Research Article

Antimicrobial, Conductive, and Mechanical Properties of AgCB/PBS Composite System

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Demand for environmentally friendly plastic materials which are obtained from renewable resources such as biomass-based polyesters is of concern. Herein, the enhanced characteristic performances of poly(butylene succinate) (PBS) by employing the fabrication of PBS-based composites with the nanosilver-coated carbon black (AgCB) using an injection-molding method are reported. The preformed AgCB additives are priorly prepared by the benzoxazine oxidation method. Phase characterization of the obtained composite materials examined by X-ray diffraction (XRD) reveals the crystalline PBS matrix and the presence of metallic silver particles, confirming the successful fabrication of the composite materials. Detailed analyses on thermal, mechanical, electrical, and antimicrobial properties of the composite materials are reported. The AgCB-PBS composite materials provide such potential features by an enhancement of electrical conductivity and the antimicrobial activity by an inhibition against *E. coli* and *C. albicans*. These AgCB-PBS composite materials show the possibility to be an option for antielectrostatic and antimicrobial applications such as for the production of smart, environmentally friendly keyboards.

1. Introduction

Nowadays, pollution caused by tremendously large amount of usage and disposal of petroleum-based plastic materials is of concern all over the world [1–3]. Moreover, demand for reusing and also value adding of industrial and agricultural by-products are crucial, especially in developing countries [4, 5]. Therefore, environmentally friendly materials obtained from renewable resources have attracted significant attention. Biomass-based polyesters such as poly(lactic acid)

(PLA) [6, 7], poly(3-hydroxybutyrate) (PHB) [8, 9], and poly(butylene succinate) (PBS) [10–12] are among such materials. In particular, PBS has received enormous attention as it is gradually biodegraded in the landfill or sea in the presence of bacteria or fungi [13]. Moreover, PBS can be processed as easily as commercially available, petroleum-based plastics by using a variety of techniques such as blown films, fibers spinning, injection molding, thermoforming, or blow molding [14]. However, its potential for extensive applications has been limited by its slow melt viscosity as well

as a high cost of production of PBS-based products [15, 16]. To reduce the production cost and to improve the characteristic features of PBS, several strategies have been employed including blended or composite materials consisting of the PBS polymer matrix and a variety of biodegradable additives such as cellulose [17, 18], various plant fibers [19–22], starch [23–25], and soy protein [26, 27].

As mentioned that additives could improve the properties of polymers, several organic and inorganic fillers have been used as the additives for the preparation of polymer-based composites. Carbon black (CB) is one of the most popular additives that are known for improving the mechanical properties of polymer matrices, such as tensile strength, tear strength, hardness, and abrasion resistivity. Apart from these properties, its inexpensive cost makes CB suitable for large-scale production. Furthermore, CB has been shown to increase the electrical [28–30] and thermal conductivities of the composite materials [31].

Silver nanoparticles are renowned for their incredible physical, chemical, and biological properties [32]. Due to its high thermal stability and ability to inhibit the growth of various microorganisms, silver nanoparticles exhibit potentially useful addition as another additive for PBS [33, 34]. Our research group has developed a new method for preparing silver nanoparticles and then coating the nanoparticles onto the different substrates using benzoxazine as a reducing agent [35]. Different types of substrates, i.e., SiO_2 , TiO_2 , glass slides, polycarbonate (PC) sheets, polyvinyl alcohol (PVA), and chitosan (CS) powder as well as carbon black (CB) powder, were successfully coated with silver nanoparticles using this method [35]. The procedure occurred easily at room temperature, and any solvent or medium can be used. Moreover, this method did not disturb the structure of substrates, so the characteristic properties of the materials remain. Therefore, the properties of silver nanoparticles can be added without altering the original features of the substrates.

In this paper, enhanced PBS characteristics for antimicrobial, conductive, and mechanical properties have been examined by preparing PBS-based composites between PBS as the polymer matrix and carbon black (CB) or nanosilver-coated carbon black (AgCB) as its additive. The AgCB additive was priorly prepared by using a benzoxazine oxidation reaction for using as a filler. Two types of PBS-based composites were fabricated, namely, carbon black blended in PBS (CB-PBS) and nanosilver-coated carbon black blended in PBS (AgCB-PBS), varying the percentage of the additives for 3, 5, 10, and 15%. Note that the notations of $x\text{CB-PBS}$ and $x\text{AgCB-PBS}$ (x is the percentages of the additives) are used throughout this article. The phase formation of the novel composites was characterized by X-ray diffraction (XRD). Details on the characterization of their thermal properties, mechanical properties, electrical properties, and antimicrobial properties were thoroughly examined and discussed. As a highlight of this work, the AgCB-PBS composites provide improved antimicrobial and electrical properties, while maintaining the mechanical stability in comparison with the pure PBS. The obtained AgCB-PBS composite materials with good antimicrobial and

antielestatic properties could be a good candidate for further use in the production of keyboards. Furthermore, these obtained PBS-based composites are also biodegradable, of which the disposal problems after the end of use can be manageable.

2. Experimental

2.1. Chemicals and Materials. 3,4-Dihydro-3,6-dimethyl-1,3,2H-benzoxazine, the reducing agent used for the synthesis of silver nanoparticles coated on CB, was synthesized by the method reported previously [35]. Poly(butylene succinate) (PBS) used in this work was an injection grade FZ71PD from Mitsubishi Co. Ltd. All chemicals and materials were used as received without prior characterization.

2.2. Preparation of Nanosilver-Coated Carbon Black (AgCB). For the preparation of nanosilver-coated carbon black (AgCB), 10 g of carbon black was weighed and then put in a beaker containing 10 cm^3 CH_2Cl_2 . The mixture was then stirred for 3 minutes. After that, the solution of the synthesized reducing agent in CH_2Cl_2 ($1\text{ g}/10\text{ cm}^3$) and 1 g of AgNO_3 were added to the reaction mixture. Then, the mixture was continuously stirred for a further 8 h to complete the coating of silver onto the carbon black. The obtained AgCB product was filtered out and then washed with CH_2Cl_2 for 2 times and additionally with acetone for 1 time to remove organic impurities. The product was dried at 75°C for 3 h. After that, the obtained powder was sieved to reduce the particle sizes to be less than $150\text{ }\mu\text{m}$. The success of nanosilver coating on the carbon black substrate was confirmed by field emission scanning electron microscopy (FESEM, obtained by a JSM-7600F, JEOL) and transmission electron microscopy (TEM, collected by a TEM-Hitachi, HT7700).

2.3. Preparation of Poly(butylene succinate)- (PBS-) Based Composite Materials. Composite materials of PBS and CB, herein called as CB-PBS, were prepared from their compound pellets. Prior to the reaction, PBS (grade 2003D) was dried in an oven at 40°C for 24 h. Next, a small amount of vegetable oil was added so that the additives CB or AgCB can stick in the PBS matrix quickly. The amount of additives (CB or AgCB) was added by varying the percentages of 3, 5, 10, and 15% with respect to the PBS amount.

Melt flow index (MFI) of PBS, CB-PBS, and AgCB-PBS composites was investigated to get the information about processing conditions. The initial temperature for the test was set to be 170°C , and the pressing force was set to be 2.16 kg. Dumbbell-shaped samples of PBS, CB-PBS, and AgCB-PBS composites were fabricated by an injection molding method using a Battenfeld machine (BA 250).

2.4. Characterization and Properties Test of PBS-Based Composite Materials. The phase formation of the composite materials was investigated by X-ray diffraction (XRD) using an X-PertPRO MPD diffractometer (Cu K_{α}

radiation, Nickel filter, scan step = 0.02°). Moreover, the crystallinity at the surface of composite materials was also characterized by using grazing incidence X-ray diffraction (GIXRD) at beamline BL1.1W (Multiple X-ray Techniques, MXT) at Synchrotron Light Research Institute (SLRI), Thailand (using synchrotron monochromatic X-ray radiation of energy 12 keV or wavelength of 1.0322 Å, two-dimensional image plate MAR345 detector, incidence angle of 0.5 degrees, and sample-to-detector distance of approximately 170 cm). The 2-dimensional diffraction patterns were further integrated in order to obtain the 1-dimensional XRD patterns using Fit2d software. The XRD pattern of the NIST standard lanthanum hexaboride (LaB₆) was collected in order to calibrate the sample-to-detector distance used in this experimental setup.

Thermal properties of the prepared composite materials were examined by thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) using a TGA/DSC 1 METTLER TOLEDO STARE SYSTEM with a heating rate of 10°C/min. Mechanical properties of the prepared composite polymers were further studied according to the standard ASTM D 638-03. Tensile strength was studied by a universal testing machine (UTM). According to the ASTM D 683-03 standard, the tensile force was created from 1000 kg weight with the moving speed of 5 mm/min. The force was continuously applied until the sample was broken so that the tensile strength can be measured in either kg/cm² or N/mm². Furthermore, % elongation at break can be calculated using the formula given below:

$$\% \text{ elongation at break} = \left[\frac{(\text{final length} - \text{initial length})}{\text{initial length}} \right] \times 100. \quad (1)$$

For the electrical conductivity test, the prepared composite materials were cut into square-shaped samples and then glued using silver paint. The resistance can be measured by applying the voltage of 1000 Volts for 30 mins. The resistivity of the materials can be calculated from the equation: $\rho = R \times A/L$, where ρ = resistivity, R = resistance, A = cross-sectional area, and L = length.

Antibacterial and antifungal tests of prepared samples were examined using the agar disk diffusion method [36]. Two types of media were used in this study: (1) sterile Mueller–Hinton agar (MHA) was used for bacteria cultivation and (2) sterile Sabouraud's dextrose agar (SDA) was used for yeast cultivation. The inoculum suspension was prepared by incubating a pure culture of the test microorganisms at 37°C for 18 h. Then, the incubated microbes were transferred to the liquid media to adjust the turbidity to be 0.5 McFarland turbidity standard. A sterile cotton swab was dipped into the prepared inoculum and was then swabbed over the entire surface of the agar. The microorganisms used as the samplings in this study were *Micrococcus luteus* (*M. luteus*) ATCC 9341, *Staphylococcus aureus* (*S. aureus*) ATCC 25923, *Bacillus subtilis* (*B. subtilis*) ATCC 6633, *Escherichia coli* (*E. coli*) ATCC 25922, *Candida albicans* (*C. albicans*) ATCC 10231, and *Pseudomonas aeruginosa* (*P. aeruginosa*) ATCC 27853. The fabricated PBS-based composite materials were cut in

square-shaped samples and then placed in the Petri dish. The Petri dish was incubated at 37°C for 18–24 h for bacteria but 72 h at 30°C for fungi. The clear zone was observed, and the size of the inhibition area was recorded.

3. Results and Discussion

The melt flow study of the pristine PBS, CB-PBS, and AgCB-PBS composite materials was carried out at 170°C using the pressing weight of 2.16 kg, following the ASTM D1238 standard method. The flow rate of pristine PBS was 32.142 g/10 min. When the CB filler was added for 10, 20, 30, and 40 phr (per hundred resin) of CB with respect to the content of PBS matrix, the flow rates were decreased to 19.158, 5.766, 2.412, and 0.768 g/10 min, respectively (Figure 1). This result suggested that the content of CB additive is limited to be not higher than 20 phr. Therefore, the variation of CB contents of 3, 5, 10, and 15 phr was used in this study, hereafter coined as 3CB-PBS, 5CB-PBS, 10CB-PBS, and 15CB-PBS, respectively. The photographic images of the fabricated products (illustrated in Figure 2) indicate the successful preparation of the homogeneous, translucent dumbbell-shaped plastics.

To get insight into the details of phase formation and crystallinity of the as-fabricated products, XRD patterns of the pristine PBS, as well as the xCB-PBS composite materials, are collected. The XRD patterns (Figure 3) reveal the formation of crystalline PBS phase in all the cases. In detail, the pristine PBS shows the characteristic diffraction peaks at 2θ of 19.5°, 21.9°, and 22.6° which are indexed corresponding to the planes (020), (021), and (110) of PBS, respectively [37]. No additional XRD peaks are observed in the xCB-PBS composite materials, indicating that the CB additive used herein is amorphous. Note that, the diffraction peaks of the PBS matrix still remain in the composites. This observation shows that the addition of the CB filler does not disrupt the crystalline structure of the PBS matrix. To identify the existence of CB filler, SEM images of the pristine PBS and the as-fabricated xCB-PBS composites are compared (Figure 4). The surfaces of the xCB-PBS composites are composed of many fine particles embedded within the rather smooth PBS matrix (as observed in the case of pristine PBS). This observation indicates the good distribution of CB particles within the whole PBS matrix. Note that, the elemental mapping analyzed by energy dispersive X-ray spectroscopy (EDS) cannot be used to identify the distribution of CB within the PBS matrix due to the similar elemental contribution, i.e., C in both species.

To further examine the enhanced characteristic features of the composites by means of the addition of CB filler, thermal properties of the pristine PBS and CB-PBS composites were studied (Figure 5). DSC curves were recorded from –50 to 200°C under a nitrogen atmosphere to study the temperature related to phase transition of polymers, while the TGA thermograms were recorded from 0 to 1000°C under an oxygen atmosphere to examine the thermal stability and the amount of residues after thermal treatment. According to DSC thermogram (Figure 5(a)), the pristine PBS shows the glass transition temperature (T_g) at –61°C and

FIGURE 1: Melting flow rate of pristine PBS and x CB-PBS composites (x = percentages of CB additive with respect to the amount of PBS matrix). Note that the solid line is only used as a guide to the eye.

FIGURE 2: Photographic images showing physical appearance of (a) pristine PBS, (b) 3CB-PBS, (c) 5CB-PBS, (d) 10CB-PBS, and (e) 15CB-PBS composite materials after proceeded by an injection molding method.

the endothermic melting peak (T_m) at 117°C . Interestingly, the addition of CB filler in the CB-PBS composite materials reduces the T_g to be -63 to -64°C and also reduce the T_m to be at 115°C . TGA thermograms in Figure 5(b) show that the decomposition of pristine PBS starts to occur at the temperature around 300°C corresponding to the breaking down of the PBS molecules. The presence of CB filler in x CB-PBS composites, especially in case of 15CB-PBS (the magenta plot (e) in Figure 5(b)) with the highest CB content herein, shifts the decomposition temperature to be higher than the one of the pristine PBS. The drastic change of weight loss is

observed at about 400°C , and the weight of the residue is completely stable at the temperature of 550°C . The further increasing of temperature up to 1000°C shows no additional change of the residual weight, ensuring the complete thermal decomposition of the PBS-base materials. There is no significant trend of the residual weights of pristine PBS, 3CB-PBS, 5CB-PBS, 10CB-PBS, and 15CB-PBS, which are 0.28, 0.12, 0.53, 0.21 and 0.44, respectively.

Mechanical properties of pristine PBS and x CB-PBS, namely, tensile strength, elongation at break, impact strength, and bending strength were examined. Tensile

FIGURE 3: XRD patterns using X-ray of Cu K_{α} radiation of the as-synthesized (a) pristine PBS, (b) 3CB-PBS, (c) 5CB-PBS, (d) 10CB-PBS, and (e) 15CB-PBS materials indicating the crystalline phase of PBS. The addition of CB shown herein as amorphous phase does not disrupt the structure of PBS matrix.

FIGURE 4: SEM images of (a) pristine PBS, (b) 3CB-PBS, (c) 5CB-PBS, (d) 10CB-PBS, and (e) 15CB-PBS. Scale bar shown here is equal to $10\ \mu\text{m}$.

strength was studied by following the ASTM D638 standard and using a crosshead speed of 50 mm/min. In detail, tensile strength of the pristine PBS is 41.47 ± 0.24 MPa, while ones of the 3CB-PBS, 5CB-PBS, 10CB-PBS, and 15CB-PBS are 36.83 ± 0.65 , 33.29 ± 0.53 , 35.98 ± 0.13 , and 34.77 ± 1.17 MPa, respectively (Figure 6(a)). In addition, elongation at break of the pristine PBS is $16.30 \pm 0.74\%$ while ones of the 3CB-PBS, 5CB-PBS, 10CB-PBS, and 15CB-PBS are $13.04 \pm 0.15\%$, $8.28 \pm 0.58\%$, $12.97 \pm 0.44\%$, and $8.12 \pm 0.77\%$, respectively (Figure 6(b)). Impact strength of the pristine PBS is 2.95 ± 0.46 kJ/m², and ones of the 3CB-PBS, 5CB-PBS,

10CB-PBS, and 15CB-PBS are 2.36 ± 0.60 , 2.08 ± 0.57 , 2.35 ± 0.20 , and 2.16 ± 0.30 kJ/m² (Figure 6(c)). In general, the addition of CB filler into PBS to form the x CB-PBS composites slightly decreases tensile strength, elongation at break, and impact strength in comparison with the pristine PBS as also observed in the literature [38]. However, there is no trend of the change with respect to the content of CB additives. Unlike the previously mentioned mechanical properties, the higher amount of CB additives leads to the lowering of bending strength from the pristine PBS (of 101.84 MPa) to the composites 3CB-PBS (of 101.69 MPa),

FIGURE 5: (a) DSC and (b) TGA thermograms of the pristine PBS and xCB-PBS composite materials. Note that the labels shown in the plots represent the materials as follows: (A) pristine PBS, (B) 3CB-PBS, (C) 5CB-PBS, (D) 10CB-PBS, and (E) 15CB-PBS.

5CB-PBS (of 92.39 MPa), 10CB-PBS (of 82.95 MPa), and 15CB-PBS (of 71.77 MPa). Even though the composites materials reveal a reduction of mechanical strengths from the pristine PBS, they are still within the acceptable range to be used in similar fashions as the PBS-based polymers.

For the further step, we additionally integrated the properties of silver nanoparticles into the PBS-based composites in order to enhance their characteristic features. Firstly, the additive so-called nanosilver-coated carbon black (AgCB) was prepared by the benzoxazine oxidation method [35]. SEM and TEM images of the synthesized products shown in Figure 7 indicate the successful incorporation of silver nanoparticles onto the carbon black substrate. As shown in the TEM image, the approximately 2 nm-sized Ag particles are embedded and well distributed within the spherical CB matrix. These obtained AgCB particles were further integrated into the PBS matrix with a variation of percentages of 3, 5, 10, and 15%. The phase formation of the as-fabricated xAgCB-PBS composite materials (x = percentages of AgCB with respect to the PBS content) was characterized by XRD. The XRD patterns of the AgCB-PBS composite materials (Figure 8) reveal the diffraction peaks of the PBS matrix as having been observed in the cases of CB-PBS composites. Moreover, there are some additional XRD peaks at 2θ of 38.41° , 43.34° , 66.37° , 77.15° , and 81.32° , which can be indexed as (111), (200), (311), and (222) of the metallic silver (JCPDS 04-0783), respectively. This evidence clearly shows the successful incorporation of AgCB additive into the PBS matrix and the remaining phase formation of all components within the composites. Moreover, SEM images of the as-fabricated xAgCB-PBS composites (Figure 9) also show that the composites consist of many fine AgCB particles embedded within the PBS matrix. The EDS elemental

mappings of C (Figure 9(f)), O (Figure 9(g)), and importantly Ag (Figure 9(h)) clearly identify the good distribution of AgCB additives within the PBS matrix.

AgCB-PBS composite materials were subjected to studying thermal properties (Figure 10). Here again, the DSC curves were recorded from -50 to 200°C under a nitrogen atmosphere to study the temperature related to phase transition of polymers. Interestingly, the incorporation of AgCB into the PBS matrix reduces the glass transition temperature to be at -63 to -64°C and the endothermic melting peak to be at 114°C , which is even lower than the case of CB-PBS composites (Figure 10(a)). TGA thermograms of AgCB-PBS composite materials decomposed under an oxygen atmosphere (Figure 10(b)) show the most considered weight loss at the temperature from 300 to 320°C . This is assigned to be the decomposition of PBS matrix phase. In the similar fashion with the DSC results, the decomposition temperature of the AgCB-PBS composites is slightly lower than the one of CB-PBS composites and also the pristine PBS since the silver nanoparticles coated on the CB surface can conduct heat very well. Therefore, it speeds up the thermal decomposition to occur at a lower temperature. Noted that, the residuals after thermal treatment are 1.90, 1.87, 1.07, and 0.99 wt.%, respectively, which are proved to be metallic silver [39, 40].

Moreover, the study on mechanical properties of AgCB-PBS was also performed in a similar way as the CB-PBS materials (Figure 11). Here again, the addition of AgCB filler into PBS slightly reduces tensile strength, elongation at break, impact strength, and bending strength of the composites in comparison with the pristine PBS in the similar fashion as the cases of CB additive. This observation clearly shows that the mechanical properties of the

FIGURE 6: (a) Tensile strength, (b) elongation at break, (c) impact strength, and (d) bending strength of the pristine PBS as well as the xCB-PBS composite materials.

FIGURE 7: (a) SEM and (b) TEM images of the nanosilver-coated carbon black (AgCB) used as the additive for fabrication of xAgCB-PBS composites (x = the percentages of AgCB filler with respect to the PBS content).

FIGURE 8: (a) Calculated XRD peaks of standard silver metal (JCPDS no. 04-0783) and the observed XRD patterns of (b) 3AgCB-PBS, (c) 5AgCB-PBS, (d) 10AgCB-PBS, and (e) 15AgCB-PBS composites.

FIGURE 9: Continued.

FIGURE 9: SEM images of (a) the pristine PBS in comparison with the (b) 3AgCB-PBS, (c) 5AgCB-PBS, (d) 10AgCB-PBS, and (e) 15AgCB-PBS composites. The corresponding EDS elemental mappings of (f) C element, (g) O element, and (h) Ag element of the 15AgCB-PBS composite illustrate the well distribution of the AgCB additive within the PBS matrix. Note that the scale bar depicted in Figures 9(a) to 9(d) is $10\ \mu\text{m}$, while the one depicted in Figures 9(e) to 9(h) is $20\ \mu\text{m}$.

FIGURE 10: (a) DSC and (b) TGA thermograms of the pristine PBS and the AgCB-PBS composite materials. Note that the labels shown in the plots represent the materials as follows: (A) PBS, (B) 3AgCB-PBS, (C) 5AgCB-PBS, (D) 10AgCB-PBS, and (E) 15AgCB-PBS.

FIGURE 11: (a) Tensile strength, (b) elongation at break, (c) impact strength, and (d) bending strength of the pristine PBS as well as the xAgCB-PBS composite materials.

composites are not straightforwardly related to the presence of Ag particles but rather the CB additive. In detail, tensile strengths for 3AgCB-PBS, 5AgCB-PBS, 10AgCB-PBS, and 15AgCB-PBS were 36.83 ± 0.65 , 33.29 ± 0.53 , 35.98 ± 0.13 , and 34.77 ± 1.17 MPa, respectively (Figure 11(a)). For the test of elongation at break, the 3CB-PBS, 5CB-PBS, 10CB-PBS, and 15CB-PBS composites reveal the value of $13.04 \pm 0.15\%$, $8.28 \pm 0.58\%$, $12.97 \pm 0.44\%$, and $8.12 \pm 0.77\%$, respectively. Moreover, impact strengths of 3CB-PBS, 5CB-PBS, 10CB-PBS, and 15CB-PBS are 2.36 ± 0.60 , 2.08 ± 0.57 , 2.35 ± 0.20 , and 2.16 ± 0.30 kJ/m². The bending strengths of 3CB-PBS, 5CB-PBS, 10CB-PBS, and 15CB-PBS gradually decrease from 101.69, 92.39, 82.95, to 71.77 MPa, respectively.

To prove the enhanced characteristic features of the composites by means of the presence of Ag particles within the composites, the electrical conductivity of the AgCB-PBS composite materials was measured in comparison with the pristine PBS. The results clearly show an improvement of the electrical conductivity of the AgCB-PBS composites for

approximately an order of magnitude. Specifically, the pristine PBS has a conductivity of $1.73 \times 10^{-7} (\Omega\text{m})^{-1}$, whereas the 3AgCB-PBS, 5AgCB-PBS, 10AgCB-PBS, and 15AgCB-PBS reveals the electrical conductivity of 2.28×10^{-6} , 3.62×10^{-7} , 9.40×10^{-7} , and $3.01 \times 10^{-6} (\Omega\text{m})^{-1}$, respectively. Note that, the electrical conductivities of AgCB-PBS composite materials observed herein are even higher for about an order of magnitude than those of AgCB-PLA composite materials prepared by using a similar procedure reported by our group previously [39]. This result highlights the possibility of using these AgCB-PBS composite materials for antielectrostatic applications, e.g., antifouling plastics.

Not only an enhancement of electrical conductivity, the antimicrobial activities of the AgCB-PBS composites were also studied by the agar disk diffusion method using different microorganisms, namely, *S. aureus* (ATCC 25923), *B. subtilis* (ATCC 6633), *M. luteus* (ATCC 9341), *E. coli* (ATCC 25922), *P. aeruginosa* (ATCC 2785), and *C. albicans* (ATCC 10231) (Figure 12 and Table 1). The bacteria being studied in

FIGURE 12: Positive tests for antimicrobial properties on *E. coli* and *C. albicans* using the agar disk diffusion method of (a) 3AgCB-PBS, (b) 5AgCB-PBS, (c) 10AgCB-PBS, and (d) 15AgCB-PBS composite materials.

TABLE 1: Antimicrobial properties of AgCB-PBS composite materials.

Nanosilver-coated material	Microbial strains (clear zone)					
	<i>E. coli</i> ATCC 25922	<i>S. aureus</i> ATCC 25923	<i>P. aeruginosa</i> ATCC 2785	<i>M. luteus</i> ATCC 9341	<i>B. subtilis</i> ATCC 6633	<i>C. albicans</i> ATCC 10231
3AgCB-PBS	–	–	–	–	–	–
5AgCB-PBS	–	–	–	–	–	–
10AgCB-PBS	–	–	–	–	–	+
15AgCB-PBS	+	–	–	–	–	+

this work have different types of cell walls. They can be classified into two groups, i.e., Gram-negative bacteria and Gram-positive bacteria. *E. coli* (ATCC 25922) and *P. aeruginosa* (ATCC 2785) are Gram-negative bacteria while *S. aureus* (ATCC 25923), *M. luteus* (ATCC 9341), and *B. subtilis* (ATCC 6633) are Gram-positive bacteria. The antimicrobial study (Figure 12 and Table 1) shows that the silver nanoparticles within the AgCB-PBS composites reveal a stronger activity against the Gram-negative bacteria than the Gram-positive bacteria, which agrees well with the previous literature [41, 42]. The difference in antimicrobial activity might be due to their different cell wall structures [43, 44]. The Gram-positive bacteria consist of the thicker peptidoglycan (20–80 nm) than Gram-negative bacteria (7–8 nm) [43]. Therefore, the thicker peptidoglycan sheet might prevent the penetration of silver nanoparticles from getting into the bacterial cells to damage them. The other type of microorganisms, namely, *C. albicans*, pathogenic yeast, was also investigated. It is observed that the fabricated AgCB-PBS composite materials illustrate greater inhibition zones than both the Gram-positive and Gram-negative bacteria. This result is similar to the study reported by Parthiban et al. [45]. Again the activity is mainly related to the cell wall structure of the microorganisms. Overall, our research work discloses the development of antimicrobial biocomposites. Among all the samples, 15AgCB-PBS shows the most effective inhibition zone against *E. coli* (ATCC 25922) and *C. albicans* (ATCC 10231), highlighting the role of Ag particles within the AgCB-PBS composites.

An additional phase analysis of the 1-year-aged CB-PBS and AgCB-PBS composite materials was performed using the synchrotron-based grazing incidence XRD (GIXRD). The GIXRD patterns (Figure 13) reveal that there is only the crystalline PBS polymer matrix phase presented at the

surface part of the materials in all the cases. This observation suggests that the AgCB-PBS composites gradually degraded due to the leaking of crystalline Ag nanoparticles as well as the reaction with the microbes especially at the surface of materials when the materials are placed in the ambient atmosphere for a longer time. This emphasizes the fact that even though the AgCB-PBS composites exhibit the antimicrobial properties, they are still biodegradable. Hence, these AgCB-PBS composites could be one of the promising candidates for environmentally friendly plastics for the future.

4. Conclusion

In this work, the pre-formed additive so-called nanosilver-coated carbon black (AgCB), which was prepared by the benzoxazine oxidation method to distribute silver nanoparticles onto the surface of carbon black, is used to blend with the PBS polymer matrix in order to enhance its characteristic features. Phase identification using XRD of the as-fabricated CB-PBS and AgCB-PBS composites fabricated by the injection molding mainly shows the XRD peaks according to the crystalline phase of the PBS matrix. The XRD peaks of metallic silver particles are additionally observed in the as-fabricated materials, indicating the successful preparation of the AgCB-PBS composite materials. Addition of the CB and AgCB fillers affects the glass transition temperature, the endothermic melting peak, and the thermal decomposition temperature of the PBS matrix. Mechanical properties in terms of tensile strength, elongation at break, impact strength, and bending strength are slightly decreased in the composites in comparison with the pristine PBS materials. However, they are still within the acceptable regions. As a highlight of this work, AgCB-PBS

FIGURE 13: One-dimensional integration XRD patterns of the 1-year-aged (a) xCB-PBS composites and (b) xAgCB-PBS composites in comparison with the pristine PBS material obtained from the two-dimensional grazing incidence X-ray diffraction (2d-GIXRD) patterns using synchrotron monochromatic X-ray of energy 12 keV (wavelength of 1.0332 Å) and grazing incidence angle of 0.5°.

composite materials, especially the 15AgCB-PBS, reveal an enhancement of the electrical conductivity for approximately an order of magnitude as well as exhibit the antimicrobial activities for inhibition against *S. aureus* and *C. albicans* using the agar disk diffusion test. The fabricated AgCB-PBS composite materials are expected to be one of the suitable candidate materials for producing smart keyboards as they have good antimicrobial and electrical properties, which could reduce the requirements for the cleaning process.

Data Availability

The data used to support the findings of this study are available upon request through Dr. Worawat Wattanathana via e-mail (fengwwa@ku.ac.th), or through Assoc. Prof. Dr. Apirat Laobuthee via e-mail (fengapl@ku.ac.th).

Conflicts of Interest

The authors declare there are no conflicts of interest regarding the publication of this paper.

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