# Flame Retardancy, Thermal, Rheological, and Mechanical Properties of Polycarbonate/Polysilsesquioxane System

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**ABSTRACT:** The polymethylphenylsilsesquioxane (PMPSQ) microspheres were used as an additive for the injection-molding of polycarbonate (PC) in order to obtain a flame retardant polymer and to damage the other properties of PC as low as possible. The results showed that PMPSQ microspheres were a kind of effective flame retardant by the LOI test and the flame retardancy of PC increased with the increase of PMPSQ content. Thermogravimetric analysis suggested that the PMPSQ could induce cross-linking reactions and accelerate the thermal degradation of PC, sequentially promote the formation of the residue char during degradation. Differential scanning calorimetry measurement showed that the glass transition temperature of PC/PMPSQ system was decreased with the increase of PMPSQ content, which maybe caused by the

## **INTRODUCTION**

Polycarbonate (PC) has excellent dimensional stability, heat resistance, impact strength, transparency, and electrical properties and is commonly used in various fields of automobile, office machinery, electron and electric equipment, architecture, and the like.<sup>1,2</sup> However, many applications require that a PC composition be flame retardant and combine ease of processing with good impact strength. To improve this, effort has been made to develop flame retardant and other plastic additives. The addition of bromine based flame retardants is a common way in thermoplastic polymer to improve the flame retardancy, but it can lead to the formation of potential toxic brominated dibenzodioxins and -furans during thermal degradation.<sup>3,4</sup> While attempts have begun to replace these halogen compounds with phosphorus compounds and metal hydrates.<sup>5,6</sup> Although various studies were carried out on them, they haven't been proved as suitable flame retardants used in industrial application.<sup>7</sup>

In response to this problem, the use of silicone derivatives as flame retardants is researched because of their high heat resistance and nontoxicity during enlarged free volume of PC-rich phase. The rheological behaviors including dynamic and steady modes, combined with the analysis of scanning electron microscopy photographs, exhibited that the dispersion and compatibility of PMPSQ in the PC matrix decreased the viscosity of the systems and the addition of PMPSQ microspheres improved the processability of PC, but still there exist aggregation of PMPSQ microspheres in PC matrix, which gave a higher impact strength but a lower flexural modulus and elongation at break than pure PC. © 2009 Wiley Periodicals, Inc. J Appl Polym Sci 115: 330–337, 2010

**Key words:** polycarbonate; polymethylphenylsilsesquioxane microspheres; flame retardancy; thermal property; rheological property; mechanical property

combustion.<sup>8</sup> Previous researches using silicone derivatives as flame retardants include investigations of polymethylphenylsilsesquioxane (PMPSQ) and polydimethylsiloxanes mixed with PC, PS, PMMA, and other plastics.<sup>9–12</sup> Recent studies on POSS have been reported indicating reinforced mechanical and thermal properties of blends of silicone derivatives and PC,<sup>13,14</sup> rarely there is systematical study on the influence of the PMPSQ contents on the comprehensive properties of PC.

Polymethylphenylsilsesquioxane (PMPSQ) was found as an effective flame retardant for polycarbonate (PC) in the previous research. Meanwhile, silica and polyhedral oligomeric silsesquioxane (POSS) also have been reported indicating reinforced mechanical and rheological properties of PC, but there is rarely systematical study on the influence of PMPSQ on the comprehensive properties of PC.

In this article, the PMPSQ microspheres with a branch structure were prepared and used as a flame retardant for PC. Moreover, its flame retardant property and mechanism are investigated by the LOI and TGA measurement. The influence of PMPSQ content on the important characteristics of PC such as rheological, thermal, mechanical properties, and dispersion state of microspheres in polymer matrix has been examined in details. Some interesting phenomenon was found and explained, such as in measurement of rheological and thermal properties.

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### **EXPERIMENTAL**

#### Materials

The PC used was obtained from SABIC Innovative Plastics (Lexan-141). The PMPSQ microspheres consisting of 82 mol % phenyltrimethoxysilane and 18 mol % methylphenyldimethoxysilane units prepared by hydrolysis and condensation reaction according to Ref. 15. The molecular weight and melt point of PMPSQ microspheres were 3.774e+4g/mol and 70°C, respectively. It has been confirmed that the end groups of PMPSQ were mainly methyls and phenyls with a few hydroxyls by the NMR spectrum, FTIR, and TGA methods. The ratio of organic groups to silicon atoms (R/Si), which were used to indicate the branched extent of a polysilsesquioxane structure was 1.18.

## Preparation

The PC pellets and PMPSQ were mixed in a SHR100 high speed mixer for 5 min and the content of PMPSQ was kept from 0 to 7.0 wt %. Then the mixture was extruded by a MAPRE 2.83.0 41 twin screw extruder through a strand die and palletized, where cylinder temperature was controlled at 290°C and screw speed was 120 rpm. The resulting pellets were dried at 120°C for 12 h, and then molded into test specimens at an injection temperature of 310°C by a CJ-80E injection-molding machine.

## Characterization

#### Limiting oxygen index

The limiting oxygen index (LOI) is defined as the minimum oxygen concentration required for sustaining the downward flame combustion of materials. LOI was measured on an oxygen index instrument JF-3 produced by Jiangning Analysis Instrument Factory and performed according to GB2406-93.

#### Thermogravimetric analysis

Thermogravimetric analysis (TGA) was carried out using a TA Q600 thermal analyzer. Around 10 mg sample was heated from  $50^{\circ}$ C to  $800^{\circ}$ C at a heating rate of  $10^{\circ}$ C/min with a nitrogen flow of 50 mL/min.

## Differential scanning calorimetry

Glass transition temperature  $(T_g)$  was examined using a Perkin Elmer Diamond Differential scanning calorimetry (DSC). The samples of about 4 mg were heated from 120°C to 180°C at a heating rate of  $10^{\circ}$ C/min under a nitrogen atmosphere. Fitting the straight lines to the DSC curve before, during, and after the transition, and taking the points of intersection as the onset and endpoint of the transition. The  $T_g$  was then located at one-half the change in heat capacity between the onset and endpoint.

### Rheological measurements

Dynamic frequency sweep test and steady state shear measurements were performed using an Advanced Rheometric Expansion System in the parallel plate mode for all rheological measurements. The linear viscoelastic range was determined from strain sweep measurements as 5-20% strain, depending on the concentration of sample. The frequency sweep tests were conducted with the frequency varied to ensure the linear viscoelastic region; we set the strain to be 1.0%, and then apparent viscosity, elastic modulus (G'), loss modulus (G"), and loss tangent (tan  $\delta$ ) were obtained. The dynamic measurements were performed at different temperatures (260, 280, and 300°C) and frequency were performed over a range of 1-500 rad/s, whereas the steady state shear measurements were carried out in the shear rate range from 0.01 to  $10 \text{ s}^{-1}$ . The temperature was maintained constant during the frequency sweep.

#### Mechanical properties

The mechanical properties were measured by a universal testing machine (DXLL-20000) according to ASTM test methods, such as D-638 for the tensile strength and D-790 for the flexural modulus. The Izod impact strength was tested on the basis of ASTM D-256.

## GPC measurements

Solutions for GPC characterization were prepared by dissolving products into THF to 0.3 wt %. Polysty-rene standards with molecular mass of 3.15e+6 were measured at the beginning to calibrate the differential refractometer detector.

#### Scanning electron microscopy

The phase structures of PC were observed on a JEOL JSM-6360LV scanning electron microscopy (SEM) operating at acceleration voltage of 15 kV. The sample was immersed into liquid nitrogen, so that its surface was not affected by any external stresses. Then, fractured and coated with gold to enhance conductivity.

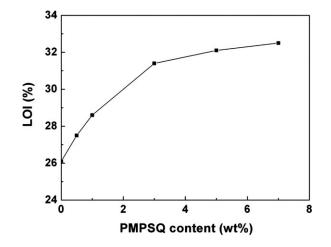


Figure 1 Effect of PMPSQ content on limiting oxygen index of PC.

# **RESULTS AND DISCUSSION**

#### Flame retardancy

The LOI test is widely used to evaluate the flame retardancy of materials and especially for flame retardant polymer formulations.<sup>10,16</sup> The influence of the PMPSQ content on the LOI value of PC is shown in Figure 1. The LOI values of PC increase with the increasing of PMPSQ content, but there is a nonlinear relationship between LOI value of PC and the content of PMPSQ. The LOI value increases quickly with the increase of PMPSQ content when the PMPSQ content is below 3 wt %. On the other hand, while the content of PMPSQ exceeds 3 wt %. the LOI value of PC increases slowly. It can be seen that the LOI value of PC is enhanced upto 31.4 from 26.1 when the content of PMPSQ reaches 3 wt %, whereas the corresponding value is 32.5 at 7 wt %.

It was reported that a special silicone with a branched structure, a phenyl-rich mixture of phenyl and methyl groups in the chain and methyl groups at the chain terminals to be effective in retarding the combustion of PC. The superior flame retardant effect owes to its excellent dispersion in PC and its rapid movement to the surface of PC to form the highly flame resistant char barrier during combustion. It was suggested that the formation of the crosslinking structures between the PC substrates and the phenyl silicone additives might play an important role in the flame retarding mechanism of the

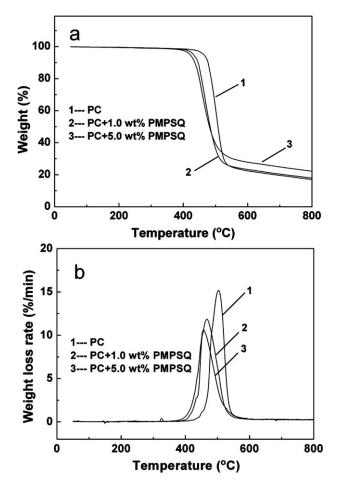


Figure 2 TGA (a) and DTG (b) curves for PC and PC/ PMPSQ systems in  $N_2$ .

PC with a phenyl-rich silicone additive.<sup>9</sup> On the other hand, higher loading of PMPSQ causes LOI to decrease, mainly caused by the presence of silanols on the PMPSQ structure. Actually, small amounts of acid or base groups can catalyze decomposition of PC at elevated temperatures, such that the material becomes less thermally stable or starts to lose flammability performance.

#### Thermal properties

The TGA and DTG curves for PC and PC/PMPSQ systems observed in a nitrogen flow are given in Figure 2, and the characteristic mass loss data are listed in Table I. It can be seen that the temperature

 TABLE I

 Resulting Data of Thermogravimetric Analysis of PC and PC/PMPSQ Systems

PMPSQ content (wt %)	Temperature (°C)				Peak rate	Residues
	$T_{1\%}$	$T_{5\%}$	$T_{50\%}$	$T_{\rm max}$	(%/min)	(wt %)
0	316	464	507	503	15.2	17.0
1	311	432	483	467	11.9	18.0
5	288	421	481	456	10.6	22.2

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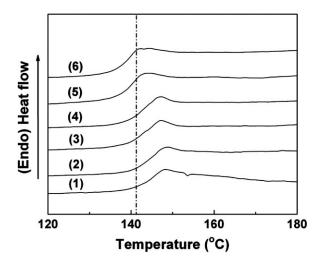


Figure 3 Effect of PMPSQ contents on glass translation temperature of PC: (1) 0, (2) 0.5 wt %, (3) 1.0 wt %, (4) 3.0 wt %, (5) 5.0 wt %, and (6) 7.0 wt %.

of 1 wt % mass loss for pure PC is 316°C and 288°C for the 5.0 wt % PC/PMPSQ system, which indicates that the mass loss onset temperatures of PC thermal degradation tend to decrease with the increase of PMPSQ content. The initial degradation stage of the PC/PMPSQ system occurs before the major degradation step is due to the decomposition of PMPSQ. As the end groups of PMPSQ to be used contain a few residual silanols, these residual end groups in polymer chain induce condensation reaction from chain ends, which are a low activation energy process.<sup>17</sup>

PC and PC/PMPSQ systems all exhibit one step degradation process and the major degradation process of PC occurs between 400 and 550°C. As evidenced from the curves, PMPSQ causes thermal destabilization and the peak rate of PC mass loss curves decreases as the amount of PMPSQ increases. Meanwhile, PMPSQ widens the temperature range of PC thermal degradation and the temperature of mass loss peak rate  $(T_{max})$  decreases with the rising of PMPSQ content. It has been confirmed that in the major degradation step, Si-O bond and Si-Ph or Si-CH<sub>3</sub> bond of PMPSQ tend to form some silvl radicals or siloxane derivatives, which can react with PC or the evolved products of PC and correspondently promote crosslinking reactions in the PC/ PMPSQ system and retard further pyrolysis during the thermal degradation process.<sup>18</sup>

After the major degradation of the PC/PMPSQ, a much slower degradation process is followed and the degradation residues of the PC/PMPSQ systems at 800°C in N<sub>2</sub> are higher than that of pure PC. With the content of PMPSQ increasing, the amount of solid residues shifts from 17.0 wt % (for PC) to 22.2 wt % (for 5 wt % PC/PMPSQ) of the initial mass. This result further confirms that the branched PMPSQ with methyl and phenyl can induce cross-

linking reactions and promote the formation of the char during the thermal degradation process, which may play an important role for the flame retardancy of PC/PMPSQ systems.

PC do not possess a unique melting point, as exhibited by typical crystalline polymers but have a high glass transition temperature ( $T_g$ ) between 140 and 150°C. The melting temperature of PMPSQ microspheres is below 200°C. Thus during processing with PC, PMPSQ would be expected to be in a molten state, improving the mixing efficiency and minimizing its domain sizes within the PC matrix.

The DSC curves for PC and PC/PMPSQ systems are given in Figure 3. It is of interest to point out that a glass transition is identified for these composites with 0–7.0 wt % PMPSQ content and the decreasing of  $T_g$  values is observed with the increase of the PMPSQ content. The  $T_g$  value of 5 wt % PC/PMPSQ system is 138.2°C, whereas the value of pure PC is 144.5°C. These phenomenon observed clearly suggest that the PMPSQ additive is well dispersed within the PC matrix. The  $T_g$  of PC/PMPSQ systems are lower than that of pure PC because of

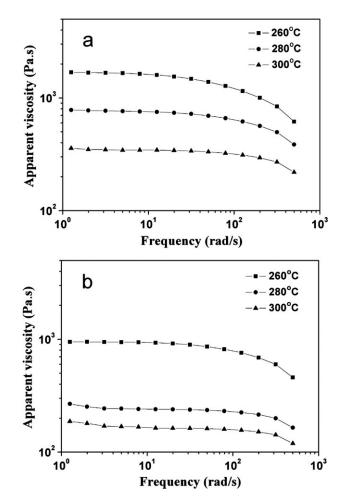
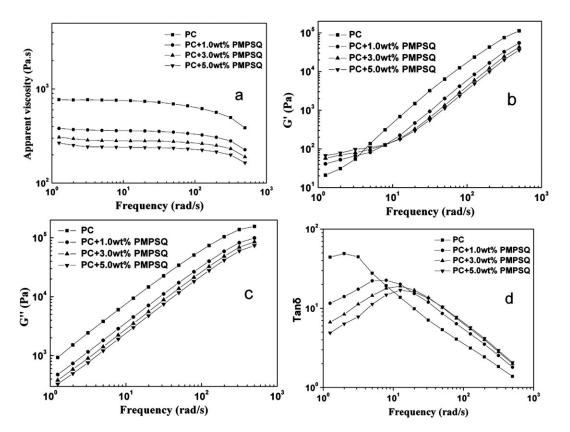


Figure 4 Rheological behaviors of PC (a) and 5 wt % PC/PMPSQ (b) at various temperatures.

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**Figure 5** Effect of PMPSQ content on dynamic rheological behaviors of PC at 280°C. (a) Apparent viscosity; (b) G'; (c) G''; (d) Tan  $\delta$ .

the enlarged free volume of the PC-rich phase, in which some PMPSQ molecules access into the void space of PC structure.<sup>19</sup> This is beautifully consistent with the analyzes result of LOI measurement.

#### **Rheological properties**

According to the previous studies, it is rather difficult to process PC, as its high melt viscosity hinders the fluidity and the residual stress resulting from the process could cause fractures.<sup>20</sup> Silicone derivatives are known to decrease the sheer viscosity of blend and used as an adjusting agent for the processability of polymers.<sup>21</sup> Thus, the rheological properties of PC modified by a mount of PMPSQ microspheres are examined, here.

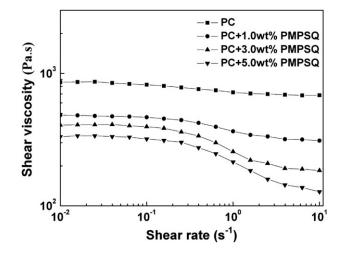
The rheological behaviors of pure PC and 5 wt % PC/PMPSQ systems as a function of frequency at temperatures of 260, 280, and 300°C are shown in Figure 4. It can be found that the apparent viscosity curves of pure PC exhibit weak pseudoplastic flow behavior; the apparent viscosity decreases slightly with increasing frequency and we can also understand that the degree of shear thinning reduces with the increase in temperature, due to the increase in mobility and fast relaxation of each component. Generally, PC falls within the category of rigid polymer owing to the numerous aryl groups in the main

chains and this leads to the high flow activation energy of PC melt; moreover, the melt viscosity is more sensitive to the variation of temperature.<sup>22</sup> Through observing the rheology curves in Figure 4(a,b), it is proved that the increasing of temperature from 260°C to 300°C both exhibit a strong influence on the rheology enhancement of PC melt, indicating that the rheological behaviors of PC melt can be regulated by the enhancing of processing temperature.

Figure 5 illustrates the influence of PMPSQ content on the dynamic rheological behaviors of PC at 280°C. Apparent viscosity versus frequency for the various blend ratios of PMPSQ and PC has been investigated in Figure 5(a). It can be seen that the apparent viscosity decreases as the frequency increases, exhibiting the characteristic pseudoplastic flow behavior of polymer systems over the entire range of frequency, for all various PMPSQ contents. The apparent viscosity also decreases sharply with the PMPSQ loading, revealing that the rheological behavior of PC melt can be regulated by the addition of PMPSQ microspheres. This maybe because the apparent viscosity of PMPSQ is much lower than that of pure PC, and the rigid chain of PC molecular can be modified by the flexible chain of PMPSQ molecular.<sup>23,24</sup> In other words, only 1 wt % of PMPSQ content in the system can alter the overall flow properties of PC and the resultant values of apparent viscosities in the system are remarkable lower than that of pure PC case. The addition of PMPSQ microspheres certainly endows PC melt with rheology enhancement. As the content of PMPSQ is increased, the apparent viscosity curves of PC systems with different content of PMPSQ approach to each other.

The storage modulus (G') and the loss modulus (G") are two important parameters in characterizing the viscoelasticity of molten polymer under oscillatory shear and G' represents the strain energy reversibly stored in the substance, whereas G'' represents the amount of energy irreversibly given off by the substance to its environment.<sup>25</sup> Figure 5(b,c) show the dependence of G' and G'' on frequency for PC systems with different PMPSQ content. It can be found that the G'' values of the PC/PMPSQ systems are lower than that of the pure PC over the entire range of frequency. They decrease when the dispersed phase (PMPSQ) content increases, which is in accordance with the trend of apparent viscosity curves. But the trend of G' appears in another way, where the values of PC/PMPSQ systems are lower than that of the pure PC at high frequencies ( $\omega > 10$ rad/s), and they also decrease with the increase of PMPSQ content. In low frequencies, it can be obviously seen that the G' values of PC/PMPSQ systems are greater than that of pure PC.

The loss tangent (tan  $\delta$ ) is a function of the ratio of the loss to the storage modulus (tan  $\delta = G''/G'$ ) and the relation between loss tangent and frequency for PC systems with different PMPSQ content at 280°C is shown in Figure 5(d). The tan  $\delta$  values of all systems are smaller than that of pure PC at low frequencies, whereas they converge at high frequencies, as illustrated in Figure 5(d). This may be a consequence of some solubility, graft, and crosslink reactions in the PC matrix, which can lead to the storage of more elastic deformation energy under oscillatory shear.<sup>26</sup> For PC systems with 1 wt %, 3 wt %, 5 wt % PMPSQ, a peak appears in the low frequency region; this peak decreases and shifts toward the higher frequency region with the increasing of PMPSQ content, then the PC/PMPSQ systems possess higher values of tan  $\delta$  than pure PC. Besides, even when the PMPSQ content in PC reaches 5 wt % the corresponding peak appears at 12.56 rad/s, and is higher than 1.99 rad/s for pure PC. Generally, the absence of tan  $\delta$  peak at lower frequency region is responsible for the existence of disorder or homogeneous structure in the filled polymer system. In other words, the appearance of tan  $\delta$  peak in the lower frequency region, together with the decreasing of tan  $\delta$  peak and shifting of tan  $\delta$  peak toward higher frequency region with the increase of PMPSQ content, have been attributed to the formation of het-



**Figure 6** Flow curves of various PC/PMPSQ systems at 280°C.

erogeneous structure/morphology.<sup>27</sup> Hence, the above phenomenon is considered to be involved in the higher level of PMPSQ aggregation in PC at 280°C, which is in accord with the results showed in Figure 5(b). On the other hand, the decrement of tan  $\delta$  with rising PMPSQ content in PC under slight frequency indicates that the decrease of energy dissipation for the PC/PMPSQ system implies the retardation of the relaxation of polymer chain by PMPSQ.

Figure 6 shows the relationship between the shear viscosity and shear rate of PC with various contents of PMPSQ microspheres in steady mode, and the shear viscosity can be used as a measure of the processability of polymer. It is found that the shear viscosity decreases remarkably with the increase in PMPSQ content at a very high shear rate of  $10s^{-1}$ . As the shear rate is decreased, the values of various PC added with different content of PMPSQ approach to each other, and the PC with high PMPSQ content still have high viscosity. Furthermore, the PC with higher PMPSQ content shows more obvious shear thinning behavior, indicating high content of PMPSQ is needed for PC to improve its processing property when applied to a small stress.

#### Mechanical properties

The mechanical properties of PC/PMPSQ systems are showed in Figure 7 as a function of PMPSQ content, which indicate that the influence of PMPSQ on mechanical properties of PC is remarkable in the range of PMPSQ contents studied.

It can be observed from Figure 7(a) that the addition of PMPSQ microspheres can improve the impact strength of PC significantly. The impact strength rises with the increasing of PMPSQ content and when the content of PMPSQ microspheres

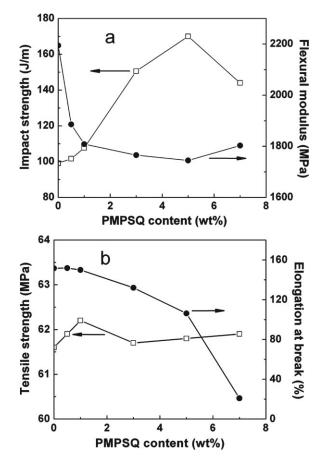


Figure 7 Effect of PMPSQ contents on mechanical properties of PC.

reaches 5.0 wt %, the impact strength value of PC is maximum (170.0 J/m). After the PMPSQ content increases more than 5.0 wt %, the impact strength decreases to 144.0 J/m at 7.0 wt % PMPSQ, which means that the addition of PMPSQ into PC no longer cause an impact strength increase of PC when the content of PMPSQ microspheres exceeds 5 wt %. To be contrary, the flexural modules of PC decrease with the PMPSQ content increasing. While the content of PMPSQ reaches 5.0 wt %, the flexural modules arrives at minimum value 1745 MPa. Subsequently, the flexural modules of PC slightly increase with the enhancing of PMPSQ content. The elongation at break of the PC/PMPSQ systems decreases with the increasing of PMPSQ content and when the PMPSQ content is 5 wt %. the value of elongation at break decreases from 151.6% to 106.2%. However, the tensile strength of PC isn't affected by the change of PMPSQ content, as exhibited in Figure 7(b). Based on this, the PMPSQ also may be having some plasticization effects, as evidenced by the decrease in  $T_g$ , loss of stiffness, and increase in impact strength as PMPSQ loading is increased.

The FTIR test of PC and PC/PMPSQ samples was carried out to see if any reactions have occurred. Unfortunately, there was rarely no difference between the two samples. Nevertheless, the molecular weight measurement of PC and PC/PMPSQ may provide some better insight into their results, which can be used to explain the above phenomenon further. The  $M_w$  of PC and PC/PMPSQ is 1.94e+4 and 1.44e+4, respectively. Thus, it can be concluded the addition of PMPSQ accelerated the decomposition of PC, which caused by the presence of silanols on this material.

# Morphology

As previously referred, with the content of PMPSQ rising, the impact strength of PC/PMPSQ system shows an obvious improvement. Nevertheless, the flexural modules and rupture elongation decline slightly compared with pure PC, while the content of PMPSQ microspheres is less than 5 wt %. This trend might be critically dependent on the dispersion and compatibility of PMPSQ microspheres in PC matrix, which would be expected to be understood. Figure 8 shows the SEM photographs of the pure PC and 5.0 wt % PC/PMPSQ systems.

It can be observed from the Figure 8(b,c) that the residual PMPSQ microspheres in the continuous PC matrix can't keep spherical shape and emerges with obvious distortion. The size of residual microspheres can be easily found in SEM photographs is about 1  $\mu$ m less than their primary diameter ( $\approx 2 \mu$ m). These morphology results are consistent with result

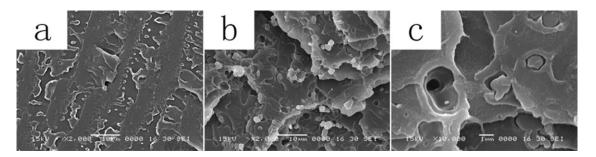


Figure 8 SEM photographs of systems: (a) PC  $\times$  2000; (b) PC/PMPSQ  $\times$  2000; (c) PC/PMPSQ  $\times$  10000.

of LOI and DSC measurement. The PC/PMPSQ systems were obtained through injection molding process and the shape of PMPSQ microspheres was changed under the high speed shear. As a result, some of PMPSQ molecular chains leave the microsphere surface and access into the void space of PC structure. In that case, the LOI increment and  $T_g$ decrement appeared. Meanwhile, some of distorted PMPSQ microspheres remained and the somewhere aggregation in PC matrix emerged, which is corresponding with the analysis results of Figure 5(d).

A morphology study on materials offers not only dispersion information, but also estimation on the interfacial interaction between additive and matrix. As exhibited in Figure 8(c), the interface between the PMPSQ phase and continuous PC phase can be found and the interfacial separation appears. Although two-phase morphology can be observed clearly from the magnified photomicrograph, there is still no credible evidence showing that a grafting or crosslinking reaction at the interphase is carried out between PMPSQ and matrix PC. It is presumed that the phase separation in our system may be due to the different contraction rate of PC and PMPSQ microsphere in the cooling of injection molding process. Meanwhile, the interface of pure PC is shown in Figure 8(a), where the fractured surface is smooth and no morphology of phase separation is observed except cavitation and fibrillation. Therefore, it is believed that the dispersion of PMPSQ microspheres in PC matrix is a main cause for the decrement of flexural modules and elongation at break of PC/ PMPSQ systems.

# CONCLUSIONS

The effect of PMPSQ microspheres used as a flame retardant for PC was investigated by LOI test and the flame retardancy of PC increased with the rising of PMPSQ content. When the content of PMPSQ reached 7 wt %, the LOI value of PC was enhanced upto 32.5 from 26.1. TGA measurement in nitrogen exhibited that the PMPSQ microspheres could induce crosslinking reactions and accelerated the thermal degradation of PC, sequentially promoting the formation of the residue char during the flame retardant process. The DSC result showed that the glass transition temperature ( $T_g$ ) of PC/PMPSQ system was decreased with the increase of PMPSQ content, which maybe caused by the enlarged free volume of the PC-rich phase by the influence of

PMPSQ molecular chain. The rheological measurements (including dynamic and steady modes) results indicated that the dispersion and compatibility of PMPSQ in PC matrix decreased the viscosity of the systems; hence, the addition of PMPSQ could improve the processability of PC. Meanwhile, PMPSQ microspheres caused a significant increase of impact strength of PC and when the PMPSQ content reached 5.0 wt %, the impact strength of PC arrived at maximum value of 170.0 J/m from 99.1 J/ m (pure PC). The aggregation of PMPSQ microspheres in PC matrix was observed from the SEM photographs, which was corresponding with the decreasing of flexural modulus and elongation at break than pure PC.

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