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Preparation, Antibacterial and Antistatic Properties of PP/Ag-Ms/CB Composites

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Abstract: Polypropylene (PP) composites that contain silver micro-particles (MILLION KILLER, denoted as Ag-Ms) and conductive carbon black (CB) have both antibacterial and antistatic properties. In the present study, the antibacterial and antistatic PP/Ag-Ms/CB composites were prepared by melt blending. The results showed that when the content was 0.8 wt%, Ag-Ms could be uniformly dispersed in the PP matrix and the mechanical properties of the composites remained stable. And the reduction percentages of Staphylococcus aureus and Escherichia coli were more than 80% which showed the good antibacterial behavior. In addition, conductive carbon black had reinforcing and toughening effects on the mechanical properties of PP/Ag-Ms/CB composites. When the content of CB was beyond 30 wt%, the surface resistance of the composite was reduced to less than 10^8 Ω which showed a remarkable antistatic property. According to the different filling content of conductive carbon black, it can flexibly regulate the resistivity of PP, and the conductive effect is durable and stable. We thus can produce permanent antistatic materials.

Key words: antibacterial; antistatic; polypropylene; conductive carbon black; surface resistivity

1 Introduction

With superior mechanical properties, easy forming process and various forming methods, polypropylene (PP) has been widely used in automotive industry to produce automotive parts such as bumpers, dashboard, engine cooling fan, sun visor, automotive interior board *etc*[1-3]. Polypropylene occupies more than 30% proportion of all the automotive plastics[4]. And inside a vehicle, most of the parts directly contacting with human bodies are made of polypropylene or modified polypropylene. Disappointedly, bacteria will adhere on the surface of these PP materials which can lead to pathogenic biofilm formation and subsequent infectious complications[5-8]. Moreover, PP is generally low in moisture absorption and high in electric insulation[9-12]. Therefore, PP products easily accumulate electrostatic charges due to the inherent high insulativity[13,14], causing troubles such as electric shocks and even creating the danger of explosions[15].

However, the preparation of polymers with both antibacterial and antistatic properties as well as their applications in PP composites was seldom reported. In order to eliminate the bacterial and static, the way of blending antibacterial and antistatic agents into the PP matrix is popular under current circumstance[16-18].

Metal micro-particles can be incorporated into PP by using the melt-blending technique. Silver-based metals are non-toxic, inorganic additives and do not cause skin irritation[19-21]. Also, they are bactericidal agents because they combine with the cellular protein of a broad spectrum of harmful microorganisms and inactivate them[22-23]. High concentrations of metal salts coagulate cytoplasm proteins, resulting in damage of the cellular metabolism and the destruction of a microorganism. Such materials have high temperature stability, low volatility, and can thereby be used to prevent bacterial infections[24,25].

Commonly, the antistatic material has a surface resistivity of 10^4-10^{11}Ω[26]. Conductive carbon black and conductive graphite are often used as antistatic agents. Although conducting graphite has better conductivity, carbon black material particles are smaller and are more

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easily mixed with PP, which can be more effectively dispersed around conductive materials to form a permeable conductive network[27,28]. Such a system can also achieve high conductivity (up to 10^9 Ω), and the conductive effect are permanent. Therefore, although the conductivity of carbon black is worse than that of graphite, it is a good option for producing permanent antistatic materials.

The main purpose of this paper is to develop new PP-based composite for applications in the automotive industry as using silver micro-particles and carbon black to improve the antibacterial and antistatic properties. Objective of the paper is also to assess the influence of silver micro-particles and carbon black on the electronical, mechanical and antibacterial properties of the composite.

2 Experimental

2.1 Materials

PP (4712E1) was purchased from ExxonMobil Corp. (America). Carbon blacks (BC-80, the diameter 35 nm and the specific surface area $70m^2/g$) were from China Petroleum and Chemical Corp. (Shanghai, China). Antimicrobial agent (MILLION KILLER, denoted as Ag-Ms) was obtained from Shanghai Wako Chemical Co., LTD (Shanghai, China). Molten maleic anhydride grafted EPDM (EPDM-g-MAH) was supplied by Nanjing Gaobiao Rubber & Plastic Co., LTD (Nanjing, China).

2.2 Composite preparation

Prior to plasticating, carbon black, PP pellets, EPDM-g-MAH and antistatic agent were dried in an oven for 3 hours at 80 ℃. Here, EPDM-g-MAH was used as compatibilizer. The plasticating processes of PP/Ag-Ms blends and PP/CB/Ag-Ms composites were carried out using a co-rotating twin screw plasticator (model BL-6175-B, Jiangsu, China) with a L/D 40 and intermeshing screw configuration. The screw speed was set at 150 rpm. The processing temperature was set in the range of 165-180 ℃ and the processing time was set in the range of 30-40 min. Further, the PP/CB/Ag-Ms composites were cooled and pulverized in a particle size of 5 mm diameter. Finally, the pellets were pumped to an injection molding machine to make mechanical-shaped samples.

2.3 Characterization

2.3.1 Tensile and bending tests

Specimens were prepared by injection molding method. Tensile and bending test was carried out with an Instron universal testing machine (model CMT5205, China) at room temperature (25 ℃, 50% relative humidity), according to GB/T 1040-2008 and GB/T 9341-2008, at a speed of 8 and 2.5 mm/min, respectively.

2.3.2 Charpy impact tests

The Charpy impact tests were performed according to GB/T 1043-2008 by using an impact testing machine (model ZBC50, Shenzhen, China) with appropriate software for processing the data. The tests were carried out for single-notched specimens at room temperature. Rectangular samples, with width of 3 mm, thickness of about 4 mm and length of 5 cm were used. The samples were cut from slabs obtained by injection molding machine, same as that in section 1.3.1. The Charpy impact strength (a_c) was calculated using Eq.(1):

$$a_c = \frac{W}{h \cdot b} \quad (1)$$

where W is the corrected energy absorbed by breaking the test specimen (J), h is the thickness of the test specimen (mm), b is the remaining width at the notch base of the test specimen (mm).

2.3.3 Surface resistance and volume resistance tests

Surface resistance (Rs) and volume resistance (Rv) were tested, according to GB/T1410-2006, by the megger (Shanghai Instrument (Group) Company, Z630) at room temperature. Antistatic persistence was tested in the following way. The samples of PP/CB/Ag-Ms composites were immersed into distilled water, and the sample surface was scrubbed with absorbent cotton. The whole process was repeated 50 times, then, the samples were exposed to air for drying under the conditions of room temperature for 24 h. Finally, the Rs and Rv values of samples were measured. The thickness of specimen, test voltage and test time was 1 mm, 250 V and 1 min, respectively. The surface resistivity of PP/CB/Ag-Ms composites was determined by using Eq.(2) and the volume resistivity was determined by using Eq.(3):

$$\rho_s = \frac{2\pi}{\ln \frac{d_2}{d_1}} \times R_s \quad (2)$$

$$\rho_V = \frac{\pi r^2}{h} \times R_V \quad (3)$$

where ρ_s is the surface resistivity (Ω), ρ_v is the volume resistivity (Ω•m), R_s is the surface resistance (Ω), R_v is the volume resistance (Ω), π is the ratio of the circumference of a circle, d_2 is the inside diameter of guard electrode (m), d_l is the diameter of main electrode (m), r is the radius of main electrode (m), h is the thickness of specimen (m).

2.3.4 Antibacterial tests

The evaluation was performed using the Standard Test Method of QB/T 2591-2003. Staphylococcus aureus and Escherichia coli were used to assess the bacterial attachment on the modified PP surface. First, the preparation of the bacterial inoculum required to grow in a sterile nutrient broth (NA composition for 1 L: 10 g of peptone, 5 g of yeast extract, 5 g of sodium chloride and 15 g of agar) was conducted at 37 ℃ for 24 h. Then a single colony of bacterium from the agar plate was used to inoculate 50 mL of NA, and cultured to exponential growth phase at 37 ℃ for 24 h. The bacteria growth broth was centrifuged at 8 000 rpm for 2 min and washed twice with phosphate-buffered saline (PBS; 0.01 M KH_2PO_4-Na_2HPO_4 containing 0.8%NaCl, pH = 7.15) to remove the supernatant. Bacterial cells were diluted with NA to the desired concentration (5.0×10^5–10.0×10^5CFUs/mL), which was calculated by testing the absorbance of the cell dispersions.

The ability of the samples to kill adhering bacteria was evaluated by the spread plate method. First, these test samples were put into the 24-well plates containing 600 μL of bacterial suspension (approximately 1.5×10^6 cell mL^{-1}), and kept for 24 h at 37 ℃. No nutrients were added in the suspension in order to preserve the existing bacteria number constant. These suspensions were used in determining antibacterial activity and bacterial adhesion. Then, these samples were washed with PBS solution 3 times to remove the non-adherent bacteria. Finally, these samples were dehydrated with a 70 vol% ethanol/water mixture for 30 min every time and finally evaporated to constant weight at room temperature under vacuum. After cultured for 4, 8, 12, 16 and 20 h, respectively, the number of viable bacterial cells was determined by colony count.

2.3.5 Scanning electron microscopy

The fracture surface of the composites was investigated using SEM, JSM-6510LV (JEOL, Japan) at an accelerator voltage of 10 kV. The fracture surface of the specimens was sputter-coated with a thin gold-palladium layer in a vacuum chamber for conductivity before examination.

3 Results and discussion

3.1 Mechanical Properties

Table 1 lists the mechanical properties of PP/Ag-Ms/CB composites with different weight content of Ag-Ms. As can be seen in Table 1, with the increase of Ag-Ms, the mechanical properties of PP/Ag-Ms/CB composite remain approximately stable. It is indicated that a small amount of Ag-Ms have little effect on the mechanical properties of the composites. But Ag-Ms have great effect on the electrical and antibacterial properties of PP/Ag-Ms/CB composite, which we will discuss later in this paper.

Table 1 Mechanical properties of PP/Ag-Ms/CB composites with different weight content of Ag-Ms (content of CB is 30 wt%)

Item	Weight content of Ag-Ms/%				
	0.2	0.5	0.8	1.0	1.5
Tensile strength/MPa	37.8	37.6	37.6	37.5	37.4
Flexural strength/MPa	37.5	37.5	37.3	37.2	37.1
Charpy impact strength /kJ·m^2	8.9	8.9	8.7	8.6	8.5

In the composite, there exists another critical component-conductive carbon black, which is a kind of conductive inorganic filler added and uniformly dispersed in PP with a co-rotating twin screw plasticator by melt mixing[29-31]. But the CB particles prefer to aggregate and cannot reach a molecular dispersion level in PP[32]. Defects with different degrees may be formed on the interface between CB particles and PP resin, which generate stress concentration to affect the mechanical properties of the composites[10,14].

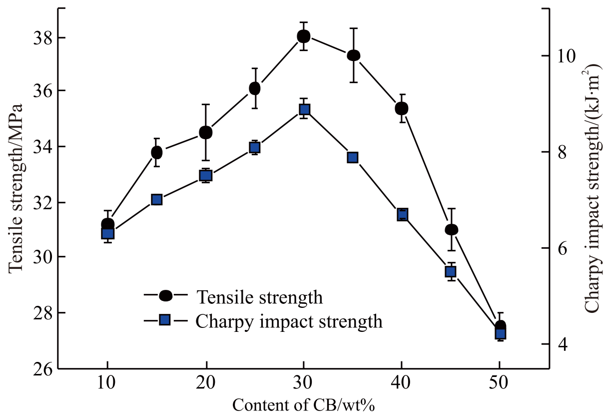

Fig.1 Relationships between the tensile strength charpy impact strength and CB content (Ag-Ms 0.8 wt%)

Fig.1 shows the effect of CB particles on the tensile strength and charpy impact strength of PP/Ag-Ms/CB composites. with the increase of the content of conductive carbon black, the tensile strength and impact strength of PP / CB composites first increase and then decrease. When the content of CB is 30 wt%, the tensile strength and the charpy impact strength reach their maximum values, 38 MPa and 8.9 kJ•m^2, and are improved by 31.0% and 37.3% compared with that of pure PP, 29 MPa and 6.5 kJ•m^2, respectively.

The increase of the tensile and impact properties of the above composites results from the reinforcing and toughening effect of CB[18]. On the one hand, the rigid CB particles are effectively dispersed in PP matrix and the stress concentration area of PP is expanded, and the PP resin around CB particles is induced to yield micro-crack to absorb a certain degree of deformation energy; On the other hand, the rigid CB particles prevent the micro-crack from further expanding to destructive crack[33-35]. When the content of CB is less than 30 wt%, CB particles can be uniformly dispersed in PP resin. Due to the small size effect and surface structural effect, physical and chemical absorption occurs between CB particles and PP resin[36-38]. When the composites are subjected to the action of external forces, CB particles and PP are physically tangled together to expand the stress concentration area to produce micro-crack, and the molecular chain of PP is inhibited to freely slide by the energy absorption of CB particles[39,40].

However, when the content of CB exceeds 30 wt%, the dispersion of CB particles in PP matrix turns to be difficult and a certain degree of agglomeration of CB particles occurs. After that, defects in PP matrix are increased gradually by the agglomerating of CB particles. When the composites are subjected to external forces, destructive cracks are produced in PP matrix and the tensile strength and charpy impact strength of the composites are decreased.

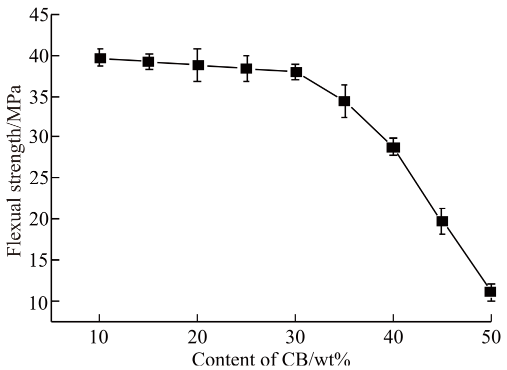

Fig.2 Relationship between the flexural strength and CB content (Ag-Ms 0.8 wt%)

Fig.2 depicts the relationship between the weight content of CB particles and the flexural strength of PP/Ag-Ms/CB composite. With the increase of CB, the flexural strength of PP/Ag-Ms/CB composite is decreased. There might exist two reasons to explain this phenomenon. Firstly, the added CB particles reduce the crystallinity of PP to decrease the rigidity of the substrate[35]. Secondly, the strength of CB particles is lower than that of PP matrix[38], and the flexural strength of PP/Ag-Ms/CB composite decreases with the increase of CB particles.

3.2 Antistatic properties

Surface resistance and volume resistance are commonly used to assess the antistatic properties of polymeric composite[9-10]. Table 2 lists the values of the surface resistance and volume resistance of pure PP and PP/Ag-Ms/CB composite with 0.8 wt% Ag-Ms.

Table 2 Antistatic properties of samples (Content of CB is 30 wt%)

Antistatic property	Pure PP	PP/Ag-Ms/CB composite with 0.8 wt% Ag-Ms
Surface resistance R_s /Ω	6.3×10^{14}	7.8×10^{15}
Surface resistivity ρ_s/Ω	4.7×10^{16}	4.2×10^{17}
Volume resistance R_v /Ω	7.5×10^{17}	8.3×10^{18}
Volume resistivity ρ_v /Ω·m	1.2×10^{17}	3.4×10^{18}

From Table 2, it is plausible to conclude that antibacterial agent (Ag-Ms) has an influence on the antistatic properties of PP/Ag-Ms/CB composite. With the addition of 0.8 wt%Ag-Ms, the surface resistance and volume resistance of PP are improved by an order of magnitude. It is represented that the leakage current of PP decreases, and the conductive properties of the material turn to be worse. Thus, the addition of 0.8 wt%Ag-Ms is negative to improve the antistatic properties of PP/Ag-Ms/CB composite.

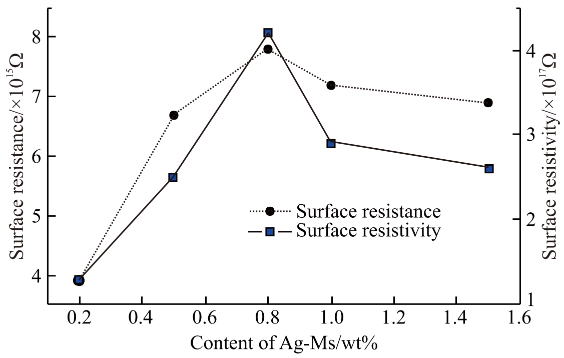

Fig.3 Effect of weight content of Ag-Ms on the surface resistance and surface resistivity of PP/Ag-Ms/CB composite (CB 30 wt%)

Fig.3 displays the effect of weight content of Ag-Ms on the surface resistance and surface resistivity of PP/Ag-Ms/CB composite. Fig.4 presents the effect of weight content of Ag-Ms on the volume resistance and volume resistivity of PP/Ag-Ms/CB composite.

As can be seen from Fig.3 and Fig.4, with the increase of Ag-Ms, the surface resistance and surface resistivity as the same as the volume resistance and volume resistivity of PP/Ag-Ms/CB composite first increase and then decrease. When the content of Ag-Ms is less than 0.8 wt%, the antistatic property of the composite performs worse. It is indicated that a small amount of Ag-Ms hinders the formation of conductive channels in PP matrix, resulting in the decrease of the antistatic properties of the composites. However, when the content of Ag-Ms exceeds to 0.8 wt%, the Ag^+ released from Ag-Ms plays a major role of conductivity. The antistatic properties of the composites are gradually improved.

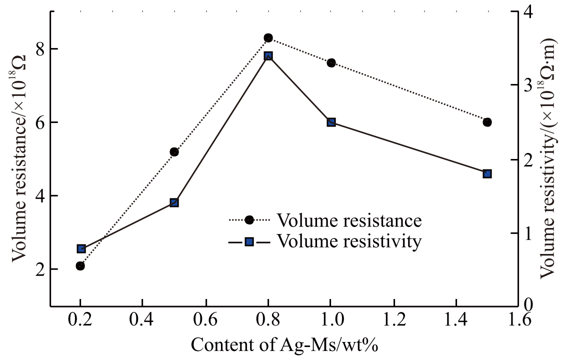

Fig.4 Effect of weight content of Ag-Ms on the volume resistance and volume resistivity of PP/Ag-Ms/CB composite (CB 30 wt%)

It is known that the contacting number and the approaching degree of conductive particles are two key factors[14,30] which influence the conductivity of Ag^+. In this work, the sizes of Ag-Ms particles are very small. They can be uniformly dispersed in PP matrix by two-roll mill. Therefore, though the content of Ag-Ms particles adding into PP matrix is very small, the contacting number and the approaching degree of Ag^+ for conductivity are easily achieved. The composite exhibits good electrical conductivity.

The addition of Ag-Ms particles can improve the antistatic properties of PP/Ag-Ms/CB composites, but the improving effect is not obvious and the cost of Ag-Ms is expensive[24]. So, in our work, the added conductive carbon black is the key filler to improve the antistatic properties of PP/Ag-Ms/CB composites. Fig.5 shows the effect of the weight content of CB particles on the surface resistance and surface resistivity of PP/Ag-Ms/CB composites and Fig.6 shows the effect of the weight content of CB particles on the volume resistance and volume resistivity of PP/Ag-Ms/CB composites.

Generally, when the surface resistance of a material is less than 10^9 Ω, this material is considered to be antistatic[1,3]. As shown in Fig.5 and Fig.6, with the increase of conductive carbon black, the surface resistance and surface resistivity as well as the volume resistance and volume resistivity of PP/Ag-Ms/CB composite are decreased. When the content of CB is 30 wt%, the surface resistance of PP/Ag-Ms/CB composite is 4.1×10^8Ω, and the composite performs antistatically. It is because of the good conductive property of conductive carbon black. The mixing of conductive carbon black and PP is a physical filling process[36,38], which does not change the natural conductivity of PP. When the conductive carbon black particles in PP are connected or close to a certain extent, conductive tunnels can be formed in the composites to perform antistatically.

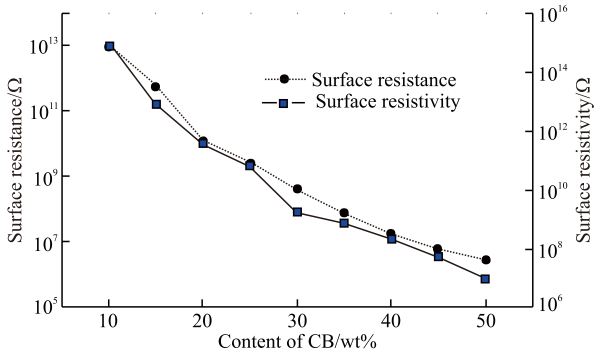

Fig.5 Effect of weight content of CB on the surface resistance and surface resistivity of PP/Ag-Ms/CB composite (Ag-Ms 0.8 wt%)

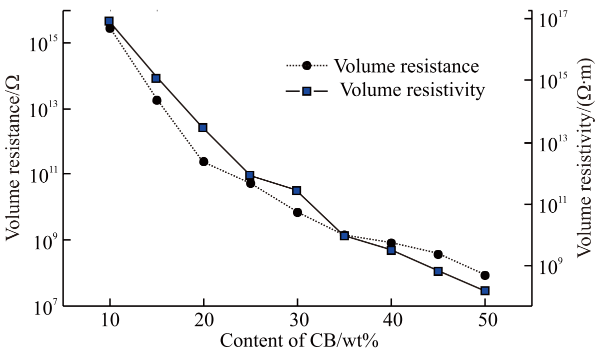

Fig.6 Effect of weight content of CB on the volume resistance and volume resistivity of PP/Ag-Ms/CB composite (Ag-Ms 0.8 wt%)

It is well known that the carbon atom of CB exists in the form of graphite structure and free electrons can move freely between the layers of graphite structure[33]. Thus, the carbon black is shown to be conductive.

When the content of conductive carbon black is less than 30 wt%, the conductive particles are separated from each other, and there is no effective conductive tunnel in the composite. At this time the conductivity of the composite depends mainly on the conductivity of the PP matrix[14], so the antistatic property can not be obtained. But when the content of the conductive carbon black exceeds 30 wt%, with the increase of CB, the distance between the CB particles is reduced, the particles contact each other to form a certain conductive tunnel, the conduction path of the conductive particles is much more, the conductive network of the composite is further improved, the conductivity of the composite is better, and the antistatic performance is better.

3.3 Antibacterial properties

Table 3 shows the antibacterial rate of Staphylococcus aureus and Escherichia coli at different culturing time of 4, 8, 12, 16 and 20 h. From Table 3, the antibacterial rates Staphylococcus aureus and Escherichia coli are above 80% which represents that PP/Ag-Ms/CB composite has a good antibacterial property. This may be due to the special chemical structure of Ag-Ms. The chemical structure of silver determines that it has a strong oxidizing property [Ag-e→Ag^+] and Ag^+ can be easily released in an aqueous environment. Ag^+ has a bad effect on the vitality of microorganisms. When Ag^+ enters the surface layer of the microbial cell membrane, the microbial cell membranes are firmly absorbed by Ag^+ with negative charges by Coulomb force. When Ag^+ penetrates the cell wall and enters the cell, rapid solidification of protein occurs and the activity of synthetase in cells is damaged. Then, the cells gradually lose the ability to proliferate and die[41-45]. In addition, the reduction potential of Ag^+ is higher and reactive oxygen surrounding Ag^+ is easily produced for sterilization. After the bacteria are killed by Ag^+, Ag^+ is separated freely from the bacteria's body and comes into contact with the other bacteria and kills them. The entire process is repeated again and again[46-49].

Discussed from the above, it is known that Ag-Ms have a good antibacterial property. Moreover, the bacteriostatic efficacy of Ag-Ms is needed to be investigated, too. Table 4 lists some values about the relationship between the antibacterial rate of Staphylococcus aureus and Escherichia coli and the weight content of Ag-Ms.

Table 3 Antibacterial activity of PP/Ag-Ms/CB composite (content of CB is 30 wt%)

Reduction of bacteria	Culturing time/h				
	4	8	12	16	20
Staphylococcus aureus/%	83	94	96	97	99
Escherichia coli/%	85	95	97	97	98

Table 4 Antibacterial efficacy of PP/Ag-Ms/CB composite (content of CB was 30 wt%)

Reduction of bacteria	Content of Ag-Ms/wt%				
	0.2	0.5	0.8	1.0	1.5
Staphylococcus aureus/%	33.2	54.7	83.5	85.2	86.9
Escherichia coli/%	35.6	56.4	85.6	86.8	88.1

It is clear from Table 4 that the percentages of bacterial reduction of Staphylococcus aureus and Escherichia coli increase with an increase in Ag-Ms content. A change of Ag-Ms content in PP/Ag-Ms/CB composite from 0.2 wt% to 1.5 wt% increases the percentage reduction of Staphylococcus aureus from 33.2% to 86.9%. Furthermore, an increase of Ag-Ms content in PP/Ag-Ms/CB composite from 0.2% to 1.5% improves the percentage reduction of Escherichia coli from 35.6% to 88.1%. When the contents of Ag-Ms are 0.2 wt% and 0.5 wt%, the antibacterial rates of Staphylococcus aureus and Escherichia coli are less than 60%. But, when the increase of Ag-Ms exceeds 0.8 wt%, the antibacterial rates of Staphylococcus aureus and Escherichia coli are more than 80%, the composite

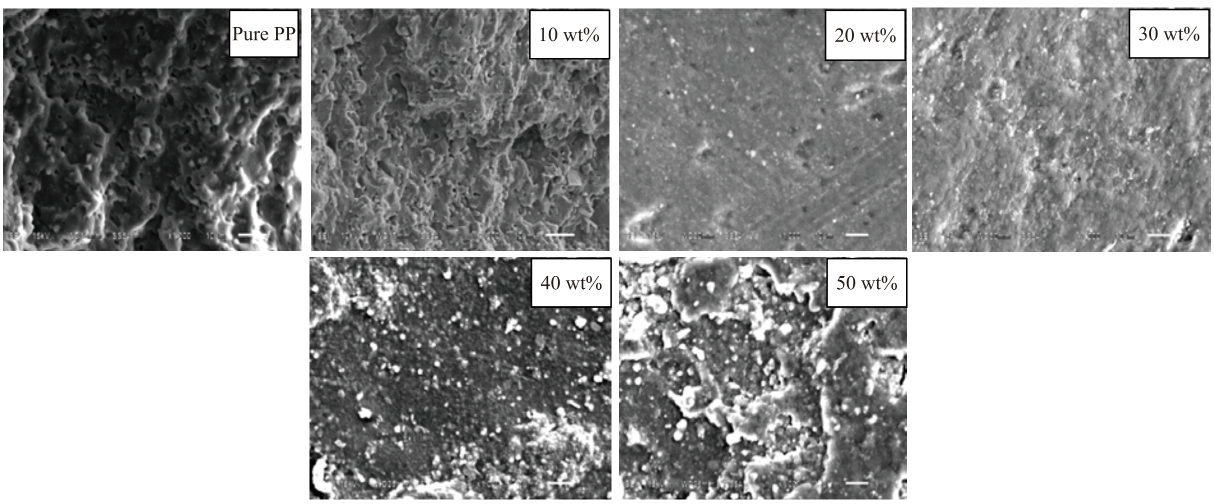

Fig.7 SEM micrographs of PP/Ag-Ms/CB composites(Content of Ag-Ms was 0.8 wt%, 15 kV, ×1 000)

exhibits excellent antibacterial property. This is also because of the Ag^+ bactericidal effect mentioned above, when Ag^+ penetrates the cell wall into the cell, the protein is rapidly solidified, the activity of the synthetize is damaged, the cells gradually lose the ability to proliferate and die[41-45], and the content of Ag^+ increases with the increase of the content of Ag-MS, and the bactericidal effect is better.

3.4 Fracture surface observation

In order to observe the distribution of CB in PP/Ag-Ms/CB composite, the fracture surfaces of samples with different content of 0,10 wt%CB, 20 wt%CB, 30 wt%CB, 40 wt%CB and 50 wt%CB are scanned by SEM. Results are shown in Fig.7.

It can be seen from Fig.7 that when the content of CB is less than 20 wt%, CB can be uniformly dispersed in the PP matrix and the composites show compatible and homogeneous phases. When the contents of CB are increased to 20 wt% and 30 wt%, CB particles are distributed evenly and densely and the conductive tunnels are formed[50,51]. The composite is transferred from an insulator to a conductor. However, when the content of CB is increased to more than 30 wt%, the dispersion of CB in PP matrix becomes worse. CB particles occur to agglomerate which increases the defects formed in the composite. Thus, the mechanical properties of the PP/Ag-Ms/CB composite become poorer and the antistatic property of the PP/Ag-Ms/CB composite is also poorer.

4 Conclusions

Antibacterial efficacy of PP/Ag-Ms/CB composite increases with an increasing Ag-Ms content. It can be concluded that Ag-Ms content in the composite has to be more than 0.8 wt% to see significant Staphylococcus aureus and Escherichia coli reduction. But, small content of Ag-Ms has little effect on the mechanical properties of the composites.

Carbon black has a great effect on the mechanical properties of the PP/Ag-Ms/CB composites. With the increase of CB, the tensile strength and impact strength of the composites first increase and then decrease. When the content of CB is 30 wt%, the tensile strength and the charpy impact strength reach their maximum values, 38 MPa and 8.9 kJ•m^2, and are improved by 31.0% and 37.3% compared with that of pure PP, 29 MPa and 6.5 kJ•m^2, respectively.

Moreover, carbon black has a great effect on the antistatic properties of the PP/Ag-Ms/CB composites, too. When the content of CB is 30 wt%, the surface resistance of PP/Ag-Ms/CB composite is $4.1\times10^8\Omega$, and the composite performs antistatically. Therefore, the resistivity of PP can be adjusted flexibly according to the different filling amount of conductive carbon black, and the conductive effect is durable and stable. It can be used to produce permanent antistatic material.

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