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Morphology and electrical conductivity of injection-molded polypropylene/carbon black composites with addition of high-density polyethylene

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Abstract

This work attempts to clarify the influence of carbon black (CB) addition on the microstructure of injection-molded high-density polyethylene (HDPE)/polypropylene (PP) blends and effect of shear-induced polymer deformation on the conductive network structure. We observed that HDPE molecules are strongly interacted with carbon surfaces and CB particles are selectively located in HDPE domains. Morphology of the injection-molded specimen consists of three parts, namely, CB–HDPE complex domain, free HDPE domain and PP domain. The volume and microstructure of the free HDPE domain are significantly influenced by HDPE and CB concentration, CB structure, and PP viscosity. We also confirmed that the CB particles are capable of self-assembly to form random conductive networks even under high shear rate within very short time. The morphological changes were finally correlated to the variation of electrical conductivity.

Keywords: Carbon black; Injection molding; Conductive polymer composite

1. Introduction

Carbon black (CB)-filled polymer composites show electrical conductivities ranging from insulators to semiconductors. Percolation takes place at a critical CB loading, called percolation concentration, where the first three-dimensional continuous CB network is built throughout the polymer matrix. The percolation concentration of such mixtures depends on the CB structure (particle size, aggregate shape and structure, porosity, and surface chemistry), on the polymer characteristics (chemical structure and crystallinity) and on processing methods and processing conditions. Generally, higher conductivities of the polymer composites can be obtained by using CBs of smaller particle size (larger surface area), lower particle density (higher particle porosity), higher structure (better aggregation), and low volatility (fewer chemisorbed oxygen groups) [1,2].

Concerning how to reduce the percolation concentration and how to control the desired conductivity of the CB-filled polymer composites, most studies focus on optimizing the degree of mixing and morphology of the matrix apart from CB structure and polymer characteristics [3,4]. Recently, we noticed [5] that the dispersed carbon particles are capable of self-agglomeration to form conductive networks and percolation is delayed by the bulk mobility of polymer layers surrounding the CB particles. Real-time trace of the dynamic percolation can be performed by measuring the time dependence of electrical resistivity during isothermal treatment of a CB/polymer compound at a melt temperature. There exists a quantitative relation among percolation time, annealing temperature, CB concentration, terminal relaxation time of the polymer matrix as well as CB–polymer interfacial energy. These findings are believed helpful to determine the best processing conditions for manufacturing the final products with high repeatability and high stability of conductivity. It is worth

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pointing out that studies of the dynamic percolation phenomenon are also of academic interests. As we know, so far, most of the theories have examined the percolation phenomenon at a state of thermodynamic equilibrium. The dynamic percolation measurement provides an effective approach to determine how percolation at an equilibrium state is correlated to percolation under experimental conditions. Moreover, since the mobility of the carbon particles due to CB agglomeration should incisively reflect the mobility of the polymer layer surrounding the carbon surface, the dynamic percolation measurements are potential for in situ study on polymer dynamics [6], particle–polymer interactions [5] and even phase fluctuations of miscible polymer blends [7] in highly filled polymer systems using the CB-like nanoparticle as a self-diagnosing probe.

A significant progress has been made in the past decade for further reduction in the percolation concentration by means of mixing CB with immiscible polymer blends [8–14]. The CB particles are found to be selectively located in one of two polymer matrices or at their interfaces. Double percolation [9] is the most adequate for describing the conductivity of the heterogeneous distributed system, where both the conductive network of CB in filler-rich phase and the continuity of this phase in the composites are basic requirements for maintaining a conductive network through the composites. Much effort has been devoted toward controlling the location of CB. It has been suggested [8,15] that the difference of the interfacial energy between CB and two polymers could be as a thermodynamic criterion to predict the selective distribution of CB in a polymer blend. The problem is how to precisely evaluate the CB–polymer interactions [16]. Our recent study revealed [17] that the surface tension of CB usually determined by small molecules is no longer valid for describing the CB–polymer interactions since CB has a very rough surface in nano-scale dimension and entropy penalty may play a main role in competitive adsorption of polymers. We found that for carbon-backbone polymer pairs, CB particles are always selectively located in a polymer having a lower glass transition temperature, T_g. That is to say, the flexibility of the polymer chains may play a dominant role in the selective location of CB in the polymer blends.

In this study, we will move attentions to a more complex and practically important system: injection molding of a CB-filled polymer blend. It is complex because during injection molding, the polymer composite must experience an inhomogeneous shear flow within very short time. Not only the morphology of the blended polymer matrices, but also the distribution of CB particles should be significantly deformed by the high pressure-driven molding flows, especially in the area near the surface of the molded specimens [3,18,19]. Generally, CB particles migrate from the region of high shear stress to the region of low shear stress. As a result, the zones near the surfaces are depleted of CB particles as migrated particles accumulate in core zones. These uneven could inevitably affect the nature of conductivity.

The objective of this study is to clarify the influence of CB particles addition on the microstructure of the injection molded polymer blends near the injection surfaces on one hand and, on the other hand, the influence of the polymer phase deformation on the conductive network structure. High-density polyethylene (HDPE)/polypropylene (PP) blends were selected as polymer matrix because the HDPE/PP blend was reported to become a miscible polymer blend during the injection molding [20] and this might be helpful for better distinguishing the relation between the CB networks and the fine structure of the spinodal decomposed matrix.

2. Experimental

2.1. Raw materials and sample preparation

Both PP and HDPE in this study were commercial polymers from Mitsubishi Chemical Co. They were dry-blended together with CB in a Henschel mixer for 1 min and melt compounded using a twin-screw extruder (PCM30 with a diameter of 30 mm, Ikegai Machinery) at a setting temperature of 240 °C with a screw speed of 200 rpm. The extruded melt was quenched in water and then chopped into pellets. Some physical characteristics of polymer and CB used in this study are given in Tables 1 and 2, respectively.

The dry pellets of above compositions were subjected to injection molding using an injection molder (Arburg) at a cylinder temperature of 240 °C and an injection pressure of 400 kg/cm^2 to prepare $100\times100\times2$ mm^3-thick sheets. The injection molding time is 10 s. For comparison, some of the specimens were hot-pressed at 200 °C for 10 min. These specimens were placed in vacuum desiccators immediately after molding for further testing.

2.2. Electrical resistivity measurements

The electrical resistivity was measured by a multi-meter (TR6845, Advantest) in the perpendicular direction of the molded sheets. Silver paste was used to ensure good contact of the sample surface with the copper electrodes. Due to no observable deviation from the four-terminal measurements, DC resistivity in this work were measured using two-terminal technique. The measurements were carried out at room temperature after a wait of 2 min in order to obtain a stationary value.

AC resistivity was determined using an impedance analyzer (4192A model, Hewlett Packard) with frequencies from 0.01 kHz to 10 MHz at room temperature.

Table 1
Characteristics of polymer used

Reference	Grade name	Density (g/cm^3)	Melt flow rate (g/10 min)	Viscosity (poise)[a]
PP	TA3	0.90	9.0	2.0×10^4
PP1	TA8	0.90	0.5	9.0×10^4
HDPE	JX20	0.96	5.0	—

[a] Measured at a shear rate of 100 s^{-1}.

Table 2
Characteristics of carbon black used

Reference	Grade name	Supplier	Specific surface area (m^2/g)	DBP adsorption number (ml/100 g)	Primary particle diameter (nm)
CB1	Furnace black #3050	Mitsubishi chemical	50	180	40
CB2	Furnace black #3250	Mitsubishi chemical	240	180	28
AB	Acetylene black	Denki kagaku	65	250	40
KB	Ketjenblack EC300	Ketjenblack int'l	800	360	30

2.3. Electron microscopy observation

Samples for TEM analysis were taken from the central portion of the specimens. They were stained by RuO_4 vapor at 40 °C for 1 h and then cut into an ultra-thin section (ca. 80–100 nm in thickness) using an ultra-microtome (Ultracut N, Reicherr). The section was cut parallel to the sheet surface at a depth of ca. 1.0 μm from the surface. TEM observation was carried out under a Jeol transmission electron microscope (JEM100CX, Nihon Denshi) at an accelerating voltage of 100 kV.

The CB dispersion and CB network structure were observed by means of a field emission type scanning electron microscope (FE S800, HITACHI). Polymers with a depth of ca. 1.0 μm from the sheet surface were erased by ion etching before the observation.

3. Results and discussion

3.1. Morphology of press-molded HDPE/PP/CB and injection-molded HDPE/PP

For better understanding the influence of injection molding on morphology of HDPE/PP/CB composites, we investigated the micro-structure of press-molded HDPE/PP/CB without experience of shear history and injection-molded HDPE/PP blends without incorporation of CB particles.

Fig. 1(a) and (b) shows typical SEM micrographs for press-molded HDPE/PP/Ketjen black (KB) with a volume ratio of 30/68/2 and 49/49/2, respectively. From the change in ratios of the HDPE/PP domain area in Fig. 1(a) and (b), one can distinguish that the KB particles are selectively located in HDPE domains. Notice that the HDPE domain size is bigger than 2.0 μm.

TEM micrographs of HDPE/PP blends with a volume ratio of 30/70 are shown in Fig. 2. One can observe a stripe pattern with periodicity of about 0.15 μm. The dark region is assigned to the HDPE-rich region, in which lamellar crystals (ca. 10 nm thick) characteristic to PE are visualized at a higher magnification, as shown in Fig. 2(b). In the bright region, one can see the cross-hatched lamellar structure, which is characteristic to the PP crystal (Fig. 2(b)). In other words, the PE- and PP-rich regions are oriented in flow direction and arranged alternatively and regularly. Further results [20] have been shown that such regularity could not be obtained by a simple mixing of immiscible systems. It may be attained only by the spinodal decomposition from a single-phase mixture prepared under a high shear rate in the injection molder. That

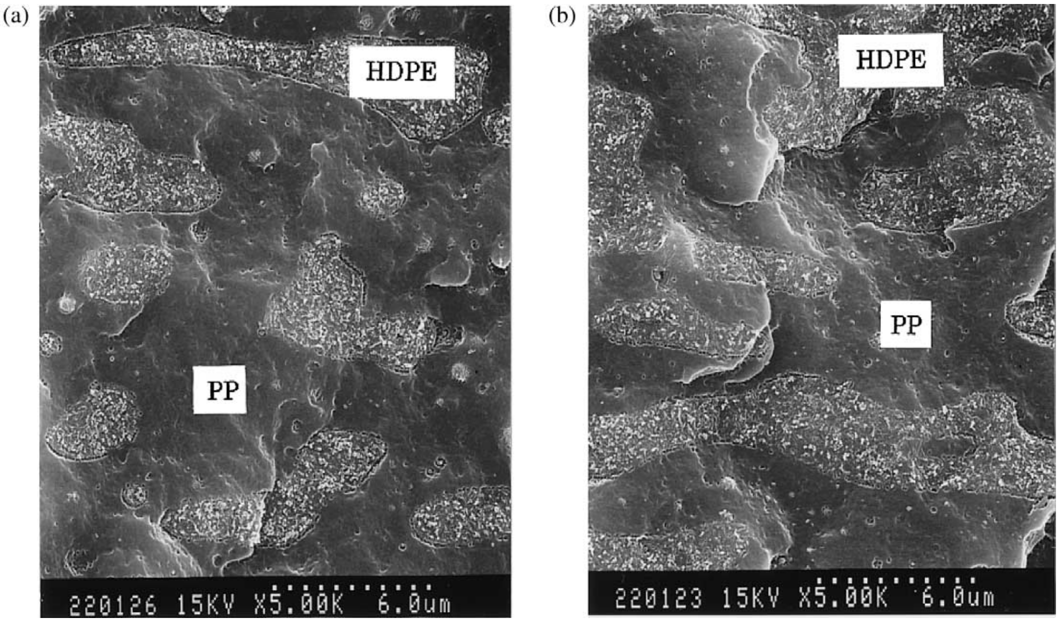

Fig. 1. SEM micrographs of press-molded HDPE/PP/Ketjen black (KB) with a volume ratio of (a) 30/68/2 and (b) 49/49/2.

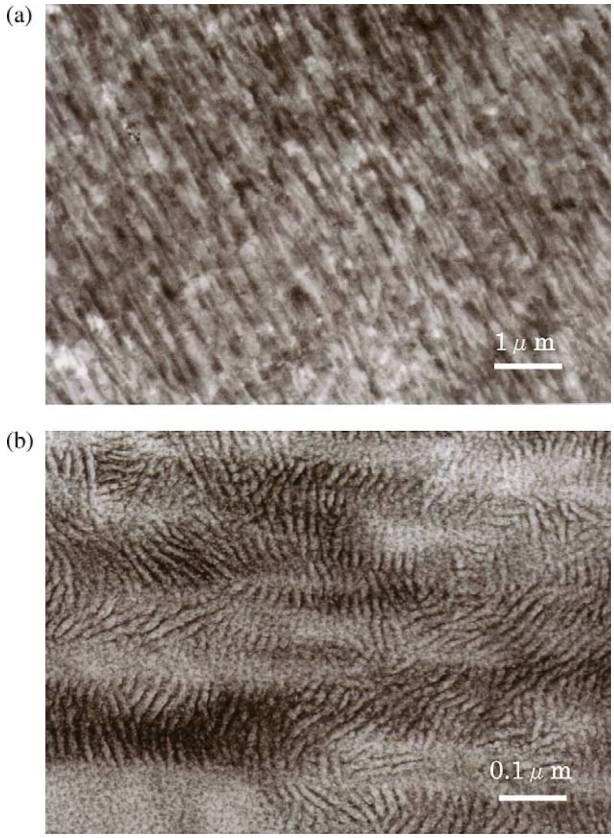

Fig. 2. TEM micrographs of injection-molded HDPE/PP (30/70) blends. (a) × 20,000; (b) × 200,000.

is, a relatively early stage of spinodal decomposition may be arrested by the crystallization to yield the regularly phase-separated structure in Fig. 2.

3.2. Influence of HDPE content

Fig. 3 (a)–(d) shows TEM micrographs for injection-molded PP/KB with addition of 0, 5, 10, and 29% HDPE, respectively. Compared to dispersion of the small KB particles in Fig. 3(a), addition of the HDPE component results in big KB agglomerates, and with increasing the HDPE content, these agglomerates are developed to contact together and form random networks, though the KB concentration in all of the samples is kept the same as 3%. This occurs because the KB particles are selectively located in HDPE domains. The increase in HDPE content raises the continuous HDPE/KB domain networks like press-molded samples shown in Fig. 1.

An interesting microstructure is observed for the sample containing 29% HDPE. As shown in Fig. 3(d), many orientated dark lines with a thickness of about 0.1 μm are distinguished from the carbon agglomerates and the PP matrix. To clarify the morphology, TEM micrographs at a higher magnification are shown in Fig. 4. One can find a typical microstructure in the 10% HDPE containing mixture (Fig. 4(c)), where the morphology consists of three parts: (A) CB–HDPE complex; (B) free PP domain (thin lamella part) and (C) free HDPE domain (thick lamella part). For composites with addition of HDPE content lower than 10%, no HDPE crystalline lamella is observed, indicating that HDPE molecules are strongly adsorbed on the carbon surface (like bound rubber) and thus

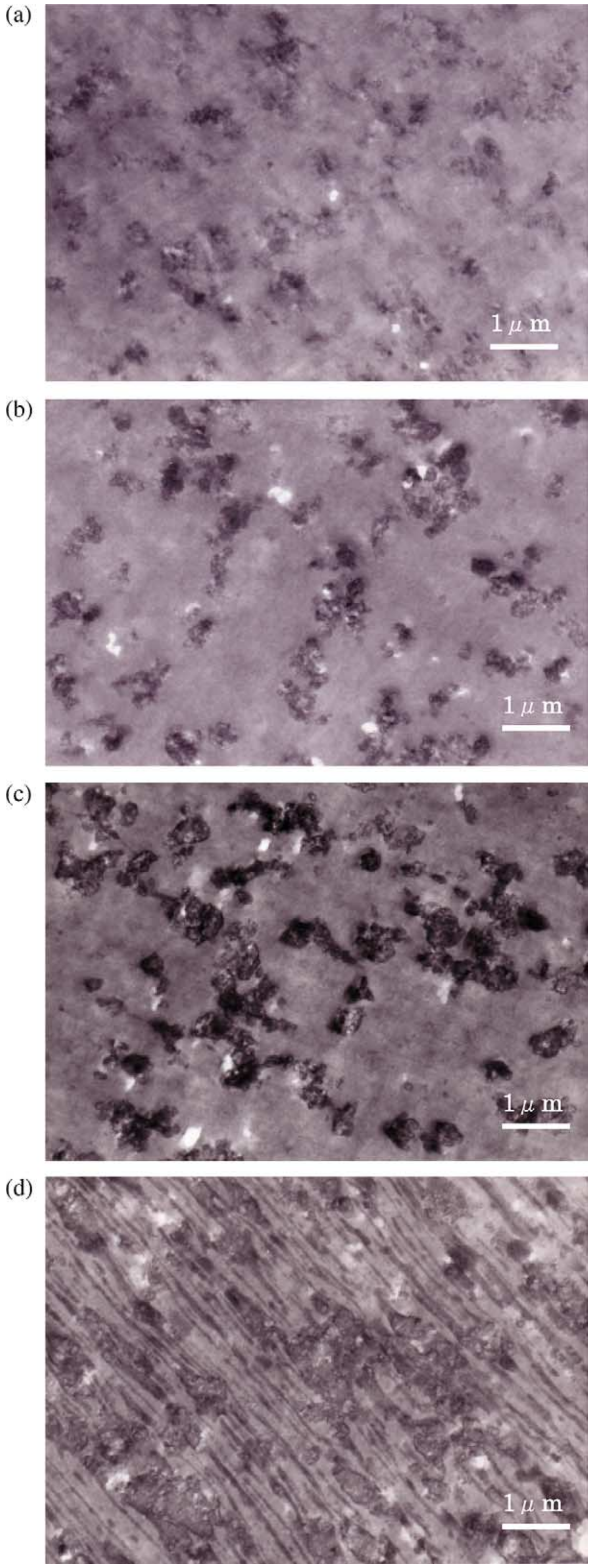

Fig. 3. TEM micrographs (×20,000) of injection-molded PP/KB with addition of (a) 0%, (b) 5%, (c) 10% and (d) 29% HDPE.

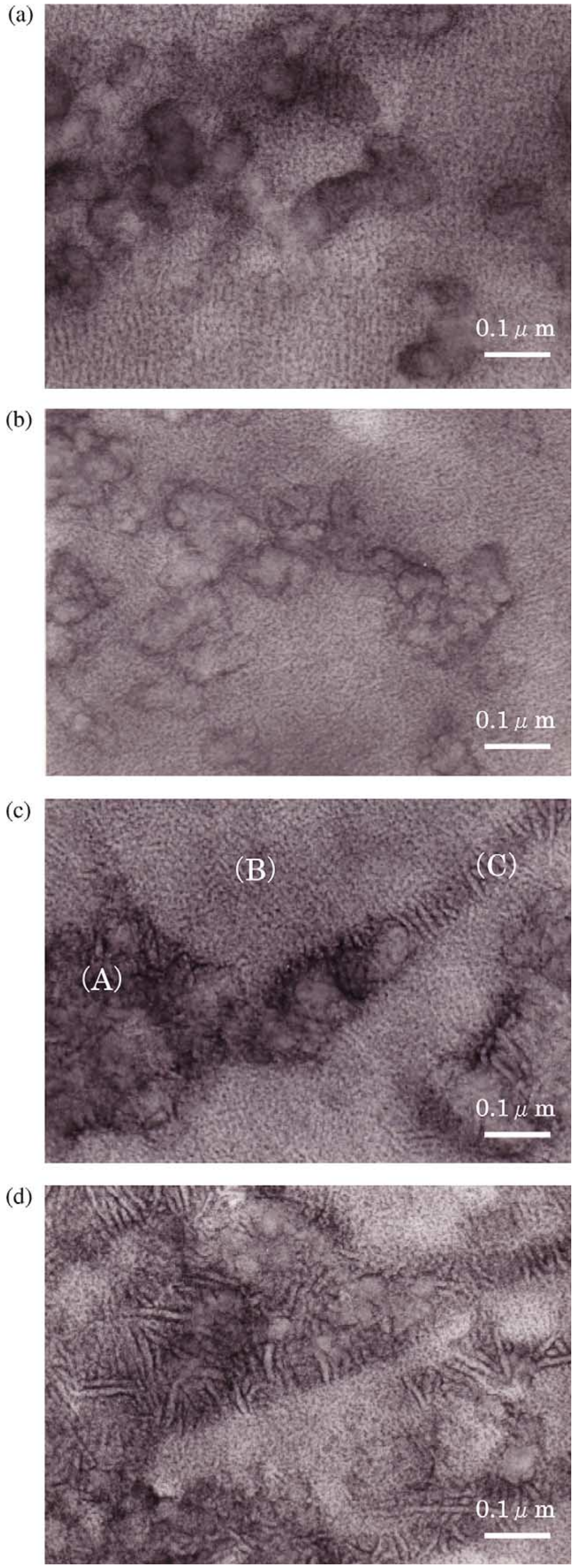

Fig. 4. TEM micrographs (×200,000) of injection-molded PP/KB with addition of (a) 0%, (b) 5%, (c) 10%, and (d) 29% HDPE.

disturbs the crystallization of the HDPE molecules. As the HDPE content is higher than 10%, the HDPE lamella is developed not only surrounding the KB particles but also at positions without KB. The orientated dark lines in Fig. 3(d) should be the crystallized free HDPE domains. This supra-organization is completely different from the morphology prepared from the press-molded samples (Fig. 1).

Shear-induced elongation and orientation of polymer domains along the flow direction is a well-known phenomenon nearby the surface of injection-molded polymer blends [21,22]. If an inorganic filler is selectively distributed in one of the polymer phases, then the distribution of the filler is thought generally following the deformed polymer domains. However, we found that the KB–HDPE networks are rather random (also see Fig. 7), and with increasing HDPE content, a large volume of HDPE can be separated from the KB–HDPE mixture forming elongated free HDPE domains. Our previous studies [5,6] have revealed that KB particles have strong tendency to self-agglomerate together. This may disturb the deformation of KB–HDPE domains along the flow direction resulting in the random KB–HDPE networks. Another fact is that HDPE is strongly interacted with PP at a high shear rate. HDPE molecules that are not adsorbed on the KB surface might solute into PP matrix and become a miscible polymer blend at the moment of injection molding and the fascinating structure in Fig. 3(d) could arise from the phase separation of the HDPE/PP blend when the high shear rate condition was removed.

3.3. *Influence of CB structure*

Suppose that the volume of free HDPE domains are determined by the total surface area of filled CB particles at a given HDPE concentration, then, the free HDPE domains would decrease with increasing the CB concentration or the specific surface area of CB. Figs. 5 and 6 show TEM micrographs for injection molded HDPE/PP/CB (28/67/5) using four types of CB (assigned as CB1, CB2, AB, and KB as listed in Table 2) with different particle size, specific surface area and *n*-dibutyl phathalate (DBP) adsorption number. Comparing the morphology of Fig. 5(d) (5% KB) to that of Fig. 3(d) (3% KB), we observe indeed that the volume of free HDPE domains decrease with the increase of KB concentration. From Figs. 5 and 6, one can also observe that the microstructure of free HDPE domains changes in two aspects with the change of CB structure, although the volume concentration of CB and HDPE in these specimens is kept the same. One is the volume of free HDPE domains. The volume of free HDPE domains from Fig. 5 seems to decrease in an order of CB1 > CB2 > AB > KB, which is not in agreement with the CB's particle size and specific surface area (we have to point out that this is our rough estimation and a more profound analysis is desirable), but in order of CB's DBP absorption number. The other is the structure of free HDPE domains. As shown in Fig. 5, the free HDPE domain is extended straightly in the sample filled with CB1 (Fig. 5(a)) while it is less straight in the CB2-filled sample (Fig. 5(b)). With increasing the CB's

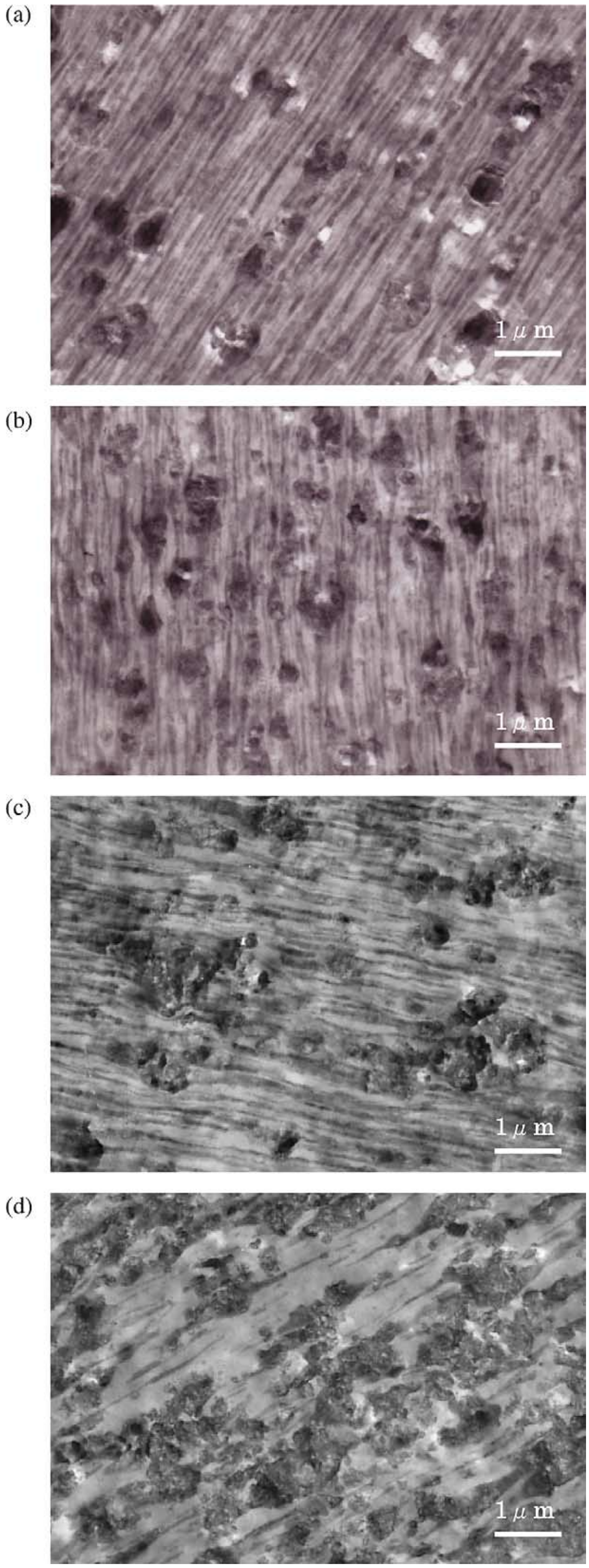

Fig. 5. TEM micrographs (×20,000) of injection-molded HDPE/PP/CB (28/67/5) mixed with (a) CB1, (b) CB2, (c) AB, and (d) KB.

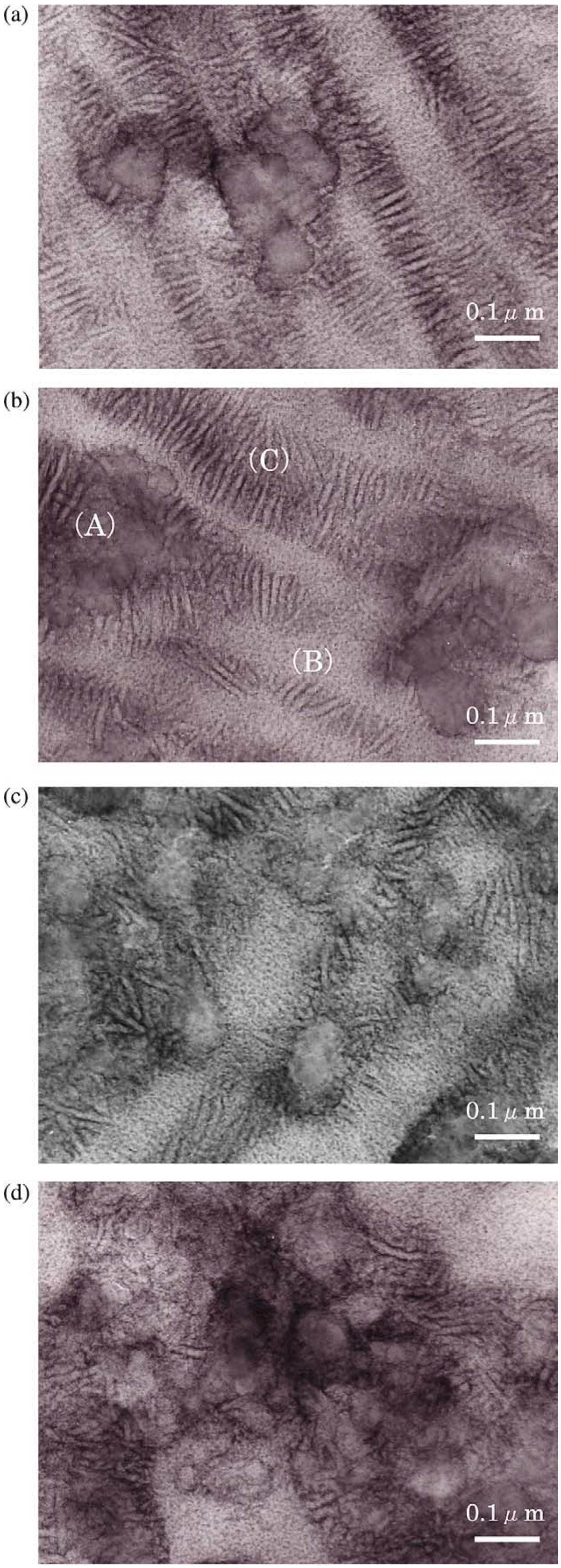

Fig. 6. TEM micrographs (×200,000) of injection-molded HDPE/PP/CB (28/67/5) mixed with (a) CB1, (b) CB2, (c) AB, and (d) KB.

DBP adsorption number, the elongated free HDPE domains become more random from the flow direction.

DBP adsorption number is a CB structure parameter characterizing the tendency to form the CB networks. A

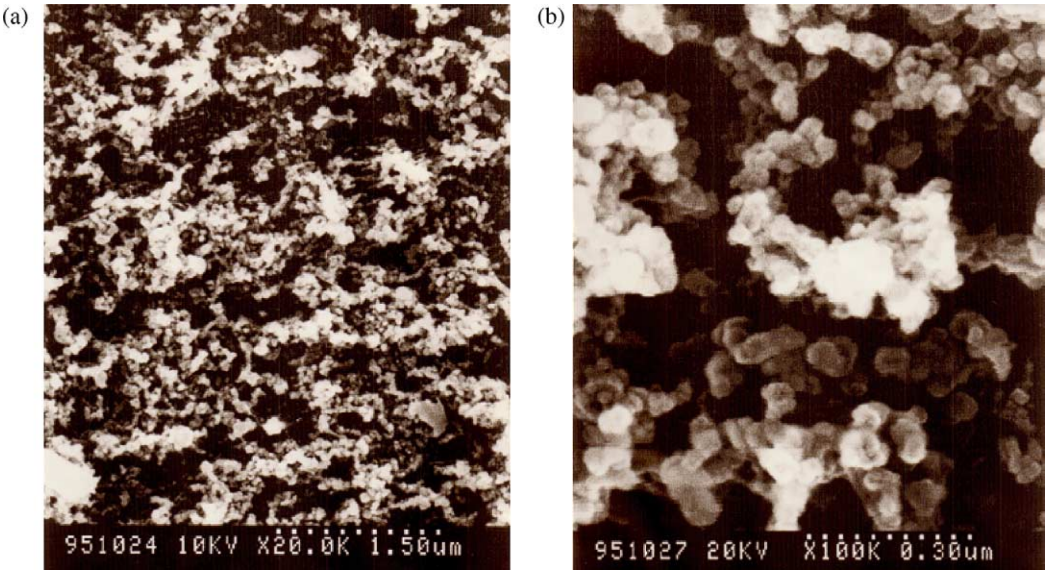

Fig. 7. SEM micrographs of injection-molded HDPE/PP/KB (28/67/5). (a)×20,000; (b)×100,000.

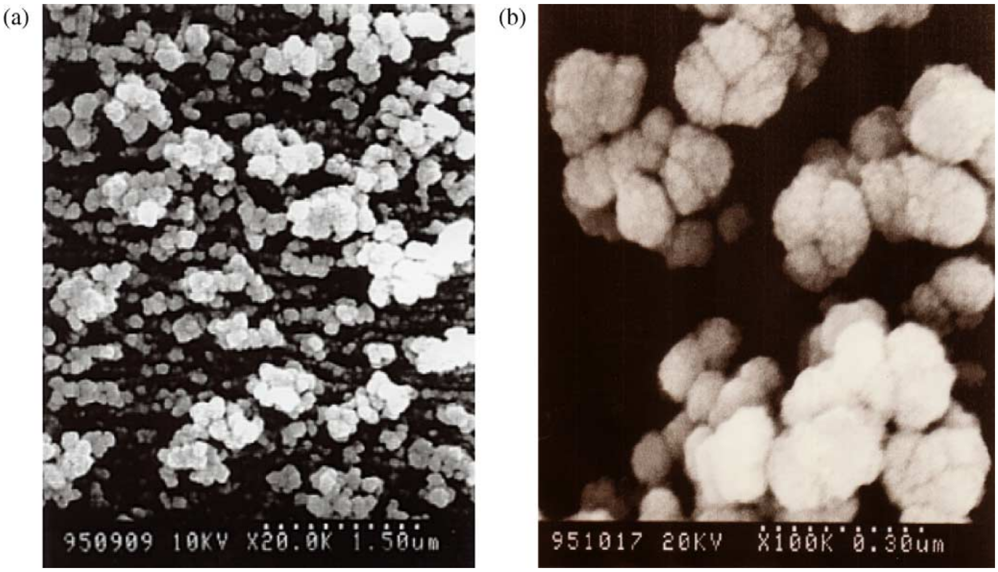

Fig. 8. SEM micrographs of injection-molded HDPE/PP/CB1 (28/67/5). (a)×20,000; (b)×100,000.

high DBP adsorption number is related to a high capability of the CB particles to aggregate together to form the 3D networks. Our results, that is, both of the volume and the structure of free HDPE domains are strongly influenced by the CB with a higher DBP adsorption number, indicate that the phase decomposition of HDPE/PP might be guided by the CB clusters, because CB is strongly interacted with HDPE and HDPE molecules should accumulate surrounding the CB particles.

Figs. 7 and 8 show SEM micrographs of HDPE/PP/KB and HDPE/PP/CB1, respectively, where polymers at the surface portion have been erased by ion etching. It is clear that KB particles agglomerate together and form random conductive clusters in the PP matrix (Fig. 7). For HDPE/PP/CB1 (Fig. 8), about 10 primary CB particles are aggregated together and these aggregates do not contact together to form networks although its composition is the same as that of the KB mixture (28/67/5).

3.4. *Influence of PP viscosity*

In order to improve the electrical conductivity, it is essential to promote the continuity of HDPE–CB complex domains throughout the PP matrix. An increase of PP viscosity might be effective since a high viscosity ratio of PP to HDPE generally reduce the critical HDPE concentration necessary for evolution from a 'sea-island' structure to a co-continuous two-phase morphology according to Jordhamo's equation [23]. However, our result getting from the injection-molded samples turns out contrary to the expectation.

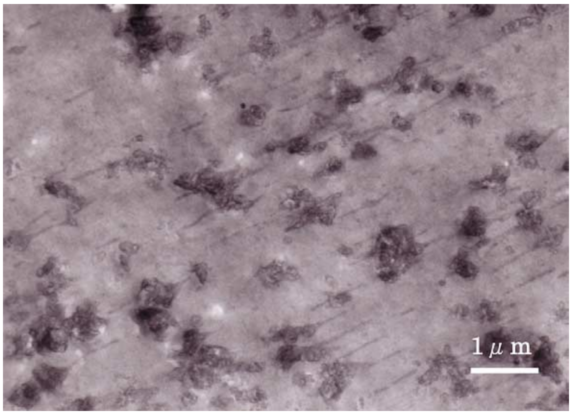

Fig. 9. A TEM micrograph (×20,000) of injection-molded HDPE/PP1/KB (10/87/3).

The morphology of HDPE/PP1/KB (10/87/3) using a relatively high molecular PP grade is shown in Fig. 9. Compared with that of Fig. 3(c), we can find that the HDPE–KB complex domains with high PP viscosity are relatively in a large size and seriously separated from each other. This can be ascribed to an intense shear experienced during injection molding of the high viscosity PP matrix. Due to the intensive shear experience, many HDPE molecules are drawn out from the HDPE–KB complex to form the free HDPE domains as seen in Fig. 9. We also believe that it is the high viscosity of PP matrix that prevents the HDPE–CB complex domains from forming the networks.

3.5. Electrical conductivity

The influence of HDPE content on the DC electrical resistivity at various KB concentrations for injection-molded specimens is summarized in Table 3. One can find that electrical resistivity of both PP/KB (3 vol%) and HDPE/KB (3 vol%) is higher than 10^7 Ω cm. However, as shown in Fig. 10, the electrical resistivity of PP/KB decreases rapidly as increasing the HDPE content, especially for mixtures with a lower KB concentration. The reduction in resistivity should be accounted for the selective location of KB in the HDPE domains and the effect of double percolation [8,9]. The increase of the HDPE content results in a higher ratio of the continuous HDPE–KB complex domains throughout the PP matrix, and thus, the KB concentration essential for percolation is reduced to a value lower than 2 vol%.

Table 3
DC volume resistivity of HDPE/PP/KB composites

Sample no.	vol%			Volume resistivity, ρ_v (Ω cm)
	PP	HDPE	KB	
1	98		2	$>1.0\times10^7$
2	97		3	$>1.0\times10^7$
3	95		5	2.8×10^1
4	92	5	3	$>1.0\times10^7$
5	87	10	3	8.2×10^4
6	77	20	3	4.0×10^3
7	68	29	3	1.4×10^3
8	68	30	2	3.7×10^5
9	85	10	5	1.9×10^1
10	75	20	5	1.6×10^1
11	68	28	5	1.8×10^1
12		98	2	$>1.0\times10^7$
13		97	3	$>1.0\times10^7$
14		95	5	5.1×10^1
15	70	30		$>1.0\times10^7$

The effect of CB structure on the DC resistivity for injection-molded HDPE/PP/CB is summarized in Table 4. As seen, a higher structure of the filled CB particles results in a lower electrical resistivity although these specimens have the same composition of 28/67/5. More exactly, the injection-molded specimen filled with the very high structure KB, that is, small particle size, high DBP absorption number and specific surface area, shows a very low resistivity in an order of 10^1 Ω cm while the low structure CB1-filled mixture is still in the range of electric insulation. A comparison between the AB and CB2-filled blends is quite difficult. AB carbon has a higher DBP absorption number and therefore it exhibits a higher tendency to self-assembly to form the conductive networks in the polymer blends as shown in Fig. 6. However, CB2 carbon owns a smaller particle size and a higher specific surface area, which are benefit for construction of more but looser conductive networks [1]. This is the reason why the resistivity of the AB and CB2-filled samples is in the same order of 10^5 Ω cm.

Frequency dependence of the electrical resistivity for the injection-molded specimens is shown in Fig. 11. One can observe that the AC resistivity of CB1, CB2, and AB-filled composites is much lower than the DC resistivity as listed in Table 4 and their resistivities decrease as increasing the frequency. Generally, conduction mechanism of the CB-filled polymer composites can be classified mainly into two types [3]. One is ohmic conduction due to the metallic contact among CB particles and, the other is non-ohmic conduction originated from existing small gaps between the

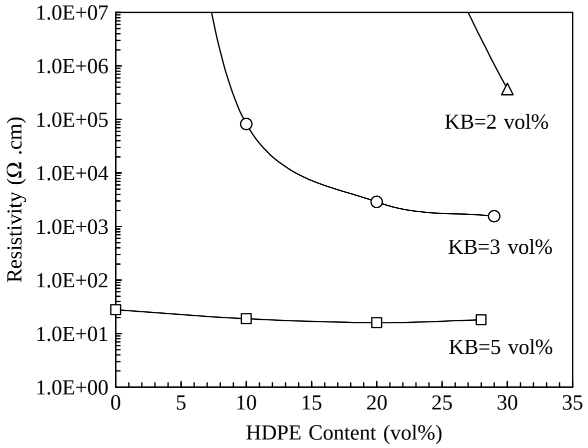

Fig. 10. Influence of HDPE content on DC resistivity for injection-molded HDPE/PP/KB composites at various KB concentrations.

Table 4
Influence of CB structure and PP viscosity on DC volume resistivity of HDPE/PP/CB composites

Sample no.	vol%			Volume resistivity, ρ_v (Ω cm)
	PP	HDPE	CB	
16	67	28	5 (KB)	1.8×10^{1}
17	67	28	5 (CB1)	$>1.0\times10^{7}$
18	67	28	5 (CB2)	1.7×10^{5}
19	67	28	5 (AB)	7.7×10^{5}
20	87	10	3 (KB)	8.2×10^{4}
21[a]	87	10	3 (KB)	$>1.0\times10^{7}$

[a] High viscosity PP1 was blended.

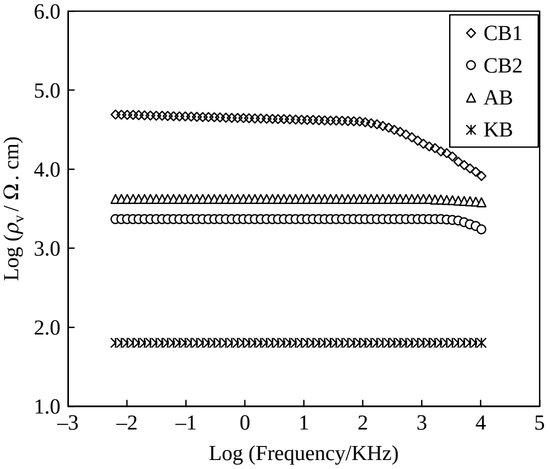

Fig. 11. Frequency dependence of resistivity for injection-molded HDPE/PP/CB (28/67/5) composites.

CB particles. Electric behavior of the small gap can be equivalent to a capacity where electrons are easier to jump under a higher applied voltage and frequency. Comparing to the non-ohmic conduction of CB1, CB2, and AB-filled HDPE/PP mixtures, KB-filled composites shows an ohmic conduction demonstrating a close contact of HDPE–KB complex in the PP matrix.

The influence of PP viscosity on AC resistivity is shown in Fig. 12. A high viscosity of PP matrix results in a high resistivity due to the separation of HDPE–KB complex as observed by TEM measurements (Fig. 9). It is interesting to note that the resistivity of the specimen blended with the high viscosity PP decreases rapidly as increasing the frequency and at very high frequencies, it is close to the value of the low viscosity specimen.

4. Conclusions

The influence of HDPE content, CB structure and PP viscosity of on morphology and electrical conductivity of injection-molded HDPE/PP/CB composites was investigated.

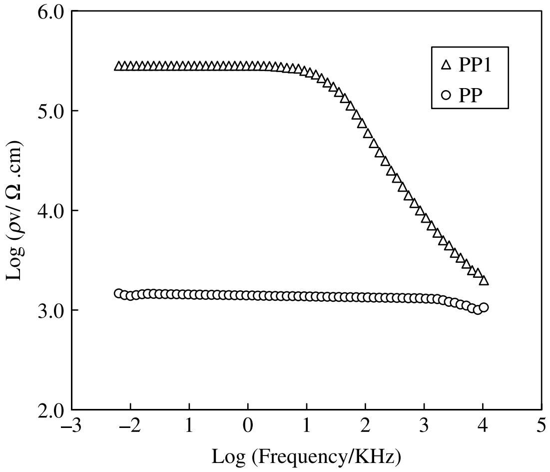

Fig. 12. Influence of PP viscosity on Ac resistivity for injection-molded HDPE/PP/CB (10/87/3) composites.

It was observed that the electrical resistivity of these mixtures decrease with the increase of HDPE content and increases with increasing the viscosity of PP matrix. The compound filled with high-structured CB, that is, small CB particle size, high DBP absorption number and high specific surface area, significantly alerts the morphology and leads to a low resistivity.

Electron microscopy observation revealed for the first time that HDPE domains near to the surface of injection-molded samples were divided into two parts due to the high shear deformation. One is the free-HDPE domain and the other is the HDPE–KB complex domain. Not only the volume but also the microstructure of the free-HDPE domains changes greatly with HDPE content, CB concentration and CB structure. It is necessary to further investigate the relationship between the free-HDPE domains and surface appearance, electrical, and mechanical properties.

A serious deformation of HDPE–KB complex domains was not observed along the shear flow, demonstrating that CB particles have strong capability of self-assembly to form random conductive networks.

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