Influence of Bioadditives Made from Sugarcane Bagasse on Interpenetrating Polymer Networks

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Received 12 March 2020; Revised 20 May 2020; Accepted 23 June 2020; Published 25 July 2020

To achieve a sustainable bioeconomy, various bio-derived additives have been developed to produce biocomposites, but only a handful of research on biocomposites focuses on the effect of bio-derived additives on interpenetrating polymer networks (IPNs). This study is aimed at understanding the interaction between bioadditives and interpenetrating polymer networks and is the first study to build the relationship between bioadditive ratio and damping factor based on dynamic mechanical analysis. The IPNs were prepolymerized in bulk by isocyanate and poly(oxypropylene) polyol (PPG) with two different molecular weights (PPG 700 and PPG 1000), and then, they were grafted with bisphenol A diglycidyl ether epoxy. The bioadditives were prepared from agricultural waste, sugarcane bagasse, and the effect of the coupling agent 3-glycidoxypropyltrimethoxysilane on a bioadditive surface was also discussed in this study. The results show that modified bioadditives have significant enhancement on tensile strength and tensile modulus of polyurethane-grafted epoxy resin interpenetrating polymer networks (PU(PPG)-EP graft-IPNs). However, the enhancement is not from a strong covalent bond between matrix and additives, that is, due to the well-dispersed bioadditives which provide stiff segments. The static and dynamic mechanical performance, water absorption ratio, and morphology of the (PU(PPG)-EP graft-IPNs) elastomers were also thoroughly discussed in this study.

1. Introduction

Billions of tons of agricultural waste are produced annually worldwide, and these wastes cause severe environmental pollution and threaten the health of all livings [1]. Thus, there is a pressing need to convert these bio-wastes into value-added products under the concept of circular economy and sustainability. From these bio-residues, we can extract chemical compounds such as cellulose fibers and lignin powders that can replace petroleum-based products. Additionally, these additives can lead us to a bioeconomy because of their renewability, versatility of use, and applications. Biowaste can often reduce costs and reinforce the desired qualities for the matrix. In this case, sugarcane bagasse presents as a novel bioadditive that was only considered as voluminous waste in the past.

Historically, sugarcane was one of the most important crops in Taiwan. During the 1940s, 1.2 million tons of sugar were produced representing 7% of the world sugar production (ST and Wang 2015); today, a variety of sugarcane-based products can be found on the market, such as sucrose, sugarcane juice, and sugarcane liquors. The sugarcane
bagasse residue is remained after extracting their juice, and
the bagasse is inedible and loose in texture resulting in high
transportation fees which are unpreferable for bioenergy
[2]. Thus, many studies have added sugarcane bagasse to
other polymers as a reinforcing material, which includes
the following polymers: polypropylene (PP) [3, 4], polyvi-
nyl chloride (PVC) [5], high-density polyethylene (HDPE)
[6–8], and polylactic acid (PLA) [9–11]. However, very
few of them focus on the effect of sugarcane bagasse on
blended polymers or hybrid polymers, especially interpen-
etrating polymer networks.

Interpenetrating polymer networks (IPNs) are defined
as a blend of two or more polymers where each material
forms a continuous network, which interpenetrates the
other [12]. IPNs are mainly composed of thermosetting
polymers, where polyurethane and epoxy resins dominate
the thermosetting polymer market. Nowadays, both are
being widely used in composite materials and often can
compensate each other to improve mechanical perfor-
manace. For example, epoxy resin has excellent thermal
resistance, great electrical insulation, strong adhesion, supe-
rior chemical resistance, and easy processing [13], but it is
relatively brittle leading to poor impact resistance com-
pared to polyurethane [14]. The combination of epoxy
and polyurethane can efficiently gain both strengths with-
out costly treatments. The theory of IPNs has been com-
monly applied in these resins, but the effect of bioadditives
on interpenetrating polymer network is seldom mentioned
in the previous work.

In this study, epoxy resin was used as the matrix to
polymerize and interpenetrate simultaneously, which is
generally called simultaneous molecular network (SIN)
[15, 16], and then formed a polyurethane-grafted epoxy
resin interpenetrating polymer network (PU(PPG)-EP
graft-IPNs), which increases the mechanical and thermal
properties of epoxy resin [17, 18] and broadens the appli-
cations [19]. A siloxane coupling agent was applied to
chemically modify the bagasse and to improve the inter-
face between additives and matrix [11, 20, 21]. A better
interface can clearly indicate the relationship between
bioadditive ratio and damping factor of IPNs based on
thorough surveying of its mechanical properties and mor-
phology [22]. This study offers a solution to tackle the
waste issue and reveals the influence of bioadditives on
IPN structures.

2. Materials and Methods

2.1. Experimental Materials. Bisphenol A type epoxy (DER
331) with an epoxy equivalent weight of 186g/eq was

| Abbreviations                      | PU   | Additives                                      |
|------------------------------------|------|------------------------------------------------|
| PU(PPG 700)-EP                     | PPG 700 | No additives                                   |
| PU(PPG 1000)-EP                    | PPG 1000 | No additives                                   |
| PU700-EP-UnSB (30-50)              | PPG 700 | Untreated sugarcane bagasse from 30 wt% to 50 wt% |
| PU1000-EP-UnSB (30-50)             | PPG 1000 | Untreated sugarcane bagasse from 30 wt% to 50 wt% |
| PU700-EP-MoSB (30-70)              | PPG 700 | Treated sugarcane bagasse from 30 wt% to 70 wt%  |
| PU1000-EP-MoSB (30-70)             | PPG 1000 | Treated sugarcane bagasse from 30 wt% to 70 wt%  |

Figure 1: FTIR spectrum of PU-modified epoxy synthesis over reaction time.
procured from EPOCONE, Taiwan. Poly(oxypropylene)glycol (PPG) with different molecular weights was provided by Bayer. TDMP (2,4,6 tri(dimethylaminomethyl)phenol, reagent grade) from Ciba-Geigy Co., Switzerland, was used as the hardener for epoxy. The sugarcane waste was received from a local supplier in Taiwan. 3-Glycidoxypropyltrimethoxysilane (GPS, reagent grade) from Acros was applied to modify the sugarcane bagasse surface. Three synthesis reagents: 4,4′-diphenylmethane diisocyanate (MDI, reagent grade), trimethylolpropane (TMP, reagent grade), and 1,4-butanediol (1,4-BD, reagent grade), were purchased from TCI Chemicals, Alfa Aesar, and Riedel de Haën, respectively. All other reagents and solvents, including industrial-grade ethanol, were used as received.

2.2. Experimental Methods

2.2.1. Preparation of Sugarcane Bagasse Bioadditives. The sugarcane bagasse was cleaned with running water, and then, it was dried in the oven at 70°C for 24 hours. The dried biomass was crushed into powders, and the powder size was selected through 60 mesh screens.

2.2.2. Surface Modification on Bioadditives [23]. The surface of the bioadditives was cleaned using 3% ethanol to remove wax or extractives, and then, 3-glycidoxypropyltrimethoxysilane was added as a coupling agent to reach a 2% concentration. The solid-liquid ratio was set at 1:10. The entire solution was treated by ultrasonic oscillations for 20 minutes and then mechanically stirred for one hour to complete the grafting reaction. After removing the solution, surface-grafted sugarcane bagasse was washed with deionized water to ensure no
2.2.3. Preparation of PU-Modified Epoxy Resins (Adopting the Method Introduced by [24, 25]). Two reagents were mixed at four neck reaction round bottles, including 1 equivalent weight of PPG diol (PPG 700 or PPG 1000) and 2 equivalent weights of MDI. The bottle was slowly heated up to 70°C in an oil bath and mechanically stirred until the intensity of the NCO functional group reaches a constant level detecting by FTIR. Excess amounts of PU prepolymer then reacted with epoxy resins in the same bottle to form PU-modified epoxy at 70°C. Its NCO functional group was monitored at hourly intervals using FTIR until the intensity of NCO remained unchanged as shown in Figure 1.

2.2.4. Addition of Sugarcane Bagasse Powders in PU-Modified Epoxy Resins. An adequate amount of acetone was added into the PU-modified epoxy resins to decrease the viscosity. TMP and 3phr TDMP were added into the resin with a ratio of 1:4 between 1,4-BD/TMP. Unmodified and modified sugarcane bagasse was added into the resins with a range of ratios (30 wt%-70 wt% increasing by 10 wt%) and then mechanically stirred until achieving a homogeneous texture. The mixed resin was poured into a Teflon mold for

coupling agents remain. After that, it was left in the oven at 90°C for 24 hours.

Figure 4: Tan δ of PU-EP grafted-IPNs added unmodified sugarcane bagasse. (a) PPG 700 PU-EP; (b) PPG 1000 PU-EP.
compression, was cured under 15 MPa of pressure at 80°C for one hour, and was finally postcured at 110°C for another one hour. There were 18 sample compositions prepared as shown in Table 1.

2.3. Characterizations

2.3.1. Tensile Mechanical Test. According to ASTM D638, the tensile test was conducted using Instron 4469 with a span speed of 10 mm/min to measure the tensile strength and tensile modulus. The sample size is $60 \text{ mm} \times 10 \text{ mm} \times 2 \text{ mm}$ with 5 replicates.

2.3.2. Izod Impact Test. The sample size was the same as that of the tensile test, and the edge was cut in a V-shape according to ASTM D256. The specimens were fixed on the instrument base (GOTECH, GT-7045-I), and the V-cut faced the pendulum. The pendulum was released at room temperature and hits the specimens to calculate the impact energy.

2.3.3. Dynamic Mechanical Test. The dynamic mechanical analysis was conducted using a dynamic mechanical analyzer (PerkinElmer, Pyris) to measure storage modulus ($E'$), loss modulus($E''$), and tan δ. The experimental mode is a 3-
point bending test, and the frequency is fixed at 1 Hz with 5 μm amplitude. The heating rate used is 10°C/min from -10 to 250°C.

2.3.4. Morphology. The morphology of specimens was observed through a scanning electron microscopy (TESCAN 5136MM) with 20 kV voltage to analyze its compatibility and fracture cross-sections. Samples were prepared by two steps: first by quenching with liquid nitrogen and then by sputtering a layer of gold through vacuum metalizing.

2.3.5. Water Absorption Test. For the water absorption analysis, specimens were cut into 10 mm × 10 mm × 2 mm pieces. They were dried in the oven at 70°C for three hours and then measured for its dry weight (Wd) using a digital balance (d = ±0.1 mg). Then, the specimens were immersed into 25°C distilled water for a time frame ranging from a few hours up to 20 days. These specimens were taken to measure their wet weight (Wa). The formula for the water absorption ratio is shown as follows: $W_t(\%) = \frac{(W_a - W_d)}{W_d} * 100$.

3. Results and Discussion

Interpenetrating polymer networks (IPNs) refer to a structure with two distinct polymers inserted into each other in the segmental level. Since the level is down to a molecular size, the glass transition temperature ($T_g$) of IPNs is expected to be uniform, which means only one $T_g$ should be found on the DMA results. As shown in Figure 2, the neat PU-EP elastomer has a sharp tan δ peak around 110-120°C with a tan δ peak value of 0.729-0.732. The peak temperature of tan δ is used to describe its $T_g$, and the magnitude of the tan δ peak is associated with the damping property of a material. Compared to the $T_g$ of PPG 700 PU-EP, the high molecular weight PPG 1000 PU-EP is around 8°C higher due to the larger space between polymer chains of PPG 1000, which allows more EP to penetrate and then increase crosslinking density [24, 25].

Apart from the single tan δ peak indicating the IPN structure, FTIR analysis was also applied to prove the grafting reaction between isocyanate and hydroxyl groups. The results show that the epoxide did not involve in the grafting reaction since the IR absorption intensity of epoxide remains consistent before and after the grafting reaction (Figure 3). The reaction occurs between the isocyanate groups in the PU prepolymer and the hydroxyl groups in the epoxy, and thus, the IR absorption peaks decrease significantly at 3450 cm⁻¹ (OH⁻) and 2270 cm⁻¹ (NCO⁻).

Since the network was confirmed to be a uniform interpenetrating polymer network, bioadditives were then combined to understand their influence. Sugarcane bagasse powders were mixed with PU-EP elastomers, and its tan δ is shown in Figure 4.

The unmodified bioadditives significantly decreased the peak value of tan δ from 0.7 to 0.3-0.4, but its $T_g$ also decreased from above 110°C down to 70°C. The storage modulus increased due to the biomass stiffness; these unmodified bioadditives have high stiffness segments which can decrease the damping of elastomer leading to a lower tan δ peak value. In previous studies, as more biomass was added into the polymers, it generally resulted in lower tan δ peak values and higher $T_g$ [13]; however, it is not consistent for our unmodified bioadditives. For example, the trend of PPG 700 tan δ peak values is SB 30 wt% < SB 50 wt% < SB 40 wt% instead of SB 30 wt% < SB 40 wt% < SB 50 wt%. Thus, we modified the biomass surface to generate a better interface between additives and the interpenetrating networks as shown in Figure 4.

![Figure 6: Linear relationship between sugarcane ratio and tan δ peak value.](image-url)
After modifying the interface by 3-glycidoxypropyltrimethoxysilane (GPS), the tan δ peak value of modified bagasse added elastomer become lower (tan δ peak value < 0.2) than that of unmodified ones (tan δ peak value = 0.41-0.34) and neat polymers (tan δ peak value = 0.7). The significantly decreased tan δ peak value indicates that GPS can be an ideal coupling agent between bioadditives and the interpenetrating polymer network because it improves the interface with less damping. The influence of GPS can be found not only on the peak value of tan δ but also on the high-temperature region (>150°C). Above 150°C, the curve of unmodified elastomer is quite fluctuant compared to that of GPS-treated elastomer due to a less friction interface.

On top of tan δ peak value analysis, tan δ area analysis is another method to characterize damping ability. As shown in

**Figure 7**: (a) Linear relationship between sugarcane ratio and tan δ peak value. (b) Logarithm relationship between the sugarcane ratio and tan δ peak value.
Figures 2, 4, and 5, the loss area of neat PU-EP graft-IPNs has the largest area which indicates higher mechanical energy absorption in a multicomponent polymer system. The tan $\delta$ peak value of neat PU-EP graft-IPNs is around 0.7. The loss area gets decreased after adding unmodified SB (tan $\delta$ peak value = 0.34) and then decreased more with modified SB (tan $\delta$ peak value < 0.2). The decrease of loss area was caused by the molecular rigidity of SB, and the modified SB has the lowest loss area because of the potential linkage between polymers and additives.

Many researchers show the loss area decreased after adding additives as we mention above. However, to the best of our knowledge, no research focuses on the effect of fiber loadings on the tan $\delta$ peak value. Surprisingly, the decrease tan $\delta$ peak value of PPG 700 PU-EP series in our study shows perfect linear relationship up to 60 wt%, while that of PPG 1000 PU-EP series also shows perfect linear relationship from 40 wt% to 70 wt% (Figure 6).

As one of our major findings, we observed that the maximum tan $\delta$ decreased linearly ($R^2 > 0.99$) within a certain range (Figure 6) when more bioadditives were added into this interpenetrating polymer network. As many of us know, rigid additives can significantly reduce the tan $\delta$ peak value of polymers [11], but this is the first work that shows the relationship between tan $\delta$ peak value and fiber amount. A previous study [26] mentioned the decrease of tan $\delta$ peak value, but to the best of our knowledge, the relationship between the two of them is still unclear.

Out of this range, the relationship between sugarcane bagasse ratio and tan $\delta$ peak is not linear (Figure 7). For PPG 700 PU-EP, tan $\delta$ peak value above 60 wt% bagasse decreased much less (0.037%) than the linear decrement (23.2%), while for PPG 1000 PU-EP, tan $\delta$ peak value below 40 wt% bagasse is extremely high. The slope of the PPG 700 PU-EP fitting curve is steeper than that of the PPG 1000 PU-EP curve. Additionally, the intercept which represents the neat polymer is only 0.28, much less than the 0.7 that was measured. That implies the linear model might not be the right model to describe our data. Also, we found many articles showing a significant tan $\delta$ peak value decrease after adding bioadditives in a single polymer matrix although the relationship is not discussed further [27]. This indicates that the logarithm model provides a better fit (Figure 7). Previous research has indicated bioadditives usually enhance the elastic part of the polymer leading to lower tan $\delta$ [11]. Our results also show more bioadditives can increase the modulus of the stiff region and decrease the tan $\delta$ peak value. Many reports have shown the positive effect of additives on the polymer tan $\delta$ peak value, but this is the first work that shows a logarithmic effect ($R^2 > 0.95$) of bioadditives on the interpenetrating polymer network.

Similar to tan $\delta$, the glass transition temperature ($T_g$) decreases in a linear relationship which indicates every addition of 10 wt% of sugarcane bagasse can decrease the $T_g$ about 5°C as shown in Figure 8. The decrease of $T_g$ implies that the interface between bioadditives and matrix lacks covalent linkage. The coupling agent improved the dispersion of bioadditives and reduced the aggregations. Since the dispersion of bioadditives improved, the surface area between additives and matrix also increased resulting in more friction generated. More friction between polymer and bioadditives led to lower temperatures of the
tan δ peak in terms of glass transition temperature. Thus, the increase in the amount of bioadditives increases the rigidity of IPNs and also increases the friction at the same time.

As with many other bioadditives, water absorption property is an important criterion to evaluate its overall quality [28]. These bioadditives were made from sugarcane bagasse, which contains plenty of hydroxyl groups on the surface leading to a higher water absorption ratio [29]. As shown in Figures 9 and 10, the untreated elastomer has a poor interface between bioadditives and the interpenetrating polymer network. The interface is very weak, and it can be destroyed within one week’s soaking process, so some unmodified elastomer curves ended in a short period of time. The water absorption ratio of the treated elastomers is only one-third of the untreated elastomers (Figures 9(a) and 10(a)), which indicates that the GPS-treated elastomer has better water absorption properties (Figures 9(b) and 10(b)). Also, PPG 1000 PU-EP shows a lower water absorption ratio compared to PPG 700 PU-EP and it is mainly due to its molecular weight.

Similar to the dynamic mechanical properties, the unmodified bioadditives has a deleterious effect on static mechanical performance compared to modified bioadditives.
The tensile strength of IPNs with unmodified additives has extremely low mechanical performance which is generally lower than 10 MPa. As showed in Figures 11(a) and 12(a), compared to IPNs with unmodified bioadditives, the tensile strength of IPNs with modified bioadditives improves by about 3 to 8 times. Similar to tensile strength, tensile modulus is also significantly improved by about 1.5 to 2 times (Figures 11(b) and 12(b)).

The effect of PPG molecular weights on IPNs has been published in our previous study [24, 25]. The low molecular weight PPG (PU(PPG 700)-EP graft-IPNs) has an average tensile strength of 57 MPa, while the high molecular weight PPG (PU(PPG 1000)-EP graft-IPNs) has an average tensile strength of 45 MPa due to the difference between crosslinking densities. The low PPG has higher crosslinking density, and the schematic diagram is shown in Scheme 1.

As the PPG 1000 has larger gaps between chains compared to PPG 700, it allows more epoxy resins to penetrate into the structure of PU and then increases the interpenetrating degree which limits the mobility of polymer chains. Thus, the $T_g$ value of PU(PPG 1000)-EP graft-IPN measured by DMA is 120°C, which is higher than that of PU(PPG 700)-EP graft-IPNs ($T_g$ is 112°C) and it is consistent to our previous study [24, 25].

Figure 10: Water absorption property of PPG 1000 PU-EP elastomers: (a) untreated bioadditive IPN and (b) GPS-treated bioadditive IPN.
Previous studies have shown that unmodified sugarcane bagasse has large amounts of impurities on the surface such as lignin, which can cause a poor interface between matrix and the bagasse. To address this issue, many researchers used ethanol to remove hydrophobic impurities and generated many surface grooves to improve the wetting properties of the matrix on additives. The overall mechanical performance can become significantly improved after adding a silane coupling agent on additives by the surface hydroxyls. The modified bioadditives can also react with the epoxy-functional groups in the matrix leading to strong and ductile IPNs.

In this study, the tensile strength reached a maximum value with 50 wt% (PPG 700) or 40 wt% (PPG 1000) of sugarcane bagasse; however, the tensile modulus shows a minimum value with 50 wt% of sugarcane bagasse. The possible reason is still uncertain which requires more future work.

The impact properties of IPNs were also investigated, as presented in Figure 13. Compared to the impact strength of unmodified sugarcane bagasse, modified sugarcane bagasse elastomer has a higher value due to its good interface with the matrix which can transfer the stress from a matrix to bioadditives. The improved interface can be observed by SEM images as shown in Figures 14 and 15.

In Figures 14 and 15, the voids and gaps of unmodified SB/PU-EP graft-IPNs can be easily observed, while those of modified SB/PU-EP graft-IPNs are much less obvious which shows much better morphology.
4. Conclusion

This study investigated the effect of bioadditives on dynamic mechanical properties, static mechanical properties, water absorption ratio, and morphology of IPNs. Our results show that the neat PU-EP, without bioadditives, has a stable interpenetrating polymer network represented by its singular sharp tan δ peak. After adding
unmodified bioadditives, there is an irregular trend between bioadditive amounts and tan δ peak value. It is difficult to derive any solid conclusions on account of the inconsistent results. Thus, the bioadditives, sugarcane bagasse, were modified using a GPS coupling agent to enhance its compatibility. The results show the tan δ peak value decreased linearly at a certain range when increasing bioadditive amounts ($R^2 > 0.99$). However, due to the intercepted value of a linear model and other concerns, we suggested that the logarithmical model is a better representation ($R^2 > 0.95$). This is also the first work to propose a logarithmical model to describe the effect of bioadditive amounts on the tan δ peak value of IPNs.

Different to tan δ values, the glass transition temperature of IPNs was linearly decreasing by around 5°C for each 10 wt% of sugarcane bagasse. Based on these findings, we believe that the coupling agent (GPS) does not create any covalent linkage between polymers and bioadditives. On the contrary, the coupling agent improves the interface by dispersing the bioadditive uniformly and reducing the aggregation of these bioadditives. We propose that the decrease of the glass temperature of the composite could also be related to the decrease of the crosslinking density; further research is suggested. Regarding the water absorption ratio, static mechanical performance, and morphology, modified bioadditives exhibit positive effects on all of them. Beyond our comprehension, the tensile

![Figure 13: Izod impact strength of PPG 700 elastomer and PPG 1000 elastomer.](image-url)
strength and tensile modulus slightly show an opposite trend; thus, more work should be done in the future to elucidate this phenomenon.

**Data Availability**

The authors confirm that the data supporting the findings of this study are available within the article.

**Conflicts of Interest**

The authors declare that they have no conflicts of interest.

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