Thermal, Dielectric, and Mechanical Properties of SiC Particles Filled Linear Low-Density Polyethylene Composites

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ABSTRACT: A thermally conductive linear low-density polyethylene (LLDPE) composite with silicon carbide (SiC) as filler was prepared in a heat press molding. The SiC particles distributions were found to be rather uniform in matrix at both low and high filler content due to a powder mixing process employed. Differential scanning calorimeter results indicated that the SiC filler decreases the degree of crystallinity of LLDPE, and has no obvious influence on the melting temperature of LLDPE. Experimental results demonstrated that the LLDPE composites displays a high thermal conductivity of 1.48 $Wm^{-1} K^{-1}$ and improved thermal stability at 55 wt % SiC content as compared to pure LLDPE. The surface treatment of SiC particles has a beneficial effect on improving the thermal conductivity. The dielectric constant and loss increased with SiC content, however, they still remained at relatively low levels ($<10^2$ Hz); whereas, the composites showed poorer mechanical properties as compared to pure LLDPE. In addition, combined use of small amount of alumina short fiber and SiC gave rise to improved overall properties of LLDPE composites. © 2009 Wiley Periodicals, Inc. J Appl Polym Sci 112: 1695–1703, 2009

Key words: composites; polyethylene (PE); thermal properties; dielectric properties; mechanical properties

INTRODUCTION

During the last few decades, in the microelectronic systems great effect has been contributed to improving higher integration density, faster performance, miniaturization of electronic devices and lower cost.[1–4] With the further miniaturization and increasing power of microelectronics, heat dissipation has become critical to the performance, reliability and further miniaturization of microelectronics since high integration of transistors has resulted in the escalation of power dissipation as well as an increase in heat flux at the devices.[5–7] However, cooling is often practically restricted by a rather low thermal conductivity of the molded plastic packaging materials which are widely used for electronic packaging and/or substrate.[8–10] The functions of molded plastic packages are to protect semiconductor devices from exposure to environmental hazards such as moisture, chemical agents, dust and light, and also to provide them with excellent mechanical strength.[11] As the power density becoming larger and larger, better properties are required for substrate and packaging materials, such as a high thermal conductivity, a low coefficient of thermal expansion, and a low dielectric constant.[12] Therefore, highly thermal conductive molding compounds are emerging as a cost-effective mean to remove heat accumulated from plastic package.[13,14] This has led to the quest for plastic packaging materials with an enhanced thermal conductivity.[15,16]

Plastics reinforced with ceramic particles with a high thermal conductivity are attractive candidates in this area because they can provide a required thermal conductivity while maintaining desired electrical insulation properties.[8,12] There are many references in the plentiful literature concerning adding thermally conductive fillers to plastics to produce a more heat conductive composites. For example, many composite plastics have been investigated including different kinds of fillers such as boron nitride,[17] aluminum nitride,[7,15,18] silicone nitride,[7,19] alumina (Al_2O_3),[20] silicon carbide (SiC),[21] silica,[22] and diamond[23]; and various polymer matrix such as high density polyethylene, linear low-density polyethylene (LLDPE), ABS, polyamide, polypropylene, etc.

The dispersion status of filler has an important effect for the thermal conductivity which is mainly

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dictated by the conductive chains or pathways formed from filler particles. Agari and Ueda[24] and Yu et al.[15] found that the use of powder mixing can lead to a special dispersion of filler in polymer matrix, i.e., filler particles surrounding or covering the matrix particles. So, it is possible to improve the thermal conductivity of composites at low filler content under this kind of special dispersion of filler as compared to using the melted mixture and other processing methods.[17] To achieve a maximum thermal conductivity at low filler content, and minimize the adverse effect of the filler on the mechanical, processing and other properties of composites, the particles mixture of polymer and filler particles is a preferable mixing way. Up until now, the investigations of the heat conductive SiC filled LLDPE composites fabricated by a powder mixing method have been little reported before. So, the aim of this study was to prepare a novel SiC/LLDPE plastic composite by a powder mixing, and investigate the thermal, dielectric and mechanical properties. To understand the fundamentals of interface effect on the thermal conductivity of polymer composite, a titanate coupler was used to functionalize the surface of SiC particles to improve the interface combinations. Additionally, to improve the mechanical properties and thermal conductivity of the SiC/LLDPE composites an attempt was made to use small amount of Al_2O_3 short fiber in the formulation to meet the dual goal.

EXPERIMENTAL

Materials

The matrix component used was LLDPE with a brand of Hyundai SR646 from Hyundai Group (Seoul, South Korea), and its physical property (offered by the producer) is listed in Table I. In this investigation the big LLDPE particles were milled into fine powder with an average size of 100 $\sim$ 150 μm for powder mixing. A light green SiC powder supplied by Qingyang Abrasives Co. (Henan, China) was used as thermal conductive filler, which has a melt point of over 2500°C, a density of 2.27 g/cm^3, a thermal conductivity of 30–50 Wm^{-1} K^{-1} and the average particle size of 5 $\sim$ 7 μm. The Al_2O_3 short fiber used has an average diameter of 6–10 μm and length of 1 $\sim$ 2 mm, purchased from Shanghai Silicate Research Institute (Shanghai, China). A titante coupler (type 201) with a purity of 99% was from Nanjing Xiangfei Chemical Co. (Nanjing, China). The polyethylene wax (type: H112) offered by Guangzhou Shencai Trade Co. (Guangzhou, China), was used to improve the processing property of the SiC/LLDPE composites.

TABLE I
Properties of the LLDPE Used

Resin properties	Values	ASTM
Melt index/g/10 min	45	D1238
Density/g/cm^3	0.930 $\sim$ 0.934	D1505
Ultimate tensile/MPa	24	D638
Ultimate elongation/%	620	D638
Vicat softening point/°C	110	D1525
Flexural modulus/MPa	570	D790
IZOD impact/kJ/m^2	65	D256

Samples preparation

Surface modification of SiC particles

The surface treatment for the SiC particles with the titante coupling agent involved the following steps: (a) dissolving the titante coupling agent (1.0% of the SiC filler mass) in methylbenzene; (b) adding SiC particles to the solution made in step (a) and stirring for 20 min; (c) heating the mixture to 80°C, refluxing it for 3 h while stirring, and then cooling it to room temperature and setting it for 5 h; (d) rinsing the mixture with methylbenzene by filtration at least three times; and (e) drying the mixture at 120°C for 10 h.

Preparation of SiC- reinforced Lldpe

The LLDPE powder, small amount of polyethylene wax and SiC particles were dry mixed in a ball-milled mixing container whose size is 200 mm in diameter and 320 mm in length (Model: AG-10, rolling at a speed of 25 rpm, Zhengzhou Xiangyu milling machine, Zhengzhou, China) at room temperature about for 1 h, until the SiC particles surrounded or covered the surface of LLDPE particles. After that, the mixture was transferred to a stainless steel die and melt pressed at 190°C in an electrically heated hot press machine (type: SL-45, Shanghai, China) with a pressure of 15 MPa for 30 min, and was allowed to cool for 30 min at room temperature. Composites with SiC powder concentrations ranging from 0 to 55 wt % were prepared.

Characterization

The differential scanning calorimeter (DSC), (Model, DSC 200PC, Netzsh Corp., Selb, Germany), was used to analyze the influence of the SiC content on the melting temperature and melting enthalpy of the LLDPE samples (5 $\sim$ 10 mg) directly cut from the molded sheet. Measurements were conducted in a nitrogen atmosphere, from 20 to 200°C, at a heating rate of 10°C/min.

Thermal conductivity (Wm^{-1} K^{-1}) measurements was carried on the specimens (with diameters of 12 mm and thicknesses of 2 mm) using the guarded

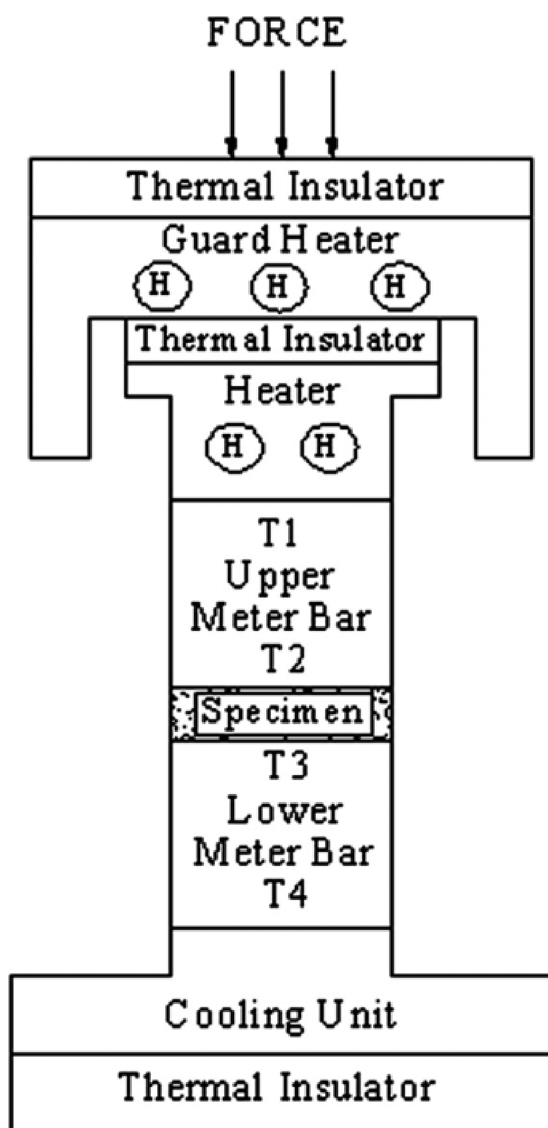

Figure 1 A schematic of the guarded heater meter.

heater meter apparatus as displayed in Figure 1. This equipment (DMR-2, Xi'an Aerospace Materials Research Institute, Xi'an, China) was fabricated according to the ASTM D-5470-06. The measurements were performed by clamping the samples between the two calorimeters and heating the heater block up to a previously set temperature, to produce an average specimen temperature. A chiller plate with circulating water at a predefined temperature was placed above the cooling calorimeter to create a thermal gradient from the heating calorimeter to the cooling calorimeter. During the measurements, an onset pressure of about 1.5 bar was applied to reduce the effect of the contact resistance between the specimen and the calorimeters due to minor surface irregularities. Readings from the thermocouples are recorded when a thermal equilibrium was achieved, whereby two successive sets of temperature readings were taken at 15 min intervals, displaying a difference of ±0.1°C. The thermal conductivity of the samples were calculated from the Fourier's law, as shown in eq. (1), based on the assumption that the heat flow is one dimensional in the perpendicular direction, and no heat loss occurs in the lateral direction:

$$k = \frac{Q \cdot \Delta L}{A[T_{\mathrm{hot_int}} - T_{\mathrm{cold_int}}]} \quad (1)$$

where k is the thermal conductivity of the samples ($\mathrm{Wm^{-1}\ K^{-1}}$), Q the average heat flux generated by the cartridge heaters (W), $T_{\mathrm{hot_int}}$ the interface temperature of the heating calorimeter (K), $T_{\mathrm{cold_int}}$ the interface temperature of the cooling calorimeter (K), A the surface area of tested samples ($\mathrm{m^2}$) and ΔL is the thickness of samples (m). The average heat flux (Q) is the amount of heat generated by two cartridge heaters, whereas the interface temperature for both the heating and cooling calorimeter were extrapolated from the temperature gradients obtained from the three thermocouples embedded in each of the calorimeter block.[9]

Weight loss of the samples (10 ~ 15 mg) upon heating was measured using a thermogravimetric analyzer (model: SDTA851, Swiss). Measurements were conducted in a nitrogen atmosphere, from 25 to 600°C, at a heating rate of 15°C/min. The observed weight loss was analyzed.

Morphological observation on the samples was performed by means of a scanning electron microscope (SEM) (model: JSM-7000F, JEOL, Japan). The observation was carried out on the cross sections of samples to study filler distribution.

The dielectric measurement was performed on a broadband dielectric spectrometer (Novocontrol Technology Company, Germany) with Alpha-A high performance frequency analyzer. The measurement was carried out in the frequency range from 10^{-1} to 10^{7} Hz under room temperature. The specimens for dielectric measurement were made in the form of circular discs with 1 mm in thickness and 20 mm in diameter. Both sides of the disks were suitably sprayed with gold powder to improve electrical contact. Generally, at least two samples were tested to check reproducibility. All measurements were carried out in the cryostat to avoid possible surrounding effects.

The volume resistivity and surface resistivity measurements were performed on an ultrahigh electric resistor (model: ZC-36, Shanghai Yiren Electric Co., Shanghai, China) at room temperature according to the GB/T1410-1992. The dimension of the samples for tests is 80 mm in diameter, 1 mm in thickness.

Tensile test was conducted on a hydraulic universal testing machine (model: ZMGI 250, Shenzhen

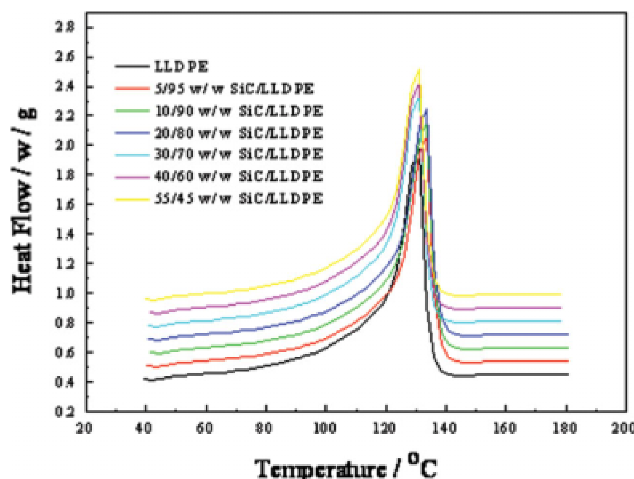

Figure 2 DSC melting curves of LLDPE and its SiC powder composites. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com].

New SANS, Shenzhen, China) at a cross-head of 50 mm/min to determine the tensile properties of the materials according to the GB/T170371.1, GB/T 1040 standards, respectively.

RESULTS AND DISCUSSION

Melting temperature and degree of crystallinity

LLDPE is a semicrystalline polymer. The portion of the crystalline phase places an important influence on almost all physical (such as thermal, mechanical) properties of polymers. It is therefore important to investigate the effect of SiC on the change of degree of crystallinity of LLDPE.[2,17]

The DSC melting curves of LLDPE and its SiC powder composites are plotted in Figure 2. It can be observed that the SiC content does not seem to have much influence on the melting temperature of LLDPE, and the peak temperature of melting slightly shifts toward a lower temperature with increasing SiC loading. This may be explained by the reason that the SiC filler probably reduces of lamellar thickness of crystallites and thus it leads to a decrease of melting temperature,[2,12] because some SiC particles will fill themselves in the interlamellar space due to a higher degree of crystallity of pure LLDPE.

The absolute crystallinity (X_c) was calculated according to eq. (2)

$$X = \frac{\Delta H_f}{(1-x)\Delta H_f^0} \tag{2}$$

where X_c is degree of crystallinity, X is mass fraction of filler, ΔH_f specific melting enthalpy of LLDPE andΔH_f^0 specific melting enthalpy for 100% crystalline polyethylene, which is set as 290 J/g[25–28] for this calculation.

The melting temperature and specific melting enthalpy of pure LLDPE and its SiC composites are summarized in Table II. It can be seen from Table II that the use of SiC particles obviously decreases the crystallinity of filled LLDPE as compared to pure LLDPE. The reason for this is that LLDPE has a relatively high crystallinity, and there is not a major amorphous part in which the SiC particles can be accommodated. At low SiC contents the filler particles locate themselves in the interlamellar spaces, which leave little room for additional crystallization, and the presence of these particles may even inhibit crystallization. At high SiC concentrations there is probably a change in crystallization mechanism, i.e., with an epitaxial crystallization on the SiC surfaces being the major contributor to polymer matrix crystallinity.[12]

It may be conjectured without detailed microscopic evidence that the crystalline domains formed by LLDPE are rendered smaller in the presence of SiC particles and reduce the overall crystallinity as the filler increases. Therefore, it is possible for the fillers to decrease the mobility of LLDPE chains in the formation of crystallites and, as a result, the domains of crystalline phase are reduced in size.[29–31] It is also likely that imperfection of crystals in the presence of the SiC inhomogeneities contributes to the decrease in crystallinity.[31]

Thermal conductivity and thermal stability

It is generally known that the amorphous polymers have similar thermal conductivity.[2] In the case of semicrystalline polymers, an increase of thermal conductivity with an increase in crystalline part content was observed as a consequence of a better transport of the heat in a crystalline phase.[2,12,32] The simple relation between thermal conductivity of semicrystalline polymers and the weight portion of crystalline phase is expressed in terms of the eq. (3)

$$k_m = k_c w_c + k_a(1 - w_c) \tag{3}$$

where k_m, k_c, k_a is thermal conductivity of polymer and its crystalline and amorphous part, respectively,

TABLE II
Specific Melting Enthalpy, Melting Temperatures, Degree of Crystallinity of LLDPE and its Composites

Material	ΔH_m/J/g	T_m/°C	X_c/%
LLDPE	144.3	130.2	58.9
5/95 w/w SiC/LLDPE	128.6	129.0	45.8
10/90 w/w SiC/LLDPE	123.0	127.5	46.3
20/80 w/w SiC/LLDPE	103.9	127.6	44.0
30/70 w/w SiC/LLDPE	98.2	127.3	47.5
40/60 w/w SiC/LLDPE	84.7	126.9	47.8
55/45 w/w SiC/LLDPE	60.4	127.0	45.5

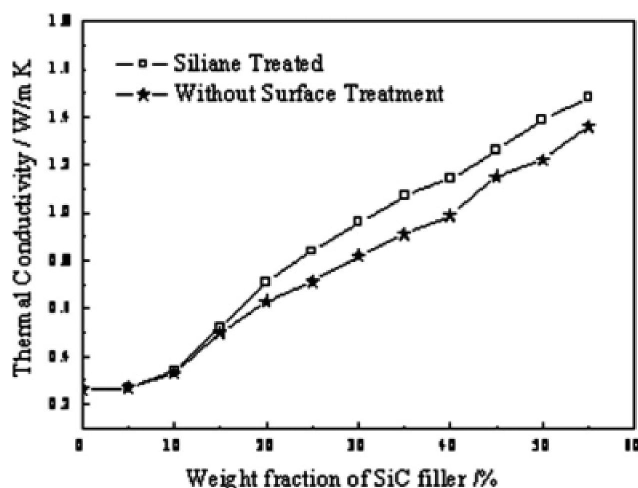

Figure 3 Thermal conductivity of filled LLDPE as a function of SiC content.

w_c is weight portion of crystalline phase of polymeric matrix.

From Table II the addition of SiC particles in LLDPE reduces the degree of crystallinity of LLDPE, thus, the thermal conductivity of LLDPE itself is lowed according to eq. (3). However, the change in thermal conductivity of LLDPE itself has negligible influence on the thermal conductivity of SiC reinforced LLDPE composites because of the big thermal conductivity values as compared to that of LLDPE. There is also little change in degree of crystallinity with increasing SiC content for filled LLDPE. For all SiC contents, the change in degree of crystallinity calculated according to eq. (3) is too small and has negligible influence on the thermal conductivity of LLDPE matrices as well as composites, especially if the average experimental error of thermal conductivity measurements being $\sim$ 5-7%.[2]

The thermal conductivity of SiC particles used is about 35–70 Wm^{-1} K^{-1} compared to 0.26 Wm^{-1} K^{-1} for LLDPE is. So, the incorporation of SiC in LLDPE will increase the thermal conductivity of composites, and the filler content has a remarkable effect on the thermal conductivity. The thermal conductivity of SiC/LLDPE composites versus filler content are presented in Figure 3. It is clear that the thermal conductivity of the composites is higher than that of pure LLDPE due to a high thermal conductivity of SiC filler, as compared to pure LLDPE. A nonlinearly increases in thermal conductivity was observed with increasing SiC concentration. At 50 wt % SiC content, the thermal conductivity reached 1.22 $\sim$ 1.37 Wm^{-1} K^{-1}, near five times that of pure LLDPE.

From Figure 3 it also can be seen that the use of titanate coupling agent to functionalize the surface of SiC obviously improved the thermal conductivity compared to the untreated filler reinforced LLDPE at SiC content greater than 15 wt %. It is well known that the transport of heat in all nonmetals (no free electrons) is by the flow of lattice vibration energy,[7] or phonons, along the temperature gradient within the specimen; for a two phase system like SiC/LLDPE composites, interfacial physical contact between polymer and filler is very critical, since phonons are very sensitive to surface defects.[33,34] Thermal resistance is caused by various types of phonon scattering processes, and the interfacial thermal barriers in composites is mainly due to the scattering of phonons resulting from acoustic mismatch and flaw associated with the matrix-filler interface.[35] The interface between the two-phase composites acts as a barrier of heat transmission. So, the surface treatment of SiC with titanate coupler improved the interfacial bonding between SiC and matrix, and reduced the air voids (k_{air} = 0.0024 Wm^{-1} K^{-1}) at the filler-matrix interface, which facilitates enhancing the thermal conductivity of composites.

The dispersion state of SiC filler in LLDPE matrix affects the thermal conductivity significantly because thermal conductivity is mainly dictated by the numbers of conductive pathways or networks formed from conductive filler in matrix. While, the different processing method for the composites often gives rise to different dispersion state of filler in composites. Therefore, the processing way has an effect on the thermal conductivity of composites, which had been proved by Agari and Ueda.[17,24]

The micro-structure images of LLDPE filled with 30 wt % and 55 wt % of filler are depicted in Figure 4. The SiC powder distributions are found to be relatively rather uniform at both low (30 wt %) and high (55 wt %) filler content, and there is even not clusters formation of SiC particles at high filler content. This can be mainly ascribed to the powder mixing because it can be seen clearly from Figure 4 that the LLDPE particles were essentially surrounded by the SiC particles although there is some connectivity between adjacent LLDPE particles in small regions. This dispersion results in obtaining connected SiC particles along the surface of larger LLDPE matrix particles. Thus, the most of adjacent SiC particles can touch each other, and form some conductive pathways in composites at high filler loading. The thermal resistance of the composites mainly due to a low thermal conductivity of LLDPE, is therefore reduced owing to the connectivity of some filler particles, thus, it results in a high thermal conductivity.[15,17]

Owing to a high thermal conductivity of SiC particles, as compared to LLDPE, when the composites are heated, just as electric charge flows where resistance is as lowest as possible, the heat will flow through the SiC particles. Thus, the conductive channels formed in the composites under this dispersion

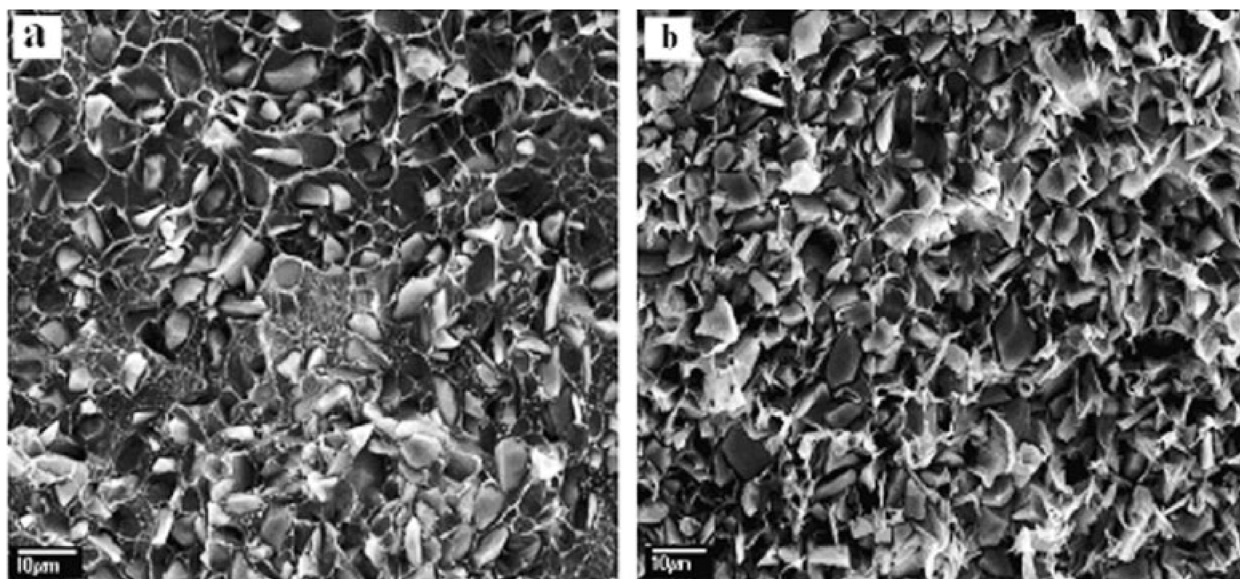

Figure 4 Microstructure of SiC/LLDPE composites at the SiC weight of (a) 30% and (b) 55%.

state of filler using powder mixing cause a higher conductivity as compared to a melted LLDPE matrix.[15] Therefore, the powder mixing gives the SiC filler a net-worked dispersion state in LLDPE, which aided to improving connectivity of filler in polymer matrix.

The TGA curves for LLDPE and its SiC particles filled composites are illustrated in Figure 5.

There is a slight increase in the thermal stability of LLDPE with increasing SiC concentration.

The reason may be ascribed to the higher heat capacity (1.27 Jg^{-1} $°C^{-1}$, as compared to 0.18 Jg^{-1} $°C^{-1}$ of LLDPE) and thermal conductivity of SiC, which will cause it to preferably absorb the heat. So, this will result in LLDPE chains starting to degrade at higher temperature.[12] Figure 5 also reveals that there is not a remarkable increase in the thermal stability of LLDPE with increasing SiC loading, for example, the decomposition temperature of LLDPE is only near 5°C lower than that of the 55 wt % SiC filled LLDPE. The thermal stability behavior of LLDPE is similar to that of the LLDPE composites containing various SiC content. This may be explained by the higher crystalinity of LLDPE. For LLDPE, there is not a major amorphous part in which the SiC particles can be accommodated[2,3,17]; therefore, the SiC particles probably locate themselves in the interlamellar spaces, bringing them in close contact with the lamellar surfaces. The higher heat capacity and thermal conductivity of SiC causes these particles to quickly reach a higher temperature than the surrounding matrix, thus resulting in degradation at lower temperature of the already strained fold surfaces.[12] It also can be seen that for pure LLDPE and filled LLDPE composites the total mass loss values are completely in agreement with the amount of SiC originally mixed into the samples.

Dielectric properties

In plastic packaging the dielectric properties of materials have an important role in device performance. Both the dielectric constant and dielectric loss greatly influence the signal-carrying capacity and the speed of the device to pass signals. Generally, low dielectric constant and loss values make a high accumulation of the device itself and a high clock rate possible.

The signal propagation delay caused in an integrated circuit can be determined by eq. (4) as a

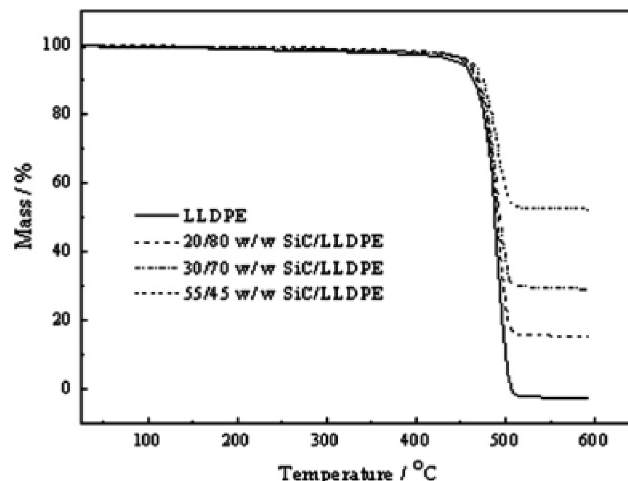

Figure 5 TGA curves of LLDPE and its SiC powder composites.

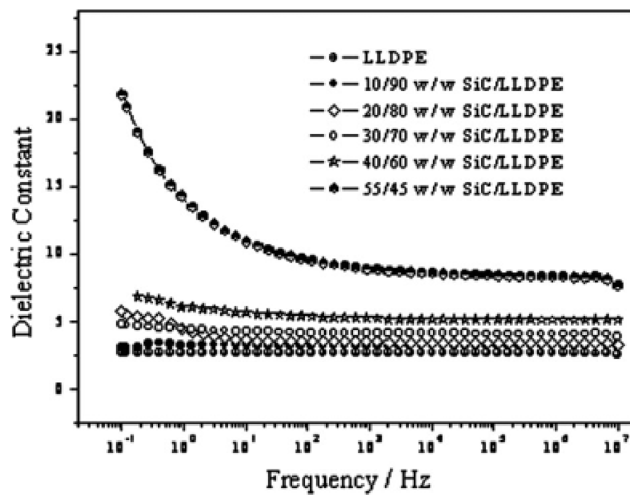

Figure 6 Frequency dependence of dielectric constant for LLDPE and its composites.

means of the dielectric constant of the packaging materials.

$$T_d = \frac{l}{c}\sqrt{\varepsilon_r} \quad (4)$$

where c is the velocity of light, l the transmission distance of the singles and ε_r the relative dielectric constant.

From eq. (4), it can be seen that a high dielectric constant of the packaging material will cause a bad effect on the signal propagation by longer the delay time,[7] and a low dielectric constant and loss is required for polymer packaging materials.

The frequency dependencies of the dielectric constant and loss of the SiC/LLDPE composite, are displayed in Figure 6 and Figure 7, respectively.

Figure 6 demonstrates that the dielectric constant of the composites increases with an increase in SiC particles loading. The dielectric constants of pure LLDPE and its SiC composites (except for 55 wt % SiC-LLDPE) are almost independent of the frequency in the measured frequency range from 10^{-1} to 10^7 Hz, and they keep at rather low levels, i.e., generally less than 7.0, whereas, that of the 55 wt % SiC filled LLDPE starts to rise under 10^2 Hz, and continues to rise to 22 at 10^{-1} Hz.

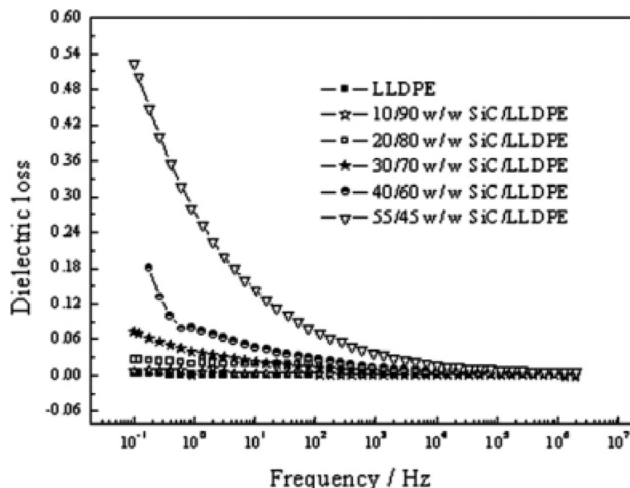

Figure 7 Frequency dependence of dielectric loss for LLDPE and its composites.

TABLE III
Volume Resistivity and Surface Resistivity of SiC/LLDPE Composites

SiC content/wt %	Volume resistivity/Ω cm	Surface resistivity /Ω
0	4.6×10^{17}	1.4×10^{17}
5	1.3×10^{17}	5.1×10^{16}
10	8.4×10^{16}	6.2×10^{15}
20	2.1×10^{15}	2.9×10^{14}
30	3.2×10^{14}	4.7×10^{13}
40	5.4×10^{13}	5.8×10^{12}
55	7.2×10^{12}	3.5×10^{11}

It can be observed from Figure 7 that the dielectric losses increase as an increase in the SiC content. Figure 7 further shows that the dielectric losses of pure LLDPE and its 10 and 20 wt % SiC composites remain very low values, and are almost independent of the frequency in the measured frequency range from $10^{-1}$to 10^7 Hz, whereas, those of the 30, 40, and 55 wt % SiC filled LLDPE composites start to rise under 10^1, 10^2, and 10^4 Hz, respectively. Generally speaking, the dielectric losses of the above three composite materials still keep at rather low levels, i.e., less than 0.025 at frequency greater than 10^1 Hz for the 30 wt % SiC filled LLDPE, and obviously less than 0.05 and 0.1 at frequency greater than 10^2 Hz for the 40 and 55 wt % SiC filled LLDPE, respectively.

Therefore, the dielectric analysis demonstrates that the SiC filled LLDPE composites have possessed rather low dielectric constant and loss at the measured frequency range from 10^2 to 10^7 Hz, and the low dielectric property is very important for the LLDPE packaging materials.

The dependence of electrical insulation properties of the SiC/LLDPE composites on the weight percentage of filler is shown in Table III. Table III illustrates that both volume resistivity and surface resistivity continuously declined with an increase in filler loading, and reached 7.2×10^{12} Ω cm and 3.5 $\times$ 10^{11} Ω at a 55 wt % SiC content. The result has proven that the SiC/LLDPE composites still possesses rather good electrical insulation properties.

Mechanical properties

Table IV summarizes the tensile properties of LLDPE and its composites at various SiC contents. It

TABLE IV
Mechanical Properties of SiC/LLDPE Composites

SiC content/wt %	Elongation at break/%		Stress at break/MPa	
	Siliane treated	Without treated	Siliane treated	Without treated
0	680 ± 13.0	664 ± 18.0	26.6 ± 1.0	25.2 ± 1.3
5	620 ± 14.2	603 ± 11.0	24.7 ± 0.7	22.7 ± 0.9
10	581 ± 16.0	557 ± 17.3	21.5 ± 0.9	20.1 ± 0.7
15	520 ± 10.7	487 ± 13.1	19.5 ± 0.3	17.5 ± 0.8
20	439 ± 15.4	403 ± 12.4	15.8 ± 0.5	13.8 ± 1.0
30	396 ± 13.5	363 ± 9.9	13.3 ± 0.7	11.5 ± 0.3
40	363 ± 11.6	331 ± 10.3	11.3 ± 0.3	9.8 ± 0.5
55	295 ± 9.8	287 ± 8.2	10.2 ± 0.5	8.7 ± 0.6

can be observed that pure LLDPE has higher values of stress and elongation at break, as compared to its SiC composites. Adding SiC filler to the pure LLDPE matrix results in a obvious decrease in both elongation at break and stress at break.

LLDPE has a high elongation at break, since when external force is applied, the LLDPE chains have enough space and time to orientate, which is the result of the chemical structure of LLDPE, when the chains are orientated, they start to from orientation crystallinity, which evokes an increase in the strength of a LLDPE sample. Incorporating SiC particles to the LLDPE matrix reduces chains mobility which leads to a rapidly decreasing elongation at break,[12] and introduces discontinuity in stress transfer to the filler-polymer interface in the composites structure. Thus, with increasing SiC filler content, both stress and elongation at break decrease obviously.[3]

Hybrid SiC/Al_2O_{3f} /LLDPE composites

From Table IV we know that the stress and elongation at break decrease remarkably at high SiC filler content. In addition, there are practical limits in loading high fraction of SiC filler particles to improve thermal conductivity, because high filler loading often causes problems in terms of processing as well as mechanical performance.[36] To improve the tensile properties of the SiC/LLDPE composites while maintain its thermal conductivity less changed, in the present investigation, an Al_2O_3 short fiber was introduced to the SiC particles as hybrid fillers to reinforce LLDPE (the weight ratio of Al_2O_3 short fiber to SiC particles is 1 : 5). The physical properties of LLDPE filled with 55 wt % total hybrid Al_2O_3 short fiber-SiC filler are summarized in Table V, and the SEM of fracture surface of Al_2O_3 short fiber-SiC/LLDPE as seen in Figure 8.

It is seen from Table V that the hybrid fillers filled LLDPE composite exhibits higher tensile properties than those of the SiC alone reinforced one; while the thermal conductivity of the former is nearly same as that of the latter. As compared to SiC particles, the Al_2O_3 short fiber has significant reinforcing effect, thus improving the tensile property of the resulting composites. Considering a relatively low thermal conductivity of the Al_2O_3 fiber compared to the SiC particles, the obtained slightly higher thermal conductivity of the samples containing hybrid fillers successfully demonstrates the usefulness of the concept of synergistic effect of hybrid filler in enhancing the thermal conductivity of the resulting composites for given filler loading,[37] since the fillers with a large

TABLE V
Properties of the LLDPE Composites Containing 55 wt % Filler

Properties	SiC	Al_2O_3 short fiber/SiC
Thermal conductivity/W/m K	1.48 ± 0.03	1.52 ± 0.02
Dielectric constant (1MHz)	8.3 ± 0.4	7.4 ± 0.3
Dielectric loss (1MHz)	0.01 ± 0.003	0.01 ± 0.005
Surface resistivity/Ω	3.5×10^{11}	9.1×10^{11}
Volume Resistivity/Ω cm	$7.2 \pm 0.5 \times 10^{13}$	$4.3 \pm 0.3 \times 10^{14}$
Tensile strength/MPa	10.2 ± 0.8	19.5 ± 1.1
Elongation at Break/%	295 ±13	385 ± 10

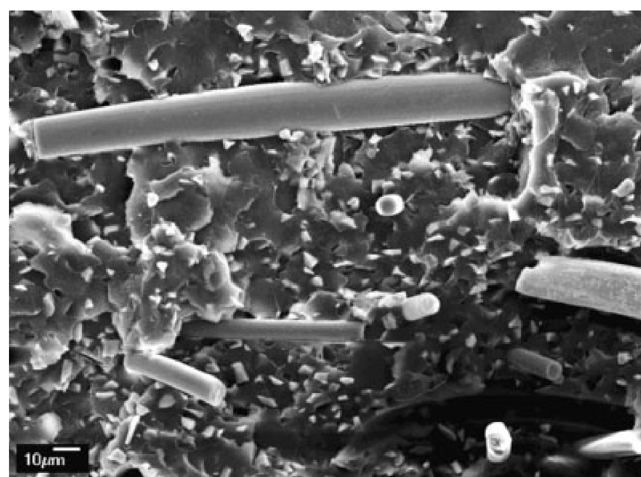

Figure 8 SEM micrograph of across-section of LLDPE filled with 55 wt.% of hybrid SiC/Al_2O_{3f}.

aspect ratio easily form the bridges between them, known as conductive network, the aspect ratio of the filler is more considerable that dictates the thermal conductivity of composites. The formation of random bridges or networks from conductive particles facilitates phonon transfer, which leads to high conductivity.

CONCLUSIONS

A thermal conductive composite plastic was fabricated from a particle mixture of LLDPE and SiC using heat press molding.

The SiC filler decreases the degree of crystallinity of LLDPE, and has no significant influence on the melting temperature of LLDPE. From SEM microimages the SiC particles distributions were found to be rather uniform at both low and high filler content owing to the powder mixing process employed.

The thermal stability and thermal conductivity of the LLDPE composites rised as SiC content, and the thermal conductivity reached 1.48 Wm^{-1} K^{-1} at 55 wt % SiC content. Moreover, the surface treatment of SiC with titanate coupler facilitated improving the interfacial bonding between SiC and matrix, and reduced the voids at the filler-matrix interface, thus enhancing the thermal conductivity.

The dielectric constant and loss rose with an increase of SiC content, however, they still remain at relatively low levels ($<10^2$ Hz). The SiC filled composites show poorer mechanical properties as compared to unfilled LLDPE.

The combined use of SiC particles and small amount of Al_2O_3 short fiber provides the LLDPE composites with improved tensile properties and thermal conductivity, as well as other properties, as compared to SiC particles used alone due to the synergistic effect of hybrid fillers for given filler loading.

Dr. Qunli An from China Northwestern Polytechnical University is recognized for performing the thermal conductivity and tensile properties measurements on our samples.

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