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Properties of Biodegradable Poly(butylene succinate) Filled with Activated Carbon Synthesized from Waste Coffee Grounds

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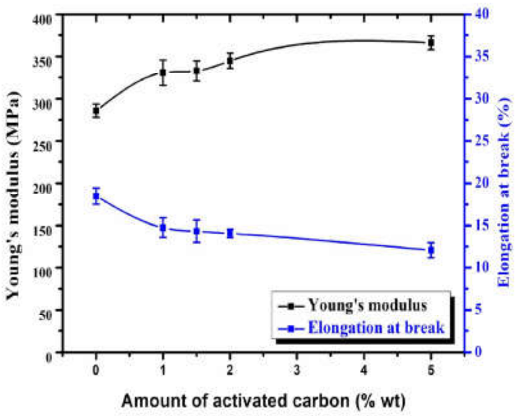
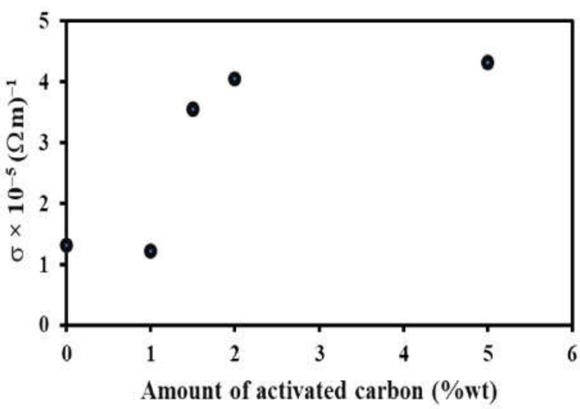
Abstract

Activated carbon (AC) synthesized from waste coffee grounds by using microwave irradiation method was mixed with poly(butylene succinate) (PBS) in a batch internal mixer. The concentrations of AC were used at 0, 1, 1.50, 2 and 5 %wt. The mechanical properties of PBS/AC composites, such as tensile strength, impact strength and hardness were investigated. The electrical and morphological properties of neat PBS and their compounds were examined. Moreover, the crystal structure of PBS/AC composites was observed by the XRD technique. The result showed that AC could enhance the Young's modulus and hardness of PBS. The impact strength, tensile strength, and elongation at break decreased with an increase in the volume of AC. The crystal structure of neat PBS was changed with the AC concentration. The AC content also affected the electrical conductivity. The concentration of AC at 1.50 %wt was the percolation threshold. On the other hand, the addition of AC led to smooth fractured surface of the composites.





Keywords: Poly(butylene succinate); Activated carbon; Mechanical property; Electrical conductivity

Introduction

Nowadays, the awareness of plastic waste pollution around the world is concerning. As well known, plastic waste takes a long time to decompose. Hence, one of the choices to decrease plastic pollution is the research and development of biodegradable polymer [1 – 3]. Poly(butylene succinate) (PBS) is generated from condensation polymerization between 1, 4-butanediol and succinic acid [4]. They are mainly produced from renewable agricultural resources such as corn, soybean [5, 6]. PBS can be a good alternative to common plastics such as polyethylene and polypropylene [5, 6]. It has gained acceptance in many applications, such as food packaging, bags, agricultural film and many other market applications [7, 8]. PBS shows biodegradability, thermal and chemical stability, and good mechanical properties. The biodegradability of PBS is an attractive attribute for single-use food packaging because it can degrade at a high rate over short periods [9]. However, the major drawback is its high cost [10]. Hence, one of the methods to reduce its price is the addition of filler. There are many kinds of research studies on organic and inorganic fillers [1 – 3, 11]. For example, Luo et al. studied the mechanical and thermal performance of distillers grains filled poly(butylene succinate) composites [3]. They stated that the distiller's grains had the potential to decrease the cost of PBS-based materials with a variety of enhanced properties.

Activated carbon (AC) is a highly porous carbonaceous material. It has a high adsorption capacity thus is used as an adsorbent in many applications, such as pharmacy, cosmetics or catalyst in industry as a part of the membrane for purification system [12, 13]. Moreover, it has been used as filler reinforcement and conducting material in a polymeric matrix [14, 15]. At present, waste coffee grounds tend to increase due to the increase of coffee shops in Thailand, hence, more waste coffee grounds are generated. Therefore, this research attempted to recycle waste coffee grounds to produce activated carbon for use as filler reinforcement in a biodegradable polymer for various applications, such as mulch film and food packaging. An example of using AC in the polymer was recently reported in epoxy resin. The nano-AC synthesized from arhar crop was mixed in the epoxy matrix. It was found that the amount of 2% filler loading showed the best mechanical properties due to the maximum tensile strength [16].

In line with the main idea above, this study focused on the effect of AC content on the mechanical and electrical properties, morphology and crystal structure of PBS. The loading of AC was in a range of 0 – 5 %wt.

Materials and Methods

In this research, two raw materials were used. The PBS (Poly(butylene succinate) (BioPBS^{TM} FZ71PM grade) was procured from PTT MCC Biochem Company Limited. The activated carbon (AC) was synthesized from waste coffee grounds using microwave irradiation. In the first step, the coffee residue was carbonized at 400 °C for 1 h. Charcoal at 400 °C was immersed in 1 molar of potassium hydroxide (KOH) solution for 24 h. After that, it was activated by microwave radiation at 1,000 W for 3 min. The microwave irradiation transmitted a high-frequency electromagnetic wave which caused vibration in the charcoal molecule and transformed it into activated carbon. The vibration simultaneously generated heat which was capable of water evaporation and volatility in charcoal. The AC phase was confirmed by X-ray diffraction method which showed the amorphous carbon phase. The average particle size of AC was 12.02 ± 6.58 µm which was measured by laser particle size instrument (ANALYSETTE 22 NeXT, Fritsch).

In the preparation of PBS/AC composites, PBS and AC had their moisture reduced in a stove at 50 °C for 24 h. Before compounding, they were shaken in a premixed bag for 10 min. Then, the PBS/AC composites were melted and mixed inside of an internal mixer (MX500–D75L90, Chareon Tut Co., Ltd., Thailand) with a rotor speed of 50 rpm at 145 °C for 15 min. After cooling, the composites were ground by plastic grinder machine. The specimen was prepared by pressing the ground composites using hot compression molding machine at 145 °C with 1,500 psi pressure. Three steps of the specimen preparation consisted of pre-heating for 5 min, then pressing for 3 min, and then cooling for 3 min.

For mechanical testing, tensile properties were investigated by the Universal testing machine (LS Plus Series, Lloyd). The dumbbell-shaped samples were prepared according to ASTM D638 Type I. The tensile testing was operated under an ambient condition with a 10 kN load cell and a crosshead speed of 50 mm min^{-1}. Five samples were executed for each ratio. For the impact test, a notched Izod impact type was managed according to ASTM D256 at room temperature by an impact tester machine (Ceast 9050, INSTRON). The hardness test was conducted according to ASTM D2240 using a durometer (GS–612, TECLOCK) and expressed as Shore D hardness value.

The approximate sample size of 1.27 × 1.27 × 3 mm^3 was obtained by hot compression molding and used for the electrical properties measurement. The sample was coated on both sides with silver paint and dried for 12 h before measurement with a precision LCR meter (E4980A, Agilent) at a frequency of 1 kHz.

X-ray diffraction (XRD-6100, Shimadzu) was used to explore the crystal structure of PBS/AC composites with a range of 2θ from 10 – 50° and CuK_α (λ = 0.15418 nm).

The scanning electron microscope (SEM Quanta 250 W7, Philips) was also used to show and investigate the fractured surface for the impact test.

Results and Discussion

Mechanical Properties of PBS/AC Composites

The tensile properties of the PBS/AC composites with various AC concentrations are reported in Table 1. Young's modulus of PBS/AC composites increased with an increase in AC quantity while the percentage elongation at break and the tensile strength decreased with an increase in the amount of AC. The deterioration of the elongation at break and tensile strength might have been due to the twining of PBS chains which destroyed the original structural integrity of PBS for elongation at break and poor interfacial adhesion of AC particles and polymer matrix for tensile strength [3]. The tensile properties were investigated and showed a similar tendency with different fillers to strengthen PBS-based composite [2, 3].

Table 1 Tensile properties of neat PBS and PBS/AC composites.

Amount of AC (%wt)	Young's modulus (MPa)	Tensile strength (MPa)	Elongation at break (%)
0	286 ± 8	35.33 ± 1.17	18.48 ± 0.96
1	331 ± 15	32.16± 0.45	14.75 ± 1.17
1.50	333 ± 12	32.13 ± 1.11	14.32 ± 1.33
2	345 ± 9	30.00 ± 1.28	14.07 ± 0.51
5	366 ± 8	29.62 ± 1.56	12.05 ± 0.88

Figure 1 shows the average hardness (Shore D) of all samples which relied on the concentration of AC. The outcome illustrated that the hardness of PBS was 67.32 ± 0.83. After adding AC content of 1%, 1.50%, 2% and 5%wt, result showed the hardness of 69.76 ± 0.95, 71.92 ± 0.73, 72.00 ± 0.65 and 73.20 ± 0.79, respectively. The hardness of the composites increased with an increase in the AC content. This enhancement might be due to the stiffness of AC influence on Shore D hardness of PBS composites.

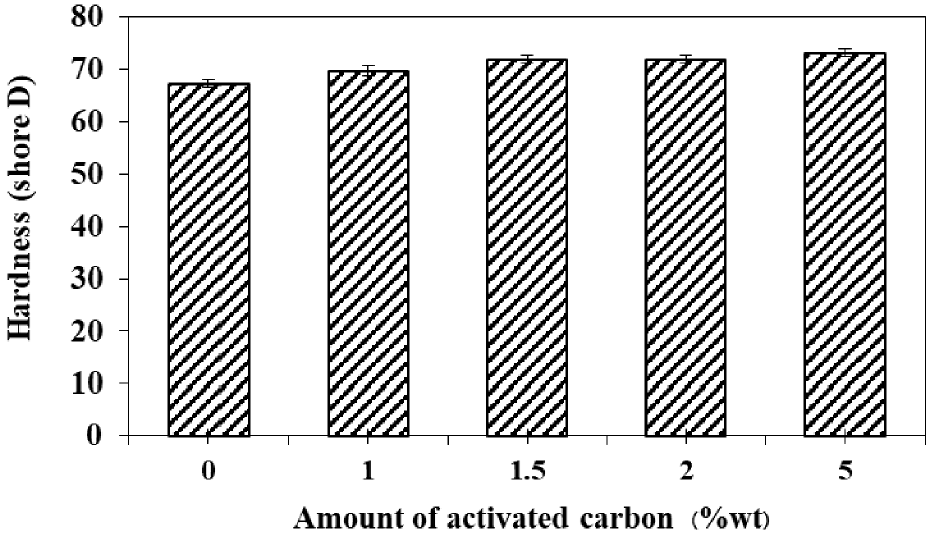


Fig. 1 Hardness of PBS/AC composites with various AC concentrations.

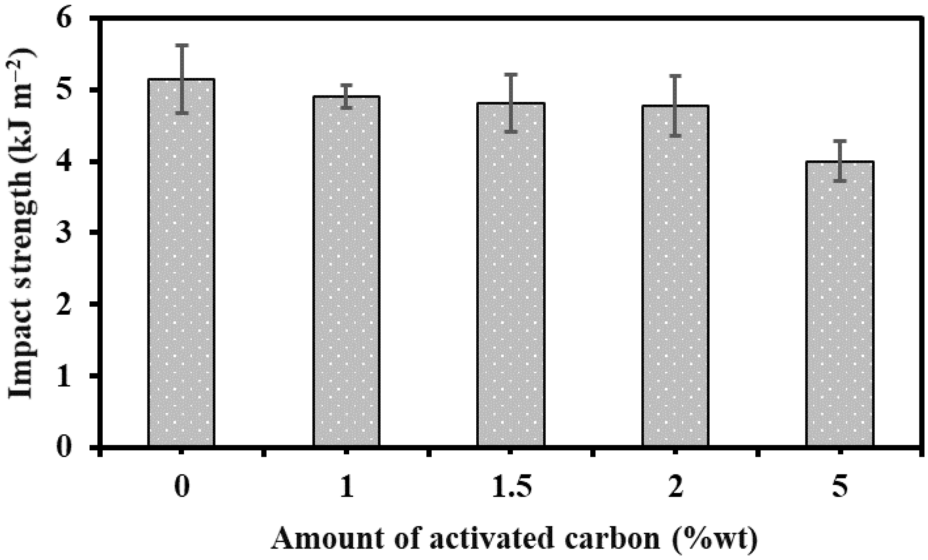


Fig. 2 Impact strength of PBS/AC composites with various AC concentrations.

Electrical Property of PBS/AC Composites

The electrical conductivity of PBS/AC composites is illustrated in Fig. 3. The result revealed that the electrical conductivity of neat PBS was 1.31 × 10^{-5} (Ω m)$^{-1}$. After the addition of AC, the electrical conductivity of PBS/AC composites at the AC content of 1%, 1.50%, 2% and 5 %wt were 1.22 × 10^{-5}, 3.56 × 10^{-5}, 4.05 × 10^{-5} and 4.32 × 10^{-5} (Ω m)$^{-1}$, respectively. It could be seen that 1%wt AC did not affect the electrical conductivity of the PBS/AC composites. The reason for this is because the content of 1%wt AC was still lower than the percolation threshold [17]. However, a further AC content could increase the electrical conductivity of the composites. This enhancement might be due to the higher electrical conductivity of AC.

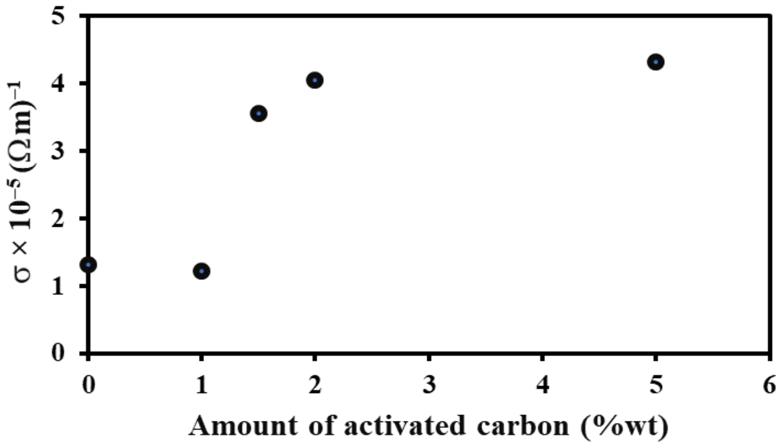


Fig. 3 Electrical conductivity (σ) of PBS/AC composites with various AC concentrations.

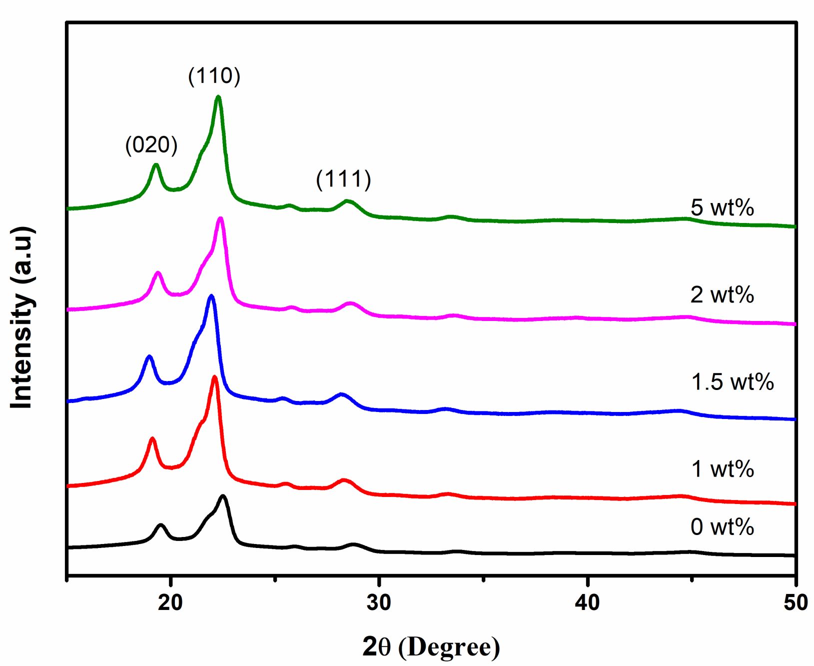


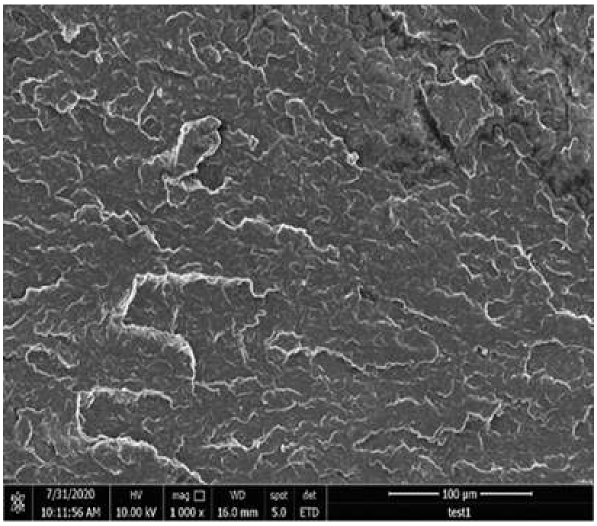
Fig. 4 XRD patterns of PBS/AC composites with various AC concentrations.

Crystal Structure of PBS/AC Composites

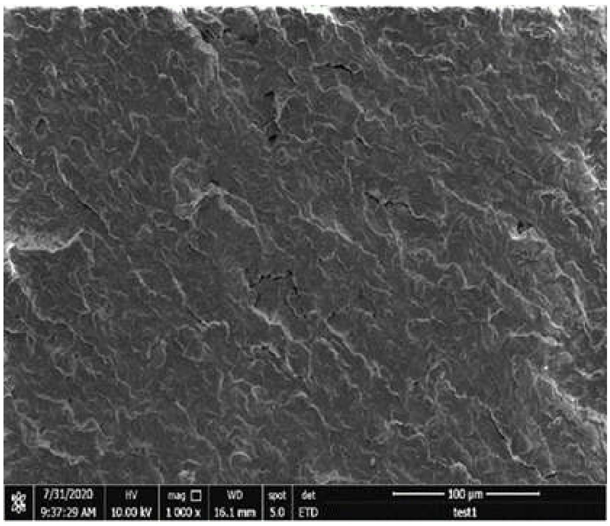
XRD patterns of PBS and its composites are shown in Fig. 4. Three main peaks located at around 19.35°, 22.38° and 28.79° are assigned to (020), (110) and (111) planes of PBS, respectively [18]. The composites showed a slight shift of all peaks to the lower angle, indicating that the d-spacing increased with an increase in the AC content. A similar result was found in the low-density polyethylene/montmorillonite nanocomposites system [19].

Morphology of PBS/AC Composites

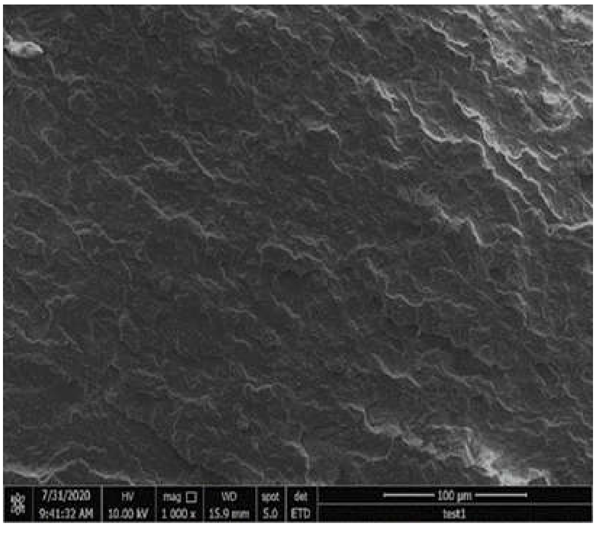
The SEM micrographs of the fractured surface of PBS/AC composites with different AC concentrations are shown in Fig 5. It could be seen that the neat PBS displayed a rough surface and many tear lines which is a character of a ductile material (Fig 5(a)). On the other hand, a smooth surface could be observed when the composites had a higher AC content as seen in Fig 5(b) – (e). This characteristic corresponded with the decrease of impact property as previously discussed.



(a)



(b)



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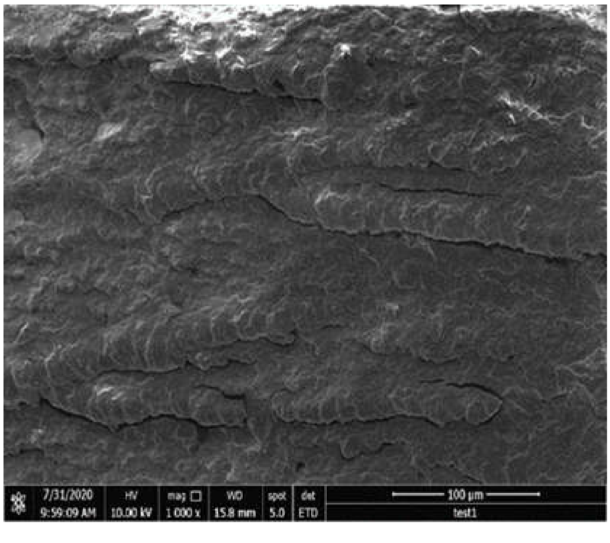

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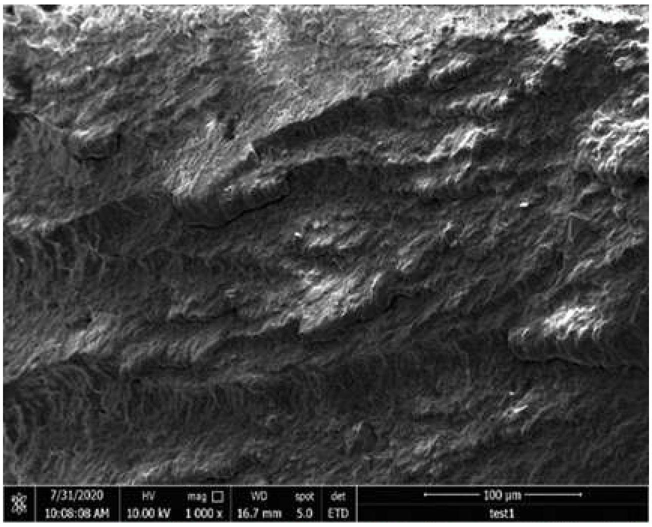

(e)

Fig. 5 SEM micrographs of PBS/AC composites with an AC content of: (a) 0 %wt, (b) 1 %wt, (c) 1.50 %wt, (d) 2 %wt, and (e) 5%wt.

Conclusion

The effects of AC content on the mechanical and electrical properties, morphology and crystal structure of PBS were investigated. The d-spacing of the crystal structure of PBS composites slightly increased when the AC content was raised. The increasing of AC content was objected to generate a smooth surface. Not only Young's modulus but also the hardness of PBS/AC composites were increased with an increase in the AC content. Hence, AC could be used as a filler reinforcement in the PBS matrix. On the other hand, the tensile strength, elongation at break, and impact strength of composites decreased. Moreover, the addition of AC could increase the electrical conductivity of PBS. The percolation threshold of PBS/AC composites was at the AC concentration of 1.50 %wt. In the future, the study of biodegradation is some of challenges for future work.

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