Biodegradable poly(butylene adipate-co-terephthalate)/wheat gluten blends: Effect of PBAT modification on morphological, mechanical and water adsorption properties

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Abstract. Biodegradable polymer blends between poly(butylene adipate-co-terephthalate) (PBAT) and wheat gluten (WG) were prepared in various ratios of 70:30, 60:40 and 50:50 by weight. Modified PBAT with maleic anhydride (PBAT-g-MA) was used as a compatibilizer in this work. Morphological, mechanical and impact properties as well as water adsorption of the PBAT/WG blends with and without the compatibilizer were investigated. Scanning electron microscopic (SEM) analysis revealed that the PBAT/WG blends with PBAT-g-MA showed a better compatibility between PBAT and WG phases. Moreover, addition of the compatibilizer into the blends significantly improved tensile strength, elongation at break and impact strength compared to the PBAT/WG blends without compatibilizer. Furthermore, water adsorption of the blends with compatibilizer is lower than that of uncompatibilized PBAT/WG blends.

1. Introduction
Poly(butylene adipate-co-terephthalate) (PBAT) is a biodegradable aromatic-aliphatic copolyester with higher flexibility and elongation than PLA and PBS [1] and suitable for various packaging and agricultural applications [2]. However, a major drawback of PBAT is its high cost. In order to overcome the drawback, blending of PBAT with low cost polymers has been an alternative approach. In this work, wheat gluten (WG) was selected to blend with PBAT because of its large availability, low price and biodegradability [3]. Nevertheless, polarity of PBAT and WG is different leading to immiscible blend between PBAT and WG. Thus, in this work, maleic anhydride grafted PBAT, which was synthesized by reactive grafting process using Luperox® 101 as an initiator, was used as a compatibilizer between PBAT and WG phases. Morphological, mechanical and water adsorption properties of PBAT/WG with and without PBAT-g-MA compatibilizer were studies.

2. Experimental
2.1. Materials
PBAT (Ecoflex© F Blend C1200) with MFI of 3.8 g/10 min was purchased from BASF Corporation. WG was supplied from Anhui Ante Food Co., Ltd, China. Maleic anhydride (MA) and 2,5-Bis(tert-butylperoxy)-2,5-dimethylhexane (Luperox® 101) were provided from Sigma-Aldrich, Inc. Glycerol was obtained from Ajax Finechem PTY Ltd.
2.2. Preparation of PBAT-g-MA

PBAT-g-MA was prepared in a twin-screw extruder at 180°C with a screw speed of 80 rpm. MA and Luperox as an initiator were used at constant amount of 5 and 0.5 phr for grafting MA on PBAT chains.

2.3. Preparation of PBAT/WG blends

WG was plasticized by glycerol at content of 20 phr before mixed with PBAT. In the case of the compatibilized PBAT/WG blends, the PBAT-g-MA with amount of 10 wt% of PBAT was used. The PBAT/WG and PBAT/WG/PBAT-g-MA blends were blended in the twin screw extruder at 150°C with a screw speed of 60 rpm. The PBAT/WG ratios were varied at 70:30, 60:40 and 50:50 by weight. All specimens of the blends were heat-pressed by compression molding.

2.4. Characterization Methods

Morphologies of cryo-fractured surface of PBAT/WG blends with and without the PBAT-g-MA compatibilizer were observed by a scanning Electron microscope (TM3030, Hitachi, Japan). Tensile properties including maximum tensile strength and elongation at break of the PBAT, WG, PBAT/WG and PBAT/WG containing the compatibilizer were investigated by Universal testing machine (Instron 5969, Instron, USA.) according to ASTM D638 type IV. Izod impact testing was performed by Pendulum impact tester (CEAST 9050, Instron, USA.), following the procedure as described in ASTM D256-10. Water adsorption of PBAT, WG, PBAT/WG and PBAT/WG/PBAT-g-MA was determined by soaking the specimens in deionized water at a set time interval until 7 days. The percentage of weight change (WC) was calculated as followed equation: $WC(\%) = \left(\frac{W_s - W_o}{W_o}\right) \times 100$ where $W_s$ and $W_o$ are the weight of a specimen after and before soaked in the deionized water, respectively.

3. Results and discussion

3.1. Morphology characterization

The SEM was used in order to investigate the effect of the PBAT-g-MA compatibilizer on the phase morphology of the PBAT/WG blends. Figure 1 showed SEM micrographs of the cryogenically fractured surface of the PBAT/WG and PBAT/WG/PBAT-g-MA blends at ratios of 70/30, 60/40 and 50/50. Regards to the SEM images of the uncompatibilized PBAT/WG (Fig.1 (a)-(c)), the dispersion of WG was non-uniformed in the PBAT matrix with the large particle size of the WG as indicated in the SEM images. Moreover, the interfacial de-bonding was observed in all uncompatibilized blends. These evidences indicated poor adhesion between PBAT and WG phases. In the case of the PBAT/WG/PBAT-g-MA blends (Fig.1 (d)-(f)), the dispersion of WG phase became uniform with a smooth surface and without a presence of WG particles in PBAT matrix. This result could be attributed to improvement of interfacial adhesion between PBAT matrix and WG by addition of the PBAT-g-MA resulting in the better compatibility between PBAT and WG phases.

| PBAT/WG | 70:30 | 60:40 | 50:50 |
|---------|-------|-------|-------|
| No added compatibilizer | ![Image](image1.png) | ![Image](image2.png) | ![Image](image3.png) |
3.2. Mechanical and impact properties
The mechanical properties including maximum tensile strength and elongation at break and impact properties of PBAT, WG, PBAT/WG blends with and without the PBAT-g-MA were represented in Figure 2 (a)-(d), respectively. Regardless of adding the PBAT-g-MA compatibilizer, elongation at break and impact strength decreased as increasing WG content in the blends. As seen in Fig.2 (a)-(c), plasticized WG had high modulus and low elongation at break compared with PBAT. These results indicated that WG showed more brittleness behaviour and PBAT possessed more ductile characteristic. Thus, incorporation of stiffness WG polymer into PBAT could reduce elongation at break and impact strength in the PBAT/WG blends with and without the compatibilizer. It was found that the mechanical and impact properties, the compatibilized PBAT/WG blends were higher than those of the uncompatibilized blends. This could be attributed to reduced interfacial tension between PBAT and WG polymers, resulting from reaction of MA on PBAT-g-MA with hydroxyl and carboxyl groups of PBAT and interaction of MA of the compatibilizer with amino and hydroxyl groups of WG.

![Figure 1. SEM images of the PBAT/WG and PBAT/WG/PBAT-g-MA blends at the ratios of 70/30, 60/40 and 50/50 at magnification x 200.](image1)

![Figure 2. Mechanical and Impact properties of PBAT, WG, uncompatibilized PBAT/WG and compatibilized PBAT/WG blends at various ratios: (a) Maximum tensile strength, (b) Elongation at break and (c) Impact strength.](image2)
3.3. Water adsorption
Figure 3 represents percentage of weight change of plasticized WG, PBAT, PBAT/WG blends with and without PPBAT-g-MA after immersion in DI water at a set time interval. Among all specimens, the plasticized WG showed the highest %weight change after soaked in water, indicating poor water resistance of the plasticized WG due to a presence of hydrophilic characteristic of polar peptides (amino and carboxyl groups) in WG responsible for interacting with water via hydrogen bonding [4] and a presence of glycerol plasticizer added in WG. On the other hand, the hydrophobic PBAT provided the lowest water adsorption. The water weight change increased as an increase in polar plasticized WG content. Compared to the uncompatibilized PBAT/WG blends, the blends containing PBAT-g-MA compatibilizer gave the lower water weight change, attributed to the higher water resistance in the compatibilized PBAT/WG blends. This is because the polar groups such as amino and hydroxyl groups of WG reacted with MA in PBAT-g-MA compatibilizer, resulting in a reduction of the hydrophilic groups in the compatibilized blends.

![Figure 3. % weight change of plasticized WG, PBAT, PBAT/WG blends and PBAT/WG/PBAT-g-MA blends at ratios of 70/30, 60/40 and 50/50.](image)

4. Conclusions
Incorporating PBAT-g-MA as a reactive compatibilizer in the PBAT/WG blends had great effect on phase morphology, mechanical and impact properties as well as water adsorption of the blends. Addition of the compatibilizer into the PBAT/WG provides the better interfacial adhesion between two polymers leading to an enhancement of tensile, impact and water adsorption properties with respect to uncompatibilized PBAT/WG blends.

5. References
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Acknowledgement
The authors are thankful for the financial support from Department of Materials Science and Engineering, Faculty of Engineering and Industrial Technology, Silpakorn University.