Morphological and Mechanical Properties of Natural Rubber Compound/Poly(butylene succinate) Blend

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Abstract. Biodegradable poly(butylene succinate) (PBS)/natural rubber compound (NRC) blend was melt mixed using a two-roll mill. A weight ratio of PBS/NRC was 100/0, 90/10, 80/20, 70/30, 60/40 and 50/50. Morphological and mechanical properties were investigated. The results showed that the elongation at break increased with an increase of NRC content until the ratio of 80/20. While the tensile strength continuously decreased with an increasing amount of NRC. Moreover, the impact strength of the PBS/NRC blend increased until the ratio of 80/20 and tended to decrease in the further ratios. Morphology of PBS/NRC blend showed that NRC had good dispersion in PBS matrix up to 80/20. When the amount of NRC was increased, it became a larger particle due to an agglomeration, resulting in the mechanical properties.

1. Introduction
Thermoplastic elastomer (TPE) is a one of the polymer blends with a combination of rubber and thermoplastic. The category of TPE can be divided into two groups namely simple blends (SBs) and thermoplastic vulcanizes (TPVs). The advantages of blending elastomer and thermoplastic are that they are easy to process in melt mixing and creating a new type of materials with more anticipated properties. Hence, there are many attractive properties of the thermoplastic elastomer [1-6]. Typically, the final morphology of thermoplastic elastomer is the dispersion of rubber particles in thermoplastics matrix.

Natural rubber (NR) is a bio-based elastomer. It is has a high elasticity and good tear strength. There are many attempts to improve the toughness of polymeric matrix by natural rubber [2-5]. The example of some studies were reported in polystyrene [2], poly(lactic acid) [3-4], poly(butylene succinate) [5-6].

Nowadays, bioplastics have attracted a considerable interest to study and research on the previous report [1, 3-6]. The raw materials which make bioplastics are renewable resources such as cellulose, casein, collagen, soy protein, corn. This is the best way to decrease the plastic pollution because bioplastics can degrade and decompose in the natural environment. The examples of bioplastics are polycaprolactone (PCL), poly(lactic acid) (PLA), and poly(butylene succinate) (PBS). The most popular material in the bioplastic industry is PLA. However, it is brittle material. On the other hand,
PBS has good mechanical properties and is easy to process. PBS is synthesized from the condensation reaction between 1,4-butandiol and succinic acid. It has a polyester and linear structure. It has thermal stability up to 200°C. After use, PBS degrades into carbon dioxide and water. However, it is still an expensive cost. Hence, there are many attempts to increase the ability application of PBS i.e. polymer blends [5-7] and/or composites [8-9].

In this research, the biodegradable PBS and natural rubber compound (NRC) in an effective vulcanization system was melt blended in a two-roll mill. The effect of NRC content on the morphological and mechanical properties was investigated.

2. Materials and method

2.1. Materials

Natural rubber (NR, STR 5L), steric acid, zinc oxide (ZnO), tetramethyl thiuram disulphide (TMTD), N-cyclohexyl-2-benzoiazole sulfenamide (CBS), and sulphur (S) were used to prepare natural rubber compound (NRC) in an effective vulcanization system. All chemicals were purchased from Chareon Tut Co., Ltd., Thailand. Biodegradable PBS, BioPBSTM FZ71PM grade, was supplied by PTT MCC Biochem Co., Ltd.

2.2. Natural rubber compound preparation

Natural rubber (NR) and chemicals were mixed via a two-roll mill (Chareon Tut co., ltd., ML-D6L12-INV) with a roller speed of 10 rpm for 30 min in order to make NRC. The order of reagents is shown in Table 1. After being mixed, the NRC was kept in room temperature for a night before being blended.

| No. | Raw materials | Composition, phr | Mixing time, min |
|-----|---------------|----------------|-----------------|
| 1.  | Natural rubber | 100           | 5               |
| 2.  | Steric acid   | 2             | 5               |
| 3.  | ZnO           | 6             | 5               |
| 4.  | TMTD          | 3.5           | 5               |
| 5.  | Sulphur       | 0.75          | 5               |
| 6.  | CBS           | 1             | 5               |

2.3. Poly (butylene succinate)/natural rubber compound blend

PBS was dried in an oven at 50°C for 24 h before use. NRC and dried PBS were mixed with a two-roll mill plastic (Chareon Tut co., ltd., ML-D6L12HT-INV) at 120°C for 20 min. The ratio of PBS/NRC was 100/0, 90/10, 80/20, 70/30, 60/40, and 50/50. The total amount of each batch was 500 g. After completely being mixed, the PBS/NRC blend was cooled to room temperature and ground with a grinder.

2.4. Hot-compression process

The ground PBS/NRC blends were dried in an oven at 50°C for 24 h before hot-compression process. Hot-compression machine (Chareon Tut co., ltd., PR2D-W300L350 PM-WCL-HMI) was used to prepare samples at a temperature of 145°C and pressure of 1500 psi in order to investigate the mechanical properties. The pre-heat time, pressing time and cooling time were 5, 5, and 3 min, respectively. The sample for mechanical testing was prepared according to ASTM D638 for tensile test and ASTM D256 for impact test.
2.5. Testing and characterization

2.5.1. Mechanical testing. For impact test, the notched Izod impact type was conducted according to ASTM D256 at room temperature by impact tester machine (INSTRON, Ceast 9050). At least ten samples were tested and evaluated. For tensile test, the universal testing machine (UTM, LLOYD INSTRUMENT, LR10K plus) was used with a crosshead speed of 50 mm/min at room temperature.

2.5.2. Morphological study. The fracture surface from impact sample was investigated by using scanning electron microscope (JEOL, JSM-6510).

3. Result and discussions

3.1. Mechanical testing.
In this research, the mechanical properties of poly(butylene succinate)/natural rubber compound (PBS/NRC) blends were investigated in terms of impact and tensile tests.

For impact test, the impact strength of PBS/NRC blends is illustrated in Figure 1. The results showed that the impact strength of neat PBS was 6.2 ± 0.6 kJ/m². After NRC blending, the PBS/NRC blends in the ratio of 90/10, 80/20, 70/30, 60/40, and 50/50 showed the impact strength of 8.61 ± 1.0, 12.09 ± 1.4, 11.3 ± 1.2, 9.2 ± 1.2, and 7.8 ± 0.8 kJ/m², respectively. The highest impact strength was the PBS/NRC blends of 80/20. This was a 2-fold enhancement compared with the neat PBS. The reason of this improvement was that the NRC acted as stress concentration in polymeric matrix. It is a toughened polymer matrix similar with a previous report [6].

![Figure 1. Impact strength of poly(butylene succinate)/natural rubber compound (PBS/NRC) blends](image)

For tensile test, the elongation at break and tensile strength are illustrated in Figure 2. The elongation at break of PBS/NRC blends showed that the NRC contributed to improve this value (Figure 2a). The highest elongation at break of the blends was 29.1 ± 4.6 percentage in the ratio of 80/20. It was enhanced to 1.7 times compared with the neat PBS. This is due to the elastic property of NRC similar to the polymer-rubber system [6]. On the other hand, the incorporation of NRC significantly decreased the tensile strength of PBS (Figure 2b). As expected, tensile strength decreased with an increasing amount of NRC.
Figure 2. Tensile properties of poly(butylene succinate)/natural rubber compound (PBS/NRC) blends; (a) elongation at break and (b) tensile strength

3.2. Morphological study.
Figure 3 illustrates the SEM micrographs of the fractured surface of PBS/NRC blends with various blend ratios. The result showed that the coarse surface of pure PBS could be observed (Figure 3a). All blends illustrated the dispersed NRC particles in the PBS matrix (Figure 3b-f). Especially, the blends with 10 and 20 wt% of NRC exhibited the dispersed particle size of 2-5 µm as shown in Figure 4b-c. When the blends had a higher NRC content, the large dispersed particle size occurred due to the agglomeration of rubber particles as seen in Figure 3d-f. These morphologies were involved with the mechanical properties as previous discussion. From the mechanical and morphological properties, the PBS/NRC of 80/20 was the best blend ratio.

Figure 3. Morphology of PBS/NRC blends with a weight ratio of; (a) 100/0, (b) 90/10, (c) 80/20, (d) 70/30, (e) 60/40 and (f) 50/50

4. Conclusions
PBS/NRC blends with various weight ratios of 100/0, 90/10, 80/20, 70/30, 60/40 and 50/50 were prepared via a two-roll mill. The amount of NRC had an effect on the mechanical and morphological properties of the blends. The highest impact strength and percentage elongation at break were achieved at 20 wt% of NRC. The tensile strength decreased monotonically with increasing NRC content. Finally, NRC might be used to enhance toughness of other brittle polymer matrix.
5. References

[1] Maccaferri E, Mazzocchetti L, Benelli T, Brugo T M, Zucchelli A and Giorgini L 2020 Mater. Design. 186 108210
[2] Asletha R, Kumaran M G and Thomas S 1999 Eur. Poly. J. 35 253-271
[3] Pongtanayut K, Thongpin C and Santawitee O 2013 Energy Procedia 34 888-897
[4] Dechatiwon Na-Ayutthaya W and Poompradub S 2014 Macromol. Res. 22 686-692
[5] Faibunchan P, Nakaramontri Y, Chueangchayaphan W, Pichaiyut S, Kummerlöwe C, Vennemann N and Nakason C 2018 J. Poly. Environ. 26 2867-2880
[6] Faibunchan P, Pichaiyut S, Chueangchayaphan W, Kummerlöwe C, Vennemann N and Nakason C 2019 Poly. Adv. Technol. 30 1010-1026
[7] Hemsri S, Thongpin C, Moradokpermpoon N, Niramon P and Suppaso M 2015 Macromol. Symp. 345 145-154
[8] Huang Z, Qian L, Yin Q, Yu N, Liu T and Tian D 2018 Polym. Test. 66 319-326
[9] Calabia B P, Ninoomiya F, Yagi H, Oishi A, Taguchi K, Kuioka M and Funabashi M 2013 Polymers 5 128-141

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