



## Compatibilizing and toughening bamboo flour-filled HDPE composites: Mechanical properties and morphologies

H. Liu<sup>a</sup>, Q. Wu<sup>a,\*</sup>, G. Han<sup>b</sup>, F. Yao<sup>a</sup>, Y. Kojima<sup>c</sup>, S. Suzuki<sup>c</sup>

<sup>a</sup>School of Renewable Natural Resources, Louisiana State University Agricultural Center, Room 107 RNR Building, Baton Rouge, LA 70803, USA

<sup>b</sup>College of Material Science and Engineering, Northeast Forestry University, Harbin, China

<sup>c</sup>Faculty of Agriculture, Shizuoka University, Shizuoka 422-8529, Japan

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### ABSTRACT

Mechanical properties and morphologies of HDPE/bamboo flour (BF) composites modified with two maleated ethylene/propylene elastomers (EPR-g-MA) and one maleated polyethylene (PE-g-MA) were investigated. The concentration of each modifier varied between 0 and 8.3 wt% based on the total composite weight. The highest values in strengths and moduli were achieved for the composites containing PE-g-MA, whereas the use of semi-crystalline EPR-g-MA (sEPR-g-MA) led to the maximum impact strength. However, the presence of amorphous EPR-g-MA (aEPR-g-MA) reduced strengths of the resultant composites. The impact toughness of the aEPR-g-MA modified composites initially increased with aEPR-g-MA loading up to 2.9 wt%, and then gradually decreased beyond this level. By simultaneously incorporating both PE-g-MA and EPR-g-MA, the strength–toughness balance of the final composites was optimized. The optimal balance of composite properties was achieved at lower percentage of sEPR-g-MA and medium BF loading level. Dynamic mechanical analysis (DMA) and morphology observations well substantiated the above-mentioned mechanical results. It was found that suitable interfacial adhesion and easiness of interlayer cavitations between HDPE and BF is beneficial to initiate massive matrix yielding in the composites, thereby achieving satisfactory impact strength.

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### 1. Introduction

Over the past two decades, extensive effort has been devoted to wood and polymer composites (WPCs) due to many emerging applications, such as decking, building materials, automobile components, and infrastructure. Compared with traditional glass fiber and mineral fillers, wood fillers are less-expensive, low density, sustainable, and less abrasive to processing machinery. Because wood undergoes degradation above 200 °C, polyethylene (PE) and polypropylene (PP) are among the commonly used thermoplastic matrix due to their relatively low price and processing temperature [1]. However, large difference in surface polarity between hydrophilic wood and hydrophobic polyolefin matrix usually leads to poor interfacial adhesion and thus poor mechanical properties of final materials. To promote the interaction between them, various coupling agents or compatibilizers (mainly functionalized polymers) have been used in the literature. Among them, maleic anhydride-grafted PE or PP is the most commonly available because of their ability to effectively enhance mechanical properties of the composites, especially tensile and flexural strengths. Such improvement has been attributed to the polar interaction and

covalently link between anhydride carbonyl and hydroxyl groups of wood surfaces [2–6], as well as their good compatibility with polyolefin matrix [7–10].

Unfortunately, low impact strength is still the major drawback of WPCs in many end-use applications, as compared to glass fiber reinforced composites. Therefore, much attention has been focused on the toughening of PE- or PP-wood composites with various polyolefinic elastomers or their maleated counterparts, such as SBR [11], styrene/ethylene–butylene/styrene triblock copolymer (SEBS) [7,8,12–16], ethylene/propylene/diene terpolymer (EPDM) [15,17], ethylene/octene copolymer (EOR) [18], ethylene/methyl acrylate copolymer (EMA) [18], ethylene/butyl acrylate/glycidyl methacrylate copolymer (EBAGMA) [19], and Surlyn ionomers [14,20]. The strength and modulus of final composites were less improved or even decreased by the addition of elastomers alone, although the impact toughness was more or less improved, depending on the nature and content of elastomers used. Ethylene/propylene copolymer (EPR) is one of polyolefin-based thermoplastic elastomers. Its fully saturated backbone results in more excellent thermal, oxidative stability, and weatherability than conventional EPDM. Therefore, it has been widely used to enhance impact strength of virgin HDPE [21], PP [22–25], or PP-based composites filled with mineral fillers [26,27]. Apart from the improved dispersion level of the fillers [28,29], functionalized EPR

\* Corresponding author. Tel.: +1 225 578 8369.

E-mail address: [wuqing@lsu.edu](mailto:wuqing@lsu.edu) (Q. Wu).

(e.g., MA group) could further promote the favorable core–shell encapsulation around them, which thus results in an increase in both filler–matrix adhesion and impact strength [30–33].

Depending on stereoregularity and composition of monomer sequence, EPR copolymers are generally classified into amorphous and semi-crystalline ones. It was demonstrated that such difference in crystalline nature had a pronounced effect on mechanical properties and phase morphology of their resulting blends [25,34,35]. Okada et al. systematically investigated the influences of crystallinity of added EPR-g-MA on morphological, thermal and mechanical properties of nylon 6 blends over the whole composition range [34]. It was found that maleated EPR with some ethylene crystallinity resulted in blends having better mechanical properties and higher crystallinity than those based on amorphous EPR-g-MA. Compared to the latter blends, the strain-hardening and cold-drawing were enhanced by ethylene crystallinity in the former ones. And some morphological difference between both EPR-g-MA in their blends with nylon 6 was observed. Such crystalline effect of EPR copolymers was also studied in PP/EPR blends [25,35].

To optimize the trade-off between stiffness/strength and toughness of final WPCs, the combination of elastomers with maleated polyolefins as combined modifiers has been reported [15,18,36]. Sumbatsompop et al. investigated the effects of incorporating both PP-g-MA and impact modifiers into PP/wood sawdust composites [18]. An addition of 2.0 wt% PP-g-MA concentration into the composites containing 11.1 wt% selected impact modifier, was suggested for optimization of the overall mechanical properties. Oksman et al. studied mechanical properties and morphology of impact modified PP/wood flour (WF) composites [15]. The composite containing both 10phr SEBS-g-MA and 2phr PP-g-MA exhibited superior impact strength to the one modified via either 10phr SEBS-g-MA or 2phr PE-g-MA, while its tensile strength was less decreased even by the incorporation of 10phr SEBS-g-MA. The same behavior was not observed in the EPDM/PP-g-MA or EPDM-g-MA/PP-g-MA modified PP/WF composites. In these literatures, however, there is lack of further understanding on how the ratio between combined modifiers affects mechanical properties and fracture behaviors of final composites.

As an important renewable resource, bamboo accounts for approximately 25% and 20% of the total biomass, respectively, in the tropics and the sub-tropical area [37]. The overall mechanical properties of bamboo are comparable to or even better than those of wood. Furthermore, bamboo can be renewed much more rapidly compared with wood. Thus, the advantages of bamboo make it highly competitive nature reinforcement in polymeric composites. In several preliminary reports, bamboo fiber or flour have been utilized as reinforcements of thermosetting resins [38–44], biodegradable polyesters [45–47], poly(vinyl chloride) (PVC) [48], and PP [49–55]. Ge et al. used bamboo flours as the reinforcements for unplasticized PVC and investigated its effect on the morphology, static mechanical properties, and thermal properties of the composites [48]. Compared with neat PVC resin, the introduction

of bamboo flour significantly improved the stiffness of the composites, while decreasing the tensile strength to some extent. Moreover, no obviously adverse effect on the thermal stabilities of these composites was observed. A series of maleated polypropylene (PP-g-MA) compatibilized PP/bamboo fiber composites were prepared by Mi and coworkers [49,50]. The results showed that the tensile strength and stiffness of bamboo fiber-reinforced PP composites were higher than those of the commercial wood pulp composite that was widely used in making office furniture. Thwe and Liao [52–55] prepared and characterized bamboo–glass fiber-reinforced PP hybrid composites (BGRP). It was shown that apart from strength and modulus, the durability and fatigue resistance of bamboo fiber-reinforced PP were enhanced by hybridization with small amount of glass fiber. Compared to the above-mentioned bamboo fiber, bamboo flour is cheaper because no complex fiber separation process is required. To our knowledge, few attempts have been made on HDPE/bamboo flour composites, although polyethylene accounts for the major share in the present WPCs' market [1]. Furthermore, most of earlier studies on bamboo-filled composites focused on reinforcement effect and other physical properties of the resultant composites. However, their compatibilizing and toughening issues are less addressed.

The objectives of this study were to investigate morphologies and mechanical properties of HDPE-based composites filled with bamboo flour as influenced (a) by the crystalline nature of maleated elastomer modifier in comparison with maleated PE; (b) by the combined EPR-g-MA and PE-g-MA modifier systems, and (c) by the loading rate of bamboo flour in the presence of combined modifiers.

## 2. Experimental

### 2.1. Materials and blend design

Table 1 lists related information for the polymer and various compatibilizers used in the study [56,57]. The two EPR-g-MAs (i.e., Exxelor VA1801 and VA1803) had almost equal MA grafting level. Exxelor VA1801 is semi-crystalline and is designated as sEPR-g-MA. Exxelor VA1803 is amorphous and is designated as aEPR-g-MA. Bamboo flour (BF) was directly obtained by grinding bamboo flakes using a hammer mill with a 28-mesh screen.

Blend design included three factorial experiments. The first experiment consisted of ten blends covering three compatibilizers (PE-g-MA, aEPR-g-MA, and sEPR-g-MA), four loading rates (0, 2.9, 5.7 and 8.3 wt% based on the total composite weight); and one HDPE/BF ratio (60/40 wt/wt). The second experiment consisted of nine blends covering two EPR-g-MAs (aEPR-g-MA and sEPR-g-MA), five PE-g-MA to EPR ratios (0/100, 33.3/66.7, 50.0/50.0, 66.7/33.3, and 100/0 wt/wt), and one HDPE/BF ratio (60/40 wt/wt). The third experiment consisted of three compatibilized blends covering three HDPE/BF ratios (70/30, 60/40, and 50/50 wt/wt) and one combined modifier system (PE-g-MA/sEPR-g-MA = 2:1 by

**Table 1**  
Characteristics of polymeric resins used in this work

| Polymer   | Commercial designation | Property   | Manufacturer                          |
|-----------|------------------------|--|---------------------------------------|
| HDPE      | HD6761.17              | MFR (190 °C/2.16 kg) = 6.1 g/10 min, density = 0.952 g/cm <sup>3</sup> [56]  | ExxonMobil Chemical Co. (Houston, TX) |
| PE-g-MA   | Epolene G2608          | MFR (190 °C/2.16 kg) = 6–10 g/10 min, M <sub>w</sub> = 65,000 g/mol, acid number = 8 mg KOH/g [57]                     | Eastman Chemical Co. (Kingsport, TN)  |
| sEPR-g-MA | Exxelor VA1801         | MFR(230 °C/10 kg) = 9 g/10 min, density = 0.87 g/cm <sup>3</sup> , 43 wt% ethylene, 53 wt% propylene, 1.21 wt% MA [34] | ExxonMobil Chemical Co. (Houston, TX) |
| aEPR-g-MA | Exxelor VA1803         | MFR (230 °C/10 kg) = 22 g/10 min, density = 0.86 g/cm <sup>3</sup> , 43% ethylene, 53 wt% propylene, 1.14 wt% MA [34]  | ExxonMobil Chemical Co. (Houston, TX) |

weight added at 15% of the BF weight); and one uncompatibilized blend (HDPE/BF = 60:40 wt/wt) and virgin HDPE as a control.

## 2.2. Sample preparation

Prior to compounding, bamboo flour was thoroughly dried at 80 °C for at least 2 days in a convection oven. Melt compounding was performed using an intermesh, counter-rotating Brabender twin-screw extruder (Brabender Instruments Inc., Hackensack, NJ) with a screw speed of 30 rpm. The temperature profile of barrels ranges from 150 to 175 °C. The extrudates were quenched in a cold-water bath and then pelletized into granules. After being oven-dried at 100 °C for overnight, the granules were injection molded into standard mechanical test specimens using an Injection Molding Machine (Batenfeld Plus 35, Batenfeld Inc., NJ). Injection and mold temperatures were 190 and 68 °C for HDPE/BF composites, respectively. Virgin HDPE control samples were directly molded at 180 °C injection temperature and 54 °C mold temperature.

## 2.3. Characterization

### 2.3.1. Static mechanical tests

**Tensile testing.** Type-I dumbbell-shape tensile specimens having a typical dimension of 165 × 13 × 3 mm, were measured using an Instron 5582 testing machine (Instron Co., Norwood, MA) according to ASTM D638. A crosshead speed of 5 mm/min and a gage length of 50 mm were used for the test.

**Flexural testing.** The specimens having a typical dimension of 80 × 13 × 3 mm were measured under three-point bending using the same Universal Testing Machine in accordance with ASTM D790. A crosshead speed of 1.3 mm/min and a span length of 50 mm were used for the test.

**Impact testing.** Izod impact strength was determined from specimens having a dimension of 63.5 × 12.7 × 3 mm using a Tinius Olsen Mode 1892 impact tester (Tinius Olsen Inc., Horsham, PA) according to ASTM D256. The notch angle of 45° and “V”-type notch depth of about 2.5 mm were used for the test.

Five replicate specimens were taken for each test and average data along with corresponding standard deviation were reported.

### 2.3.2. Dynamic mechanical analysis (DMA)

Selected samples having a dimension of 27.4 × 3.1 × 3 mm were subjected to DMA test using a Q800 Dynamic Mechanical Analyzer (TA Instrumens, New Castle, DE, USA). The measurements

were carried out with a three-point bending mode at a frequency of 1 Hz and the corresponding relaxation behaviors were recorded as a function of temperature. The temperature range used in the present study varied from –40 to 120 °C under nitrogen flow.

### 2.3.3. Scanning electron microscopy (SEM)

The scanning electron micrographs of the selected composite samples were recorded with a Hitachi S-3600N VP-SEM (Hitachi, Japan) operated at an acceleration voltage of 15 kV. Prior to observation, the fractured surfaces of the impact test specimens were sputtered with gold to avoid electrical charging during examination.

### 2.3.4. Data statistical analysis

Duncan's multiple range tests for pairwise comparison were used to test the effect of various treatments using *Statistical Analysis Software SAS/STAT*. Statistical ranking at the 5% significance level was provided among the treatments for each property.

## 3. Results and discussion

### 3.1. Mechanical properties

#### 3.1.1. Effect of individual modifier type and content

Table 2 summarizes measured strength and modulus properties for the HDPE/BF composite systems, together with the statistical ranking information for each property. As a control, mechanical properties of virgin HDPE were also given. Without addition of modifiers, tensile strength of BF-filled composite was 17.5 MPa, which is lower than virgin HDPE (18.9 MPa). This clearly indicated transfer failure of applied force from the matrix to rigid BF due to poor interfacial adhesion. The distinct situation was observed when three modifiers were individually added. Among them, PE-g-MA contributed to the maximum enhancement in tensile strength. When 2.9 wt% PE-g-MA was added, the tensile strength was significantly increased by 64% compared to the uncompatibilized binary composites. A much less improvement (about 11%) is also seen for the composites containing the semi-crystalline ethylene/propylene elastomer functionalized with MA (i.e., sEPR-g-MA). For both systems, tensile strength was always higher than that of virgin HDPE. An increase in tensile strength with respect to matrix implies that the applied load was transferred through the interface between HDPE matrix and rigid BF. In many polymer-filled systems, the tensile strength can be usually improved using compatibilizers or coupling agents via the enhanced

**Table 2**  
Summary of mechanical properties of virgin HDPE and HDPE/BF composites<sup>a</sup>

| Systems           | Modifier content (wt%) <sup>b</sup> | Strength property <sup>c,d</sup> |                         |                                      | Modulus property <sup>c,d</sup> |                        |
|-------------------|-------------------------------------|----------------------------------|-------------------------|--------------------------------------|---------------------------------|------------------------|
|                   |                                     | Tensile strength (MPa)           | Flexural strength (MPa) | Impact strength (kJ/m <sup>2</sup> ) | Tensile modulus (GPa)           | Flexural modulus (GPa) |
| HDPE              | —                                   | 18.9(0.2)                        | 21.3(0.1)               | 8.12(0.21)                           | 0.30(0.10)                      | 0.80(0.01)             |
| HDPE/BF           | 0                                   | 17.5(0.3)F                       | 32.9 (1.6)D             | 2.88(0.10)E                          | 3.64(0.26)A                     | 3.12(0.14)A            |
| HDPE/BF/PE-g-MA   | 2.9                                 | 28.7(0.4)A                       | 49.5(0.4)A              | 4.58(0.45)D                          | 3.38(0.38)BC                    | 2.98(0.11)AB           |
|                   | 5.7                                 | 28.1(0.2)B                       | 48.1(0.5)B              | 4.90(0.24)D                          | 3.46(0.08)AB                    | 2.95(0.12)AB           |
|                   | 8.3                                 | 27.5(0.5)C                       | 47.4(0.7)B              | 5.62(0.39)BC                         | 3.15(0.11)CD                    | 2.78(0.19)BC           |
| HDPE/BF/sEPR-g-MA | 2.9                                 | 19.5(0.3)D                       | 35.6(0.6)C              | 5.39(0.32)C                          | 3.00(0.20)DE                    | 2.60(0.23)CD           |
|                   | 5.7                                 | 19.4(0.3)DE                      | 35.4(0.8)C              | 5.97(0.21)B                          | 2.76(0.11)EF                    | 2.56(0.22)CD           |
|                   | 8.3                                 | 19.1(0.3)E                       | 34.9(0.7)C              | 7.19(0.41)A                          | 2.78(0.12)EF                    | 2.32(0.13)E            |
| HDPE/BF/aEPR-g-MA | 2.9                                 | 16.1(0.1)G                       | 29.3(0.7)E              | 5.63(0.51)BC                         | 2.61(0.18)F                     | 2.43(0.15)DE           |
|                   | 5.7                                 | 14.0 (0.1)H                      | 25.2(1.3)F              | 4.79(0.15)D                          | 2.86(0.08)EF                    | 2.43(0.19)DE           |
|                   | 8.3                                 | 13.7 (0.2)H                      | 24.1(1.0)F              | 4.64(0.04)D                          | 2.85(0.16)EF                    | 2.27(0.15)E            |

<sup>a</sup> HDPE/BF = 60:40 wt/wt for all composites.

<sup>b</sup> The content of each modifier was based on the total composite weight.

<sup>c</sup> Numbers in the parentheses are standard deviation based on five specimens.

<sup>d</sup> Mean values with the same capitalized letter for each property are not significantly different at the 5% significance level.

adhesion between the components and the improvement of the nature of the matrix–filler interface [58]. It is, therefore, expected that analogous to PE-g-MA, sEPR-g-MA also promote interfacial bonding between hydrophilic BF and hydrophobic HDPE matrix, thereby resulting in improved tensile strength. Similar improvement in the strength via addition of functionalized polyolefin elastomers was also observed in SEBS-g-MA modified PE/wood flour [7,8,13,14] or PP/wood flour (or fiber) [15,16], and EBAGMA modified PP/wood flour composites [19], respectively.

With the further increase of the modifier content (up to 8.3 wt%), tensile strength was decreased progressively in both cases above. This trend is consistent with that observed in compatibilized HDPE/wood flour [7,8] and impact modified PP/wood sawdust [18] systems. This result may indicate that there seemingly exists a critical concentration of compatibilizers, beyond which the wetting saturation of the filler is achieved. Thus, a higher concentration may be not necessary to improve interface quality [18,59]. Unlike the formers, however, the introduction of the amorphous maleated elastomer (aEPR-g-MA) led to a completely opposite trend. The strength was firstly lowered with increased its concentration up to 5.7 wt% and then no significant decrease was observed with further addition of aEPR-g-MA. With almost equal MA grafting level for both EPR-g-MA, the following two reasons may contribute to this failure. The one possibility is that aEPR-g-MA failed to provide an interphase strong enough for stress transfer from the matrix to the filler to occur, or only existed as a third separate phase in the HDPE matrix rather than the encapsulating interface. Additionally, the inferior strength of aEPR-g-MA itself with respect to sEPR-g-MA might be partially responsible for this difference between both of EPR-g-MA modified systems.

Interestingly, the type and content of added modifiers affected the flexural properties in a manner similar to their effects on the above-mentioned tensile strength. At the same modifier's content level, the order of magnitude of flexural strength was ranked as follow: PE-g-MA > sEPR-g-MA > aEPR-g-MA. Also, the statistical ranking revealed that no significant difference in flexural strength when the concentration of each modifier varied from 5.7 to 8.3 wt% in three modified systems. For a given modified composite system, the value of flexural strength was much higher than the corresponding tensile strength.

Since polymer composites reinforced with natural fibers are mainly used in structural applications, their impact resistance is also one of the important concerns. Regardless of types of modifiers, the improvement in impact toughness with respect to the binary HDPE/BF composite was seen in all modified composites. The impact strength of composites containing either PE-g-MA or sEPR-g-MA increased with increasing content of the modifier and but the latter always exhibited superior values. Unexpectedly, with aEPR-g-MA as a modifier, the impact strength of the composites initially reached a maximum level at 2.9 wt% and then further addition of aEPR-g-MA caused a gradual decrease instead, although the overall impact strength of aEPR-g-MA modified composites was universally higher than that of unmodified one. However, it needed to note that the sources of enhanced toughness were probably different for both EPR-g-MA modified composite systems, if the variation of tensile and flexural strength between them was compared. It is known that the enhanced toughness of polymer–filler composites containing elastomers or rubbers can be realized by toughening either matrix or interfaces around fillers. Toughening of matrix tends to reduce ultimate strength of composites due to inferior bulk modulus of added elastomers, while strengths of final composites in the toughening of interfaces might be either decreased or increased, depending on interface nature. To recall that tensile and flexural strength of the composites increased with an increase in the concentration of sEPR-g-MA, the increased toughness should be largely associated with the formation of more flexible interphase

around bamboo flour in the latter case, thereby resulting in more energy absorption during impact fracture [13,15,16]. In the aEPR-g-MA modified composites, however, the lower strength with respect to the unmodified one implied that the enhanced impact strength could mainly result from the toughening of HDPE matrix.

Unlike tensile strength, tensile modulus is known to be less sensitive to interfacial interaction [9,60,61]. The addition of individual modifier led to the reduction in modulus to different extent, respectively. The composites modified with PE-g-MA exhibited a moderate reduction in tensile modulus, which is still much higher than that of the other modified systems over the whole concentration of the modifier. In contrast, even with addition of only small amounts of elastomeric EPR-g-MA, the modulus of HDPE/BF composites was considerably decreased. With further increase of their concentration, however, such reduction became less dependent on concentration of individual modifier. Likewise, the flexural modulus of the composites decreased with the addition of each modifier. Also, compared to PE-g-MA modified composites, much more reduction in the modulus was found in EPR-g-MA modified ones. This suggested that the excessive addition of maleated EPRs had an adverse effect on stiffness of the final composites.

DMA is a useful means to provide information on mechanical behavior, molecular relaxations as well as interactions taking place in the produced materials as the temperature is varied. The dynamic mechanical properties of composites are significantly dependent upon the amount of fiber, the presence of additive-like compatibilizer, filler, impact modifier, and fiber orientation [62]. For simplicity, Fig. 1 shows the temperature dependence of the storage modulus ( $G'$ ) and loss modulus ( $G''$ ) for HDPE/BF samples without modifiers and with various modifiers at the concentration of 5.7 wt%.

As expected, the magnitude of  $G'$  were remarkably increased with the incorporation of rigid BF into HDPE due to the enhanced stiffness. Among these composites, the sample containing no modifiers exhibited the maximum  $G'$  value over the whole temperature range, whereas the lowest  $G'$  was observed for aEPR-g-MA modified sample. This trend was in agreement with that obtained from the static mechanical data (Table 2). The curves of PE-g-MA and sEPR-g-MA modified HDPE/BF composites lied between both of former composites and differed slightly above around 40 °C. Usually, the enhanced adhesion between fillers and matrix tended to increase storage modulus of composites. In the case of either PE-g-MA or sEPR-g-MA modified composites, the improved interfacial

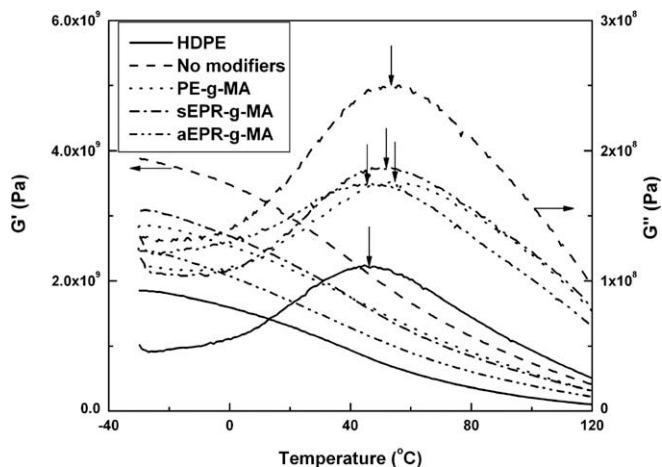


Fig. 1. Storage modulus ( $G'$ ) and loss modulus ( $G''$ ) of the unmodified and modified HDPE/BF composites with the concentration of modifier at 5.7 wt% (based on the total composite weight) as a function of temperature, together with virgin HDPE. For each composite, HDPE/BF = 60:40 (wt/wt).

adhesion was clearly confirmed via the mechanical properties and subsequent SEM observation. However, it was not this case based on the DMA results. Therefore, it was very likely that the adversely softening role of the low-modulus modifiers themselves exceeded over the positive reinforcing one resulting from improved interfacial adhesion on account of relatively higher content of added modifiers (5.7 wt%).

The dynamic loss modulus is a measure of the absorbed energy due to the relaxation and is useful in clarifying the mechanisms of internal motions. As shown in Fig. 1, a relaxation transition peak that occurred at about 50 °C was known as  $\alpha$ -relaxation of HDPE. It was thought to be related to a complex multi-relaxation process which was mainly concerned with the molecular motion of PE crystalline region [63]. The loss modulus at this relaxation temperature was markedly increased with the inclusion of BF. This enhancement was probably due to the presence of BF that reduced the flexibility of the material by introducing constraints on the segmental mobility of polymer molecules at the relaxation temperatures [64]. Likewise, the unmodified composite also exhibited the highest  $G''$  magnitude among the BF-filled composites, while the magnitude of  $G''$  differed less remarkably for modified composites. The  $\alpha$ -relaxation peak of the composites other than aEPR-g-MA modified one universally shifted to the higher temperature region as compared to virgin HDPE, while the addition of aEPR-g-MA caused the  $\alpha$ -relaxation position of HDPE almost unchanged. With the modification with PE-g-MA,  $\alpha$ -relaxation peak of HDPE matrix appeared to be the highest, suggesting the strongest restriction effect of the relaxation behavior.

The damping factor,  $\tan \delta$ , is the ratio of the loss modulus to the storage modulus. It should be noted that this parameter is independent of the material's stiffness and hence is a very good parameter when the differences in viscoelastic response of the material are desired. [65]. Fig. 2 presents the corresponding  $\tan \delta$  curves of virgin HDPE and the composites. It was found that  $\tan \delta$  is less distinctive than  $G''$  with regard to the  $\alpha$ -transition process. Over the investigated temperature range, aEPR-g-MA modified composite unexpectedly exhibited the highest magnitude of  $\tan \delta$ , followed by virgin HDPE. However, more broadening transition region of the former still implied inhibition effect of the relaxation process with the addition of BF. The damping factor curves between sEPR-g-MA modified composite and unmodified one almost overlapped, while PE-g-MA modified one had lowest magnitude of  $\tan \delta$ . The nature of interfacial layer and modifier location may be responsible for the difference. As indicated in previous mechanical

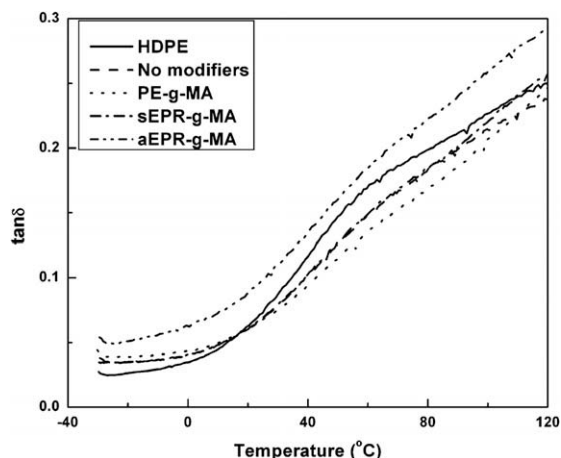


Fig. 2. Damping factor ( $\tan \delta$ ) of the unmodified and modified HDPE/BF composites with the concentration of modifier at 5.7 wt% (based on the total composite weight) as a function of temperature, together with virgin HDPE. For each composite, HDPE/BF = 60:40 (wt/wt).

data and the subsequent SEM images, PE-g-MA modified one achieved the strongest bonding between BF and HDPE, which allowed stiff BF other than the interface to bear a great extent of stress and less energy was dissipated in this case [64]. More rigid interfacial layer and better compatibility with the matrix could be also contributing factors in this composite. The elastomeric nature of sEPR-g-MA compromised the positive contribution, although the less improved interfacial adhesion was achieved. Therefore, sEPR-g-MA modified composite showed a similar  $\tan \delta$  as the unmodified one. However, the addition of aEPR-g-MA resulted in a weak interface situation. Based on the inferior strength results above, it was likely that aEPR-g-MA failed to act as an effective interface modifier but mainly separately disperse in the matrix, which in turn further coarsened final morphology of the composite due to the polarity difference between aEPR-g-MA and HDPE. As a result, the more viscous energy dissipation was evoked due to the enlarged interphase area.

### 3.1.2. Effect of combined modifiers

Data showing the effect of combined PE-g-MA and EPR-g-MA with a fixed concentration of both modifiers (i.e., 5.7 wt%) on mechanical properties of both composite systems is listed in Table 3. Statistical ranking showing treatment effect on each property is also shown in Table 3. As a control, mechanical properties of virgin HDPE and unmodified HDPE/BF composite were also given.

By adding a relatively small percentage of PE-g-MA, tensile strength of HDPE/BF/PE-g-MA/sEPR-g-MA composites initially increased significantly and then tended to level off, which was almost comparable to that of the HDPE/BF composite compatibilized with the same concentration of PE-g-MA alone. Flexural strength continuously increased with PE-g-MA percentage based on statistical ranking. The most noticeable feature was found for the result of notched impact strength. When relatively low percentage of sEPR-g-MA (i.e., 33.3 wt%) in combined modifiers was introduced, the composite even yielded higher impact strength than the composites modified with the same amount of sEPR-g-MA alone. But the tensile strength of the former was much superior due to the incorporation of PE-g-MA. At a higher percentage of sEPR-g-MA, the impact toughness also remained in an acceptable level (5–6 kJ/m<sup>2</sup>). Both moduli values followed a similar increasing trend except for the relatively lower value at 33.3 wt% sEPR-g-MA level. In the case of HDPE/BF/PE-g-MA/aEPR-g-MA composites, the same improvement in strength and moduli was visible with increasing PE-g-MA percentage and the superior impact strength was also achieved via the combination of PE-g-MA and aEPR-g-MA. At the same lower percentage of EPR-g-MA ( $\leq 50$  wt%), the aEPR-g-MA system showed somewhat lower impact strength than the sEPR-g-MA one, while the moduli were higher for the latter in this case. Based on the above results, the combination of PE-g-MA with sEPR-g-MA appeared to result in a better synergism in simultaneously maximizing both strengths and impact toughness of HDPE/BF composites.

In order to investigate effects of matrix nature of maleated EPR on dynamic mechanical properties of HDPE/BF composites containing combined PE-g-MA and EPR-g-MA (sEPR-g-MA vs. aEPR-g-MA), Figs. 3 and 4 present their viscoelastic moduli and damping factor as a function of temperature, respectively. In both cases, the total concentration of combined modifiers was fixed at 5.7 wt% and weight ratio of PE-g-MA to EPR-g-MA is kept at 2:1. It was found that larger  $G'$  value was achieved via the combination of PE-g-MA and sEPR-g-MA, indicative of higher stiffness. The peak temperature of  $\alpha$ -relaxation also shifted to a higher temperature by 8 °C, as compared to HDPE/BF composite modified by both PE-g-MA and aEPR-g-MA. On the other hand, the latter composite showed higher magnitude of  $\tan \delta$ . Based on the above discussion, this may suggest stronger interfacial interaction for the former composite [66].

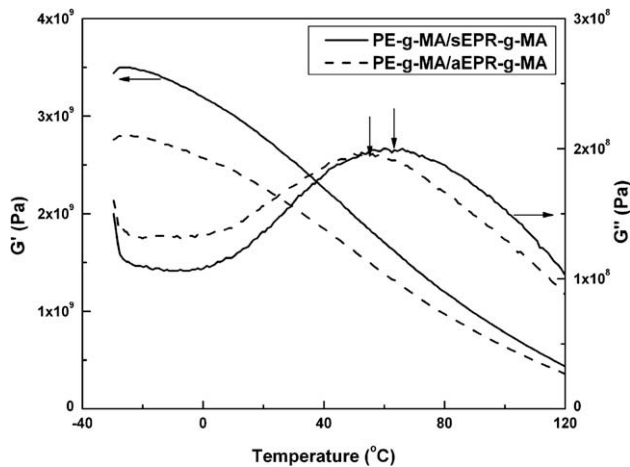
**Table 3**  
Effect of PE-g-MA percentage on mechanical properties of HDPE/BF composites modified with combined PE-g-MA and EPR-g-MAs<sup>a</sup>, together with those of virgin HDPE and the uncompatibilized HDPE/BF composite (60/40 wt/wt)

| PE-g-MA percentage (wt%)         | Strength property <sup>b,c</sup> |                         |                                      | Modulus property <sup>b,c</sup> |                        |
|----------------------------------|----------------------------------|-------------------------|--------------------------------------|---------------------------------|------------------------|
|                                  | Tensile strength (MPa)           | Flexural strength (MPa) | Impact strength (KJ/m <sup>2</sup> ) | Tensile modulus (GPa)           | Flexural modulus (GPa) |
| HDPE                             | 18.9(0.2)                        | 21.3(0.1)               | 8.12(0.21)                           | 0.30(0.10)                      | 0.80(0.01)             |
| HDPE/BF                          | 17.5(0.3)                        | 32.9(1.6)               | 2.88(0.10)                           | 3.64(0.26)                      | 3.12(0.14)             |
| <i>HDPE/BF/PE-g-MA/sEPR-g-MA</i> |                                  |                         |                                      |                                 |                        |
| 33.3                             | 25.9(0.2)F                       | 41.2(1.8)D              | 5.29(0.42)BC                         | 3.45(0.09)BC                    | 2.68(0.23)CD           |
| 50.0                             | 27.1(0.2)D                       | 44.4(0.8)BC             | 5.79(0.52)B                          | 3.50(0.16)ABC                   | 2.79(0.17)BCD          |
| 66.7                             | 27.6(0.2)C                       | 45.5(0.7)B              | 6.36(0.55)A                          | 3.18(0.08)C                     | 2.63(0.22)D            |
| <i>HDPE/BF/PE-g-MA/aEPR-g-MA</i> |                                  |                         |                                      |                                 |                        |
| 33.3                             | 23.4(0.1)G                       | 37.5(1.0)E              | 5.57(0.18)B                          | 3.50(0.19)ABC                   | 2.88(0.14)ABC          |
| 50.0                             | 26.6(0.5)E                       | 43.2(1.5)C              | 5.56(0.19)B                          | 3.57(0.53)AB                    | 2.87(0.22)ABC          |
| 66.7                             | 28.6(0.4)A                       | 45.7(1.1)B              | 5.45(0.29)B                          | 3.85(0.34)A                     | 3.04(0.15)A            |
| 100                              | 28.1(0.2)B                       | 48.1(0.5)A              | 4.90(0.24)C                          | 3.46(0.08)BC                    | 2.95(0.12)AB           |

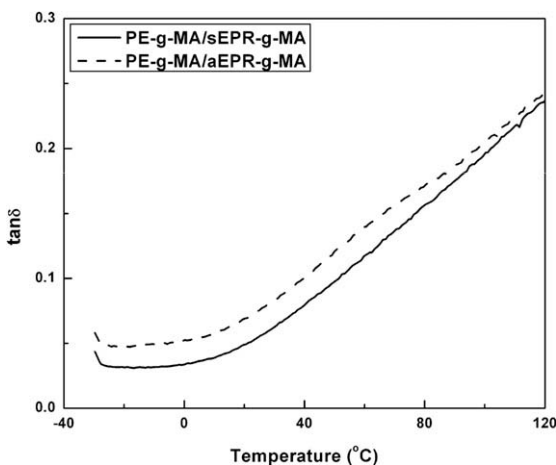
<sup>a</sup> The total concentration of the modifiers was fixed at 5.7 wt% (based on the total composite weight); HDPE/BF = 60:40 (wt/wt) for all composites.

<sup>b</sup> Numbers in the parentheses are standard deviation based on five specimens.

<sup>c</sup> Mean values with the same capitalized letter for each property are not significantly different at the 5% significance level.



**Fig. 3.** Storage modulus ( $G'$ ) and loss modulus ( $G''$ ) of HDPE/BF (60/40 wt/wt) composites modified with combined PE-g-MA and EPR-g-MA (sEPR-g-MA vs. aEPR-g-MA) as a function of temperature. For each case, the total concentration of combined modifiers was fixed at 5.7 wt% (based on the total composite weight); PE-g-MA/EPR-g-MA = 2:1 (wt/wt).



**Fig. 4.** Damping factor ( $\tan \delta$ ) of HDPE/BF (60/40 wt/wt) composites modified with combined PE-g-MA and EPR-g-MA (sEPR-g-MA vs. aEPR-g-MA) as a function of temperature. For each case, the total concentration of combined modifiers was fixed at 5.7 wt% (based on the total composite weight); PE-g-MA/EPR-g-MA = 2:1 (wt/wt).

### 3.1.3. Effect of bamboo flour loading

Data showing the effect of BF loading on mechanical properties of HDPE/BF composites with combined PE-g-MA and sEPR-g-MA system loaded at 15% of BF weight is summarized in Table 4, together with the statistical ranking information for each property. As a control, mechanical properties of virgin HDPE were also given.

Both strengths and moduli of the compatibilized composites significantly increased with bamboo flour loading according to statistical ranking results. However, the maximum level of impact toughness of the composites was observed at the medium loading of BF (i.e., 40 wt%). The reason may be attributable to relatively inadequate wetting or insufficient distribution of bamboo flour at the higher level of BF (50 wt%), which was caused by considerable agglomeration between the flour and remarkably increased viscosity of the composite [67–69]. As a consequence, the resulting voids at interfaces or large BF particles may more easily give rise to catastrophic crack growth during a high-speed impact test in contrast to a low-speed tensile measurement. On the other hand, at the lower BF loading (30 wt%), the relatively few BF bundles would perhaps contribute to the total energy dissipation during the impact fracture. The same optimum filler loading for notched impact strength was also reported for medium melt flow index HDPE (MMFI)-based composites filled with wood flake [67] and abaca fiber-reinforced PP composites without or with addition of PP-g-MA [69], respectively. At the same loading level of BF (40 wt%), the strengths and toughness of the modified composite with combined modifiers were higher than the corresponding uncompatibilized one. This was evidently as a result of compatibilizing role. But higher values in tensile and flexural moduli were true for the latter.

Storage modulus ( $G'$ ) and loss modulus ( $G''$ ) of modified HDPE/BF composites with different BF loading are shown in Fig. 5. As a comparison, unmodified HDPE/BF composite having 40 wt% BF is also included. A negligible effect on the magnitude of storage modulus was seen when BF loading level was raised from 30 to 40 wt%. With further addition of BF up to 50 wt% (about 51 vol %), the  $G'$  value was rapidly increased. Considering that the maximum packing fraction ( $\phi_m$ ) of randomly oriented fibers was 0.52 vol % [58], the formation of BF network due to their partial contact may be responsible for such a remarkable increase at the higher loading level. At the same BF loading level, the  $G'$  of the modified composite was lower than the corresponding unmodified one. As stated above, this was probably due to the inferior modulus of the added modifiers themselves. Like storage modulus, loss modulus of the composites showed a similar trend. Compared to the unmodified composite, however, the  $\alpha$ -relaxation temperature of HDPE in various modified ones was shifted toward higher temperature more or less.

**Table 4**

Effect of BF loading level of on mechanical properties of uncompatibilized and compatibilized HDPE/BF composites, together with those of virgin HDPE

| Systems                     | BF loading (wt%) | Strength property <sup>b,c</sup> |                         |                                      | Modulus property <sup>b,c</sup> |                        |
|-----------------------------|------------------|----------------------------------|-------------------------|--------------------------------------|---------------------------------|------------------------|
|                             |                  | Tensile strength (MPa)           | Flexural strength (MPa) | Impact strength (kJ/m <sup>2</sup> ) | Tensile modulus (GPa)           | Flexural modulus (GPa) |
| HDPE                        | –                | 18.9(0.2)                        | 21.3(0.1)               | 8.12(0.21)                           | 0.30(0.10)                      | 0.80(0.01)             |
| Uncompatibilized            | 40               | 17.5(0.3)D                       | 32.9(1.6)D              | 2.88(0.10)C                          | 3.64(0.26)A                     | 3.12(0.14)A            |
| Compatibilized <sup>a</sup> | 30               | 25.3(0.3)C                       | 39.8(1.4)C              | 5.25(0.29)B                          | 2.40(0.32)C                     | 2.09 (0.08)C           |
|                             | 40               | 27.6(0.2)B                       | 45.5(0.7)B              | 6.36(0.55)A                          | 3.18(0.08)B                     | 2.63 (0.22)B           |
|                             | 50               | 28.8(0.3)A                       | 49.6(1.1)A              | 5.21(0.19)B                          | 3.62(0.25)A                     | 3.20(0.13)A            |

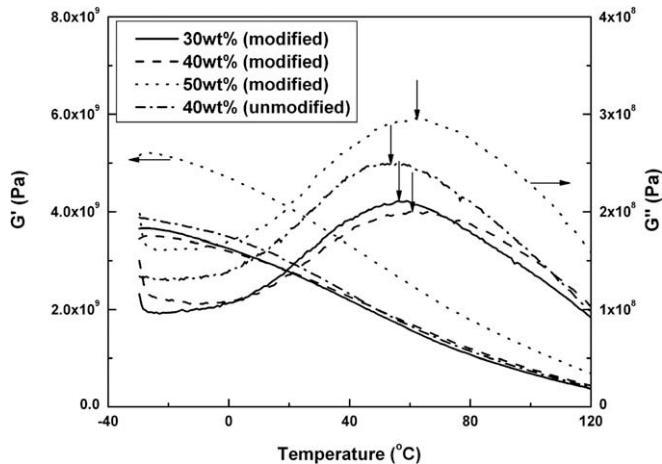
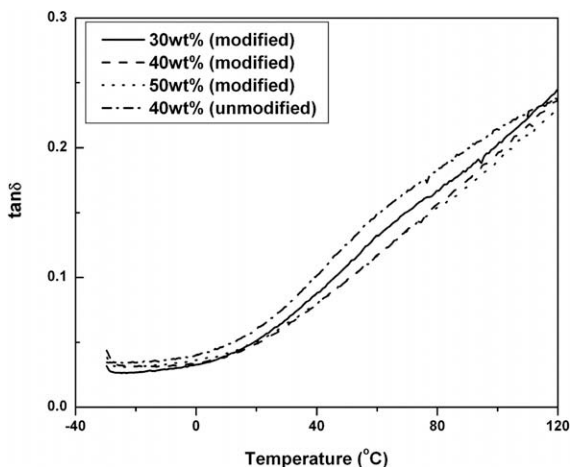
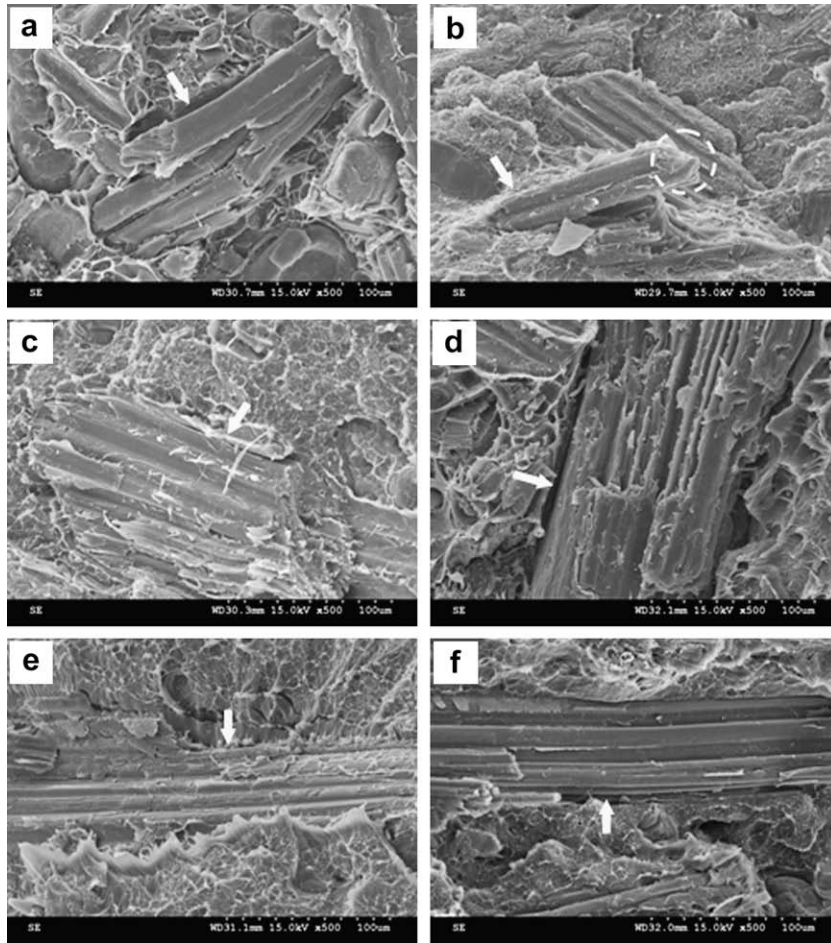
<sup>a</sup> PE-g-MA/sEPR-g-MA = 2:1 (wt/wt) for all compatibilized composites and the weight ratio of the combined modifiers to added BF was fixed at 15 wt%.<sup>b</sup> Numbers in the parentheses are standard deviation based on five specimens.<sup>c</sup> Mean values with the same capitalized letter for each property are not significantly different at the 5% significance level.**Fig. 5.** Effects of BF loading level on storage modulus ( $G'$ ) and loss modulus ( $G''$ ) of modified HDPE/BF composites with combined PE-g-MA and sEPR-g-MA, together with the unmodified HDPE/BF (60/40 wt/wt) composite. For each modified composite, the total concentration of both modifiers was fixed at 15 wt% (based on added BF weight); PE-g-MA/sEPR-g-MA = 2:1 (wt/wt).**Fig. 6.** Effects of BF loading level on damping factor ( $\tan\delta$ ) of modified HDPE/BF composites with combined PE-g-MA and sEPR-g-MA, together with the unmodified HDPE/BF (60/40 wt/wt) composite. For each modified composite, the total concentration of both modifiers was fixed at 15 wt% (based on added BF weight); PE-g-MA/sEPR-g-MA = 2:1 (wt/wt).

Fig. 6 shows the variation of  $\tan\delta$  with temperature for different composite samples. The  $\tan\delta$  was found to be decreased with an increase in BF loading from 30 to 40 wt%. The reason could be attributed to the restriction of the chain mobility by BF reinforcement, which raised the storage modulus values and reduced the

viscoelastic lag between the stress and the strain and hence decreased the  $\tan\delta$  values with an increase in the BF loading. With the further addition up to 50 wt%, the loss factor slightly changed instead. At the similar BF loading level, the  $\tan\delta$  values of the unmodified composite were seen to be higher than that of the modified composite. This result could be interpreted that for the unmodified composite, the poor interfacial bonding between BF and matrix tended to dissipate more viscous energy, thus exhibiting high magnitude of  $\tan\delta$  in comparison to the modified composite showing strongly bonded interface. All these differences in mechanical properties between three modified composite systems were closely correlated with their microstructures to be discussed in the later sections.

### 3.3. Morphological observation

Observation of the fracture surfaces of the composites by SEM can provide an insight into information related to interfacial adhesion and impact energy dissipation mechanisms involved during impact testing. Fig. 7 shows SEM graphs of impact fractured surfaces of HDPE/BF composites. Without addition of any modifiers, fiber bundles with smooth and clean surfaces were devoid of HDPE matrix, indicative of indeed poor interfacial adhesion between the filler and the matrix (Fig. 7a). Also, some traces can be visible where BF was pulled-out. With the addition of 5.7 wt% PE-g-MA, the broken fibers were seen to be embedded in the matrix without evident gap in the interfacial area and also their surfaces were seemingly covered with some polymer residuals (Fig. 7b). This was evidence of good interface bonding. In addition, there was less pronounced plastic deformation of the surrounding matrix involved. It suggests that the fracture of BF itself other than debonding is the main energy dissipation mode in this case. As shown in Fig. 7c, the increase in interfacial adhesion between BF and HDPE was not as effective as that achieved by PE-g-MA modified one, when sEPR-g-MA was added. And some voids can be seen at the interfaces. It should be noted that the signs for plastic deformation of local matrix appeared more evident for the latter case. The cavitations mechanism of elastomeric EPR-g-MA particles is probably responsible for it [16]. However, the introduction of aEPR-g-MA yielded a very coarse fracture surface and the interfacial situation seemed to quite resemble that of the uncompatibilized system, suggesting a formation of weak bonding between HDPE and the filler (Fig. 7d). When 3.8 wt% PE-g-MA was coupled with 1.9 wt% sEPR-g-MA as combined modifiers, the pull-out or breakage of fibers was not observed and the interfacial adhesion appeared strong, as indicated in Fig. 7e. Moreover the matrix deformation around the fillers became more pervasive, which was consistent with the highest impact strength among these composites. Compared to Fig. 7e, only less matrix deformation and relatively weaker interfacial bonding were observed when both PE-g-MA and aEPR-g-MA were used as combined modifiers (Fig. 7f). In order to better understand the difference in their



**Fig. 7.** SEM micrographs of impact-fractured surfaces of HDPE/BF composites. (a) Uncompatibilized HDPE/BF composite, (b) composite with PE-g-MA, (c) composite with sEPR-g-MA, (d) composite with aEPR-g-MA, (e) composite with PE-g-MA/sEPR-g-MA = 2:1 (wt/wt), and (f) composite with PE-g-MA/aEPR-g-MA = 2:1 (wt/wt), respectively. The total concentration of modifiers was fixed at 5.7 wt% (based on the total composite weight); HDPE/BF = 60:40 (wt/wt) for all composites.

mechanical properties of the above modified HDPE/BF composites, Fig. 8 shows the schematic morphological representations based on the above results and analysis. In Fig. 8a, the unmodified composite showed the poor adhesion between BF and HDPE matrix surfaces, and there are gaps around BF particles. In Fig. 8b, the enhanced interfacial adhesion was achieved when PE-g-MA, sEPR-g-MA or their combination were used as modifiers. The difference between both modifiers lied in the difference in the achieved degree of adhesion and nature of the resulting interphases (i.e., rigid vs. soft). Fig. 8c shows the microstructures for the composite with aEPR-g-MA, in which both BF and the majority of aEPR-g-MA were separately dispersed in HDPE matrix in this case. The poor adhesion existed not only at the BF/HDPE interface, but also at the one between polar aEPR-g-MA and non-polar HDPE. In the case of the combined PE-g-MA and aEPR-g-MA, the intermediate morphology is shown in Fig. 8d. The enhance adhesion between HDPE and BF was achieved, while poor adhesion was true for the aEPR-g-MA/HDPE interphase.

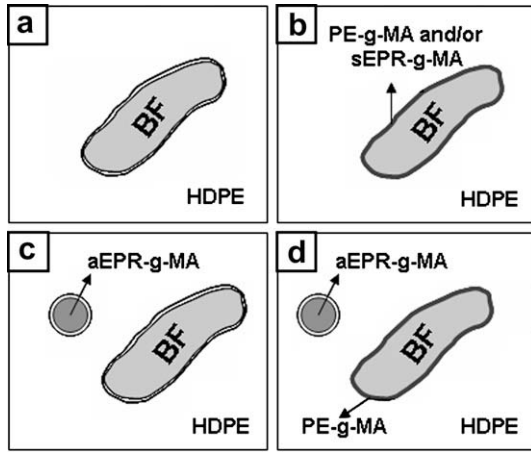
Fig. 9 shows the SEM graphs of impact-fractured surfaces of compatibilized HDPE/BF composite with varying BF loading levels. At the BF loading of 30 wt%, there were less noticeable gaps at the interfaces, even when some of fractured BF with incomplete coating with the resin was observed (Fig. 9a), implying still enough interfacial adhesion in this case. This may suggest that the imposed plastic constraint during fracture was less relieved or relaxed in this case, thereby exceeding the fracture strength of the fibers. When BF loading was increased to 40 wt%, more complete wetting

and homogeneous fractured surfaces were observed at the medium loading level. Moreover, compared to the former, fewer fibers were fractured, maybe suggesting less contribution of fiber-related energy in the latter. At higher BF loading (50 wt%), however, the fracture surface became inhomogeneous and the poorly encapsulated fibers were visible, suggesting less uniform dispersion of BF (Fig. 9b). Presumably, the remarkably increased melt viscosity at the high-BF loading may be responsible for this observation. This assumption has been partly confirmed by higher injection pressure needed for injection molding of test specimens. Therefore, it also confirms that the wetting and distribution of BF are critical for improving the impact performance of the composites [67]. These morphological observations well support the aforesaid mechanical results.

#### 3.4. Analysis of deformation mechanism

It is well-established that the fractured toughness of polymer composites arises from various sources of energy dissipation mechanisms, including fiber–matrix debonding, fiber pull-out, matrix shear yielding, and fracture of matrix and fibers [70,71]. Some of the energy dissipating events is fiber and interface related and the others are matrix related. However, major sources of toughness for the composites consisting of ductile matrix and short fibers or particle reinforcements have been attributable to shear deformation of matrix [16,72,73]. Kim et al. studies micromechanical deformation processes in toughened and particle-filled semicrystalline



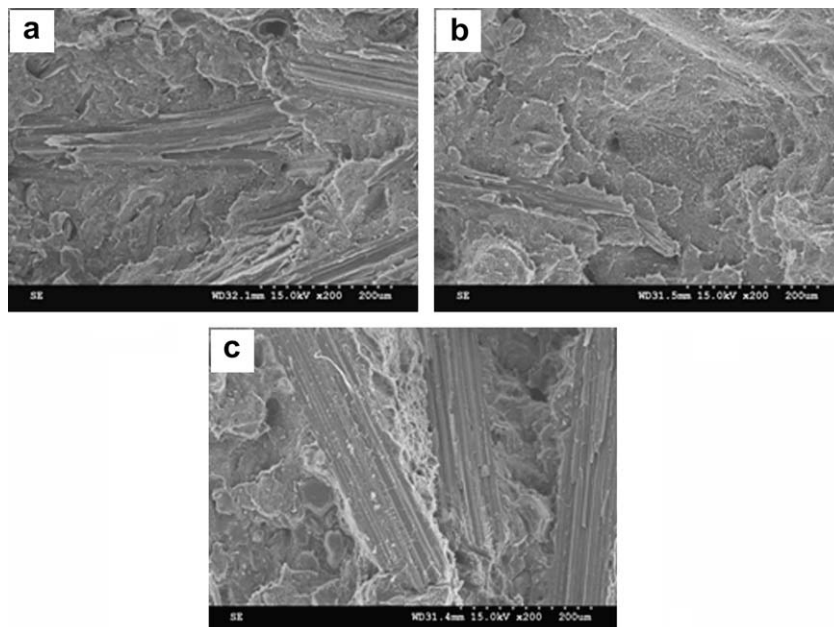


**Fig. 8.** Schematic representations of the unmodified and various modified HDPE/BF composites: (a) No modifiers; (b) PE-g-MA and/or sEPR-g-MA; (c) aEPR-g-MA; (d) PE-g-MA/aEPR-g-MA.

polymers and found that the microvoid formation played an important role for the activation of further plastic deformation of matrix material during deformation processes [72,73]. In rigid particulate-filled polymer systems, this microvoid formation is mainly caused by debonding at the interface or cavitation of rubbery shell layers around rigid fillers. In most cases, toughening effects depend largely on the degree of interfacial bonding and nature of interfaces. For a given composite system, the interfaces must provide enough adhesion for matrix deformation to be induced before the occurrence of pre-fracture. Namely, there existed critical interfacial bond strength.

By acting as stress concentrators, the rigid BF reduced the resistance to crack initiation in the composites. In the case of no modifiers, the cracks would rapidly propagate along the weakly bonding interfaces between BF and HDPE matrix, resulting in an ultimate breakage with very low-energy dissipation. As mentioned in the previous section, however, the addition of PE-g-MA yielded the maximum interfacial adhesion among three modifiers. More-

over, in this case, a relatively rigid coupling layer around the fillers was expected to be formed on account of backbone characteristics of the grafting polymers. Therefore, when the composite was subject to impact loading, plastic deformation of HDPE matrix was suppressed by the high-strain rate. Materials in front of the crack tip under this circumstance were subject to plane strain condition. The crack would propagate through the matrix with less plastic deformation. Because of strong interfacial bonding, some bamboo fibers on the crack path couldn't be even pull out but fractured, as shown in Fig. 7b. The fracture toughness may be mainly caused from BF-related energy, but less from matrix-related one. When sEPR-g-MA was added as a modifier, however, the premature crack propagation at the early stage of impact can be stabilized due to a certain degree of adhesion at interfaces and the relaxation of formed elastomeric interface. And the interlayer with the low bulk modulus was able to then cavitate under triaxial tension, which, in turn, locally released the plastic constraint imposed by high strain rate and the rigid fibers, thereby inducing plastic deformation in the HDPE matrix. As shown in Fig. 7c, the presence of sEPR-g-MA failed to afford interfacial adhesion as strong as PE-g-MA, which also assisted the occurrence of debonding at interfaces. It may also contribute to development of the above plastic deformation. On the other hand, the intrinsic characteristics of elastomeric sEPR-g-MA inevitably caused unsatisfactory strengths and a significant drop in stiffness. When PE-g-MA was coupled with small amounts of sEPR-g-MA as combined modifiers, however, the overall interfacial strength was somewhat weakened. It would allow the occurrence of microvoids to release the plastic constraint before the triaxial stresses reach the fracture stress of the matrix to cause brittle failure. Therefore, the impact toughness was significantly improved by massive plastic deformation. Meanwhile, the applied load was also transfer through interfaces due to the existence of majority of rigid PE-g-MA coupling layer and thus strengths and moduli of the composite were well maintained. On the contrary, in the absence of suitable adhesion strength, the cavitations or debonding failed to be effectively prevented and would result in ultimate failure of materials prior to matrix deformation. This may also provide possible explanation for relatively less matrix deformation in the case of HDPE/BF/aEPR-g-MA (Fig. 7d) and



**Fig. 9.** SEM micrographs of impact-fractured surfaces of HDPE/BF composite modified with combined PE-g-MA and sEPR-g-MA systems. (a) 30 wt% BF, (b) 40 wt% BF, and (c) 50 wt% BF. The total concentration of both modifiers was fixed at 15 wt% (based on added BF weight); PE-g-MA/sEPR-g-MA = 2:1 (wt/wt) for all composites.

HDPE/BF/PE-g-MA/aEPR-g-MA composites (Fig. 7f). In the former, weak adhesion was likely to prevail within the whole composite, whereas the poor adhesion could only exist between aEPR-g-MA and HDPE. As indicated in the previous section, the toughening of HDPE matrix resulting from the separated aEPR-g-MA phase is the fractured energy dissipation. Aside from the above energy dissipation, some part of fiber-related energy may be also involved in the latter owing to enhanced bonding of BF with the matrix.

Therefore, good interfacial adhesion between components is prerequisite to improve both strength and toughness of final composites. Neither very strong nor very weak interface was advantageous to achieve high fracture toughness, which confirmed the viewpoint of Kim and Mai [74]. Also, the nature of interfacial layers was another important factor to be considered.

#### 4. Conclusions

Semi-crystalline and amorphous EPR-g-MA, PE-g-MA, and their combinations were incorporated into HDPE/bamboo flour composites as interfacial modifiers. It was found that the incorporation of either PE-g-MA or sEPR-g-MA enhanced both strengths and impact toughness of the resulting composites, while aEPR-g-MA had a negative effect on tensile and flexural strengths despite moderate enhancement in impact toughness with respect to the uncompatibilized composite. Strength of the resultant composites modified by either PE-g-MA or sEPR-g-MA increased with the amount of the added compatibilizer. However, the effect began to level-off when its concentration exceeded 2.9 wt%. The maximum strength and impact toughness were achieved in PE-g-MA and sEPR-g-MA modified composites, respectively. The introduction of each individual modifier caused a drop in modulus of HDPE/BF composites to a different extent. At fixed loading level of BF (40 wt%) and total concentration of modifiers, the combination of PE-g-MA with small percentages of EPR-g-MA improved impact toughness while simultaneously maintaining strength of the composite at an acceptable level as compared to that compatibilized with either PE-g-MA or sEPR-g-MA alone. An optimum strength/toughness balance of composite properties can be achieved at lower percentage of sEPR-g-MA and medium BF-loading level. Dynamic mechanical analysis and SEM micrographs were well consistent with the abovementioned mechanical results. Based on the proposed morphological models, toughening mechanisms of various modified composites were discussed in detail. It was found that suitable interfacial adhesion and easiness of interlayer cavitations between HDPE and BF is beneficial to initiate massive matrix yielding in the composites, thereby achieving satisfactory impact strength.

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