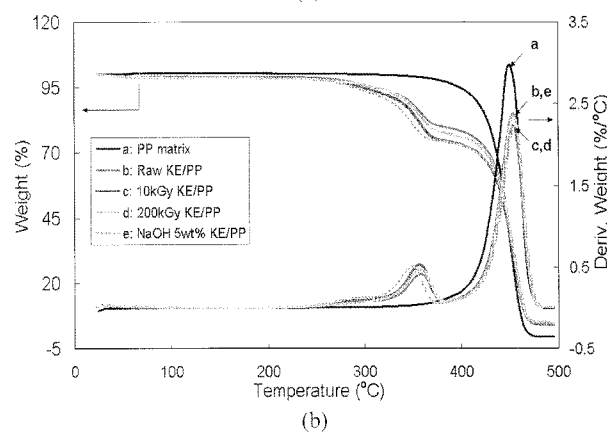
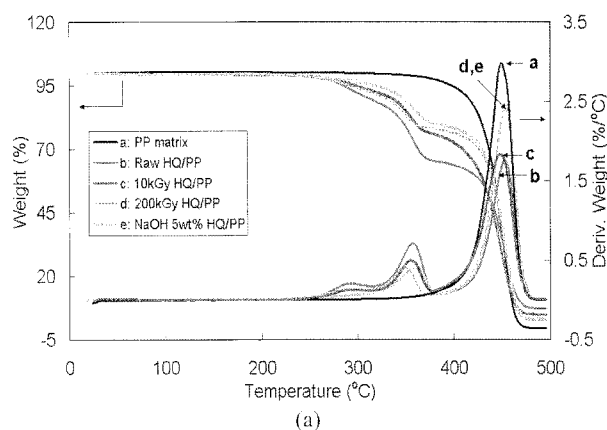


**Figure 8.** Tan  $\delta$  of natural fiber/UP biocomposites: (a) HQ/UP and (b) KE/UP.

**Table V. Decomposition Temperature of Natural Fiber/UP Biocomposites**

	Hemicellulose	Cellulose	UP Matrix
Natural Fibers (HQ, KE)	280-320 °C	360 °C	
UP Matrix			390 °C
<b>Biocomposites</b>			
EB Treated HQ	280-320 °C	356-364 °C	394-397 °C
NaOH Treated HQ	Removed	364-365 °C	390-398 °C
EB Treated KE	280-320 °C	355-367 °C	395-397 °C
NaOH Treated KE	Removed	362-363 °C	394-396 °C

**Natural Fiber/UP Biocomposite:** Table V and Figure 10 show TGA curves of UP, HQ/UP and KE/UP biocomposites. The decomposition peaks due to hemicellulose and cellulose components in the natural fiber were found in the range of 280-320 and 340-365 °C, respectively. The maximum peak of UP shifted from 390 to 395 °C in the HQ or KE/UP biocomposite. This can be explained by suggesting that the natural fibers were incorporated into the UP matrix of the biocomposite.

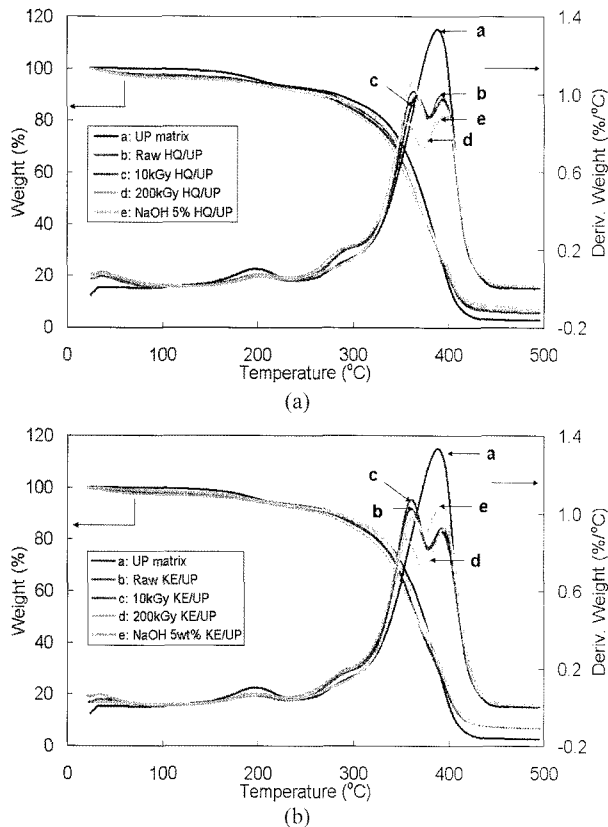


**Figure 9.** TGA and DTG thermograms of natural fiber/PP biocomposites: (a) HQ/PP and (b) KE/PP.

## Conclusions

The treatment of henequen and kenaf fibers by electron beam and alkali solution (NaOH) was conducted in this study. The EB treatment of natural fibers showed no change in functional groups and crystallinity of fibers. In contrast, the NaOH treatment of natural fibers formed glycoside bonding and removed hemicellulose. The DMA results showed that kenaf fiber had a greater reinforcing effect on the dynamic mechanical properties of both PP and UP biocomposites than henequen fiber. Also, the EB irradiation with 200 kGy and the 5 wt% alkali solution for natural fibers (HQ and KE) contributed to good interfacial adhesion between a polymer matrix (PP or UP) and natural fibers through surface modification of fibers.

Especially, the storage modulus of KE/PP biocomposite treated with NaOH 5% was the greatest in the natural fiber/PP biocomposites. Also, the storage modulus of KE/UP biocomposite treated at both 10 and 200 kGy doses was the greatest in the natural fiber/UP biocomposites. The EB treatment on the natural fiber contributed to improving the dynamic mechanical properties of natural fiber/polymer biocomposites due to the efficient surface treatment of natural fiber for biocomposites.



**Figure 10.** TGA and DTG thermograms of natural fiber/UP bio-composites: (a) HQ/UP and (b) KE/UP.

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