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Thermal and mechanical properties of polyhedral oligomeric silsesquioxane (POSS)/polycarbonate composites

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Abstract

A series of composite materials were produced incorporating polyhedral oligomeric silsesquioxane (POSS) derivatives into polycarbonate (PC), by melt blending. Significant differences in compatibility were observed depending on the nano-scale filler's specific structure: trisilanol POSS molecules generally provided better compatibility with PC than fully-saturated cage structures, and phenyl-substituted POSS grades were shown to be more compatible with PC than fillers with other functional groups. Trisilanolphenyl-POSS/PC composites possess the best overall performance among the POSS materials tested. The high compatibility between the trisilanolphenyl-POSS and polycarbonate matrix results in generation of transparent samples up to 5 wt% POSS content. Slightly enhanced mechanical properties including tensile and dynamic mechanical modulus are observed with the increase of trisilanolphenyl-POSS loading at the cost of decreasing ductility of the nanocomposites. Importantly, upon orientation of the PC/POSS nanocomposite, crystallization of POSS within the oriented material results—this observation is consistent with a growing number of observations which suggest that 'bottom-up' formation of structures incorporating multiple POSS cages result from orientation of these nanocomposites, and that the hybrid organic–inorganic inclusions may be at the heart of observed nano-scale reinforcement.

Keywords: Nanocomposite; Blend; Polycarbonate

1. Introduction

Polyhedral oligomeric silsesquioxane (POSS™) molecules possess a cage-like structure (1–3 nm in size) and a hybrid chemical composition ($RSiO_{1.5}$) which is intermediate between silica (SiO_2) and silicones (R_2SiO) [1]. Their well defined structure contains a stable inorganic Si–O core surrounded by substituents which can be modified to present a wide range of polarities and reactivities. POSS molecules can be incorporated into polymer systems through blending [2], grafting or co-polymerization [3–9] aiming at nanostructured polymeric materials whose properties bridge the property space between organic plastics and ceramics.

Recent studies on POSS-containing hybrid copolymers and thermosets have been reported indicating reinforced mechanical [6–8] and thermal properties [7,9]. The present study focuses on the thermal and mechanical properties of blends of POSS derivatives and polycarbonate (PC). Here POSS is expected to act as a nano-scale filler to modify the polycarbonate matrix and potentially result in nanocomposites with new or improved properties. Compared with traditional fillers, POSS is known in some cases to exhibit enhanced compatibility resulting from a match between its organic substituents and the matrix polymer. In this manner, control of the microstructure of the POSS nanocomposites can be achieved and is key to the performance and properties of such materials [3,9]. It is generally believed that nanoscale POSS domains with ordered and self-assembled features in a polymer matrix are highly desirable and lead to the observed improvement in material properties [10–13].

The current work focuses on the compatibility of different five different POSS structures (Scheme 1) with a polycarbonate matrix and the effect of POSS concentration on the thermal and mechanical properties of POSS/PC nanocomposites. Differences between composites containing fully-condensed POSS cages, and those possessing a hybrid inorganic–organic 3D partial cage-like structure bearing three silanol (Si–OH) groups (Scheme 1(b)) will be examined. It was recently reported that the trisilanol POSS grades, especially isooctyl trisilanol POSS, exhibit better compatibility with PET, as judged by single phase melts and optically transparent extrudate [14].

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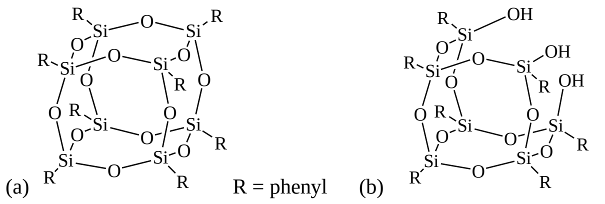


Scheme 1. Typical chemical structures of POSS molecules: (a) octaphenyl-POSS (MS0840); (b) trisilanolphenyl-POSS (SO1458) when R = phenyl, and trisilanolisooctyl-POSS (SO1455) when R = *i*-octyl.

Additionally, the properties of a blend of trisilanolphenyl-POSS with a polycarbonate/polysulfone polymer blend (HTLT Z-1050) was briefly investigated in this study.

2. Experimental section

2.1. Materials

Dow CALIBRE® 200-14 and BASF Makrolon 2405 Polycarbonate grades were used as received. POSS derivatives were supplied by Hybrid Plastics, Inc. and used as received. A polycarbonate–polysulfone thermoplastic polymer blend (HTLT Z-1050) was supplied by Suncolor Co. and used as received. Prior to compounding the polymer pellets were dried for at least 6 h under vacuum at 100 °C.

2.2. POSS/PC sample preparation

Polycarbonate was blended with different levels of POSS using a DACA model 20000 twin-screw co-rotating micro-compounding extruder (13.75 mm screw diameter/108 mm length). The blending was carried out at 290 °C and a screw speed of 100 rpm for 5 min prior to extrusion. During the extrusion process, the extruded samples were visually inspected for qualitative transparency. The samples were then compression molded into films (not constrained inside of a mold) for mechanical testing using a Carver model C press. The PC composites were first heated at 280 °C for 3–4 min and a rapid compression (5–6 ton), followed by release of pressure to remove any trapped gas bubbles. The samples were then molded at 280 °C under 6–7 ton pressure for 3–4 min and cooled rapidly between two water-chilled aluminum plates. The PC composite films (0.2–0.3 mm thickness) were obtained and evaluated for transparency. For the trisilanolphenyl-POSS/HTLT Z-1050 composite, the compounding was performed at 320 °C and 100 rpm screw speed/4 min prior to extrusion, and the compression molding was carried out under 6–7 ton pressure for 3–4 min at 310 °C.

2.3. Differential scanning calorimetry (DSC)

The thermal behaviors of the POSS/PC composites were measured using a TA Instruments DSC 2910 equipped with a pressure DSC Cell. Samples (4–8 mg) were tested at a heating rate of 10 °C/min and results from the second heating are reported. Thermal data analysis was performed using TA Instruments Universal Analysis Software.

2.4. Dynamic mechanical analysis

A Tritec 2000 DMA (Triton Technology Co.) was used in tensile mode at an oscillatory frequency of 1 Hz with applied 1% strain for all samples. The temperature scan was performed at 3 °C/min heating rate in the range from room temperature to around 180 °C for PC composites and 280 °C for HTLE Z-1050 composite. Sample dimensions were typically 4.9 mm long, 9.8 mm wide and 0.2–0.3 mm thick.

2.5. Tensile testing

Sample films were die cut into standard ASTM D 638V dog bone shaped test specimens with the geometry shown in Scheme 2. The microtensile tests were carried out on an Instron model 5565 at an extension rate of 2 mm/min at room temperature (~23 °C). Five specimens of each composite were tested and then averaged results and standard deviations were reported.

2.6. Thermal gravimetric analysis (TGA)

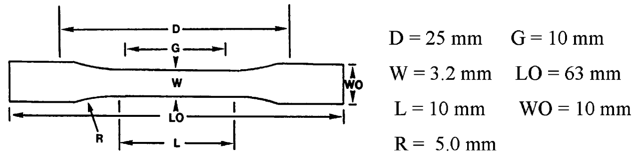
The thermal stability of the samples was measured by thermogravimetric analysis using a TA Instruments 2950 High Resolution Thermogravimetric Analyzer. Sample sizes ranged from 5 to 15 mg. Weight loss was traced as samples were heated at a rate of 10 °C/min from room temperature to 900 °C under a dry nitrogen purge of 90 ml/min.

2.7. Wide-angle X-ray diffraction (XRD)

The XRD measurements were carried out at room temperature on a Rigaku X-ray diffractometer in normal reflection mode with Ni-filtered Cu Kα radiation ($\lambda = 1.54$ Å). After the sample was loaded, the alignment procedure was followed to ensure the accuracy of the θ angle. For oriented samples, the sample specimens were rotated to obtain the diffraction profiles at parallel and perpendicular directions.

2.8. Izod impact tests

Two grades of PC, Bayer Makrolon 2405 and Dow Calibre 200-14, as well as two nanocomposites, PC/3 wt% PM 1271



Scheme 2. The test specimen for tensile measurement. $D = 25$ mm; $G = 10$ mm; $W = 3.2$ mm; LO = 63 mm; $L = 10$ mm; WO = 10 mm; $R = 5.0$ mm.

and PC/3 wt% SO1458 were scaled up for impact testing. The composition of SO1458 is trisilanolphenyl-POSS. And PM1271 is a partially polymerized trisilanolphenyl-POSS. The nanocomposites were produced by Hybrid Plastics, Inc. on a MP-2019TC twin-screw extruder (BPProcess; diameter = 19 mm; $L{:}D = 25{:}1$; 300–400 rpm) at 240–250 °C using the Makrolon grade as the base resin.

Samples of PC and PC/POSS were injection molded on a Boy 22-S machine into a two cavity mold containing an ASTM flex bar and a type V tensile bar. A barrel temperature profile of 280/305/315 °C (feed/mid/nozzle), mold temperature 100 °C, and total molding cycle time of 80 s was used. The flex bars were cut into halves using a band saw, and notched using a Tinius Olsen notcher. Both notched and un-notched samples were then tested using an izod impact tester (Tinius Olsen) using a 5 lb hammer; five samples of each material were tested.

3. Results and discussion

3.1. Optical clarity

The extrusion observations and sample appearances are summarized in Table 1. Trisilanolphenyl-POSS (SO1458)/PC composites generally exhibited transparency similar to that of control PC. A slight decrease in transparency was observed as POSS concentrations were increased from 2 to 10 wt% (Table 1). This implies that trisilanolphenyl-POSS is quite compatible with PC and dispersed domains should be very small (nanoscale), giving rise to the optical clarity. In contrast, the optical transparency of PC was significantly reduced for PC composites with increasing addition of octaphenyl-POSS (5–10 wt%), despite the structural similarities between trisilanolphenyl-POSS and the octaphenyl derivative (Table 1). The presence of the silanol groups (Si–OH) on POSS structures can be postulated to generate particle–polymer interactions (either polar or covalent), which contribute to this enhanced compatibility. Similar enhancements of compatibility between trisilanol POSS grades and polymers have been observed with PET [14] and with cellulose propionate [15].

As a viscous fluid with high viscosity, trisilanolisooctyl-POSS (SO1455) is different from the crystalline POSS materials examined in this study. The opaque extruded samples and translucent films for trisilanolisooctyl-POSS/PC composites indicate a modest level of compatibility between these two materials.

For Al-POSS/PC composites (one corner of the POSS cube substituted with an aluminum atom; Table 1), coloration generation was clearly observed at low filler concentrations (2.5 wt%): brown for Al-phenyl-POSS/PC and orange for Al-isobutyl-POSS/PC. This coloration may possibly result from an undefined reaction between the Al on the POSS cage and additives in the PC matrix. Alternatively, the aluminum-capped POSS itself may have degraded under processing conditions. The aluminum-capped POSS grades, which may in fact be dimeric in nature [16], were of interest because of the potential for coordination between Lewis acidic aluminum and the C carbonyl oxygens. While POSS grades are relatively stable up to 300 °C, they are known to degrade given sufficient time/temperature [17]. In addition to color generation, a significant number of bubbles were observed in the extruded samples of these two composites as well as in octaphenyl-POSS/PC composites; these were very difficult to remove during compression molding. The presence of bubbles within the composites also suggests chemical reactions may occur in within these systems. We note there that the 2.5 wt% Al-isobutyl-POSS/PC films were very brittle and therefore, were not subjected to any mechanical tests.

Table 1
Extrusion observation and film appearance of POSS/PC composites

POSS/PC samples	Extrusion observation	Optical clarity and observable properties of films through compression molding[a]
Polycarbonate control	Transparent	Transparent, tough
Trisilanolphenyl-POSS		
2.5 wt%	Transparent	Transparent, tough
5 wt%	Nearly Transparent	Transparent, tough
10 wt%	Translucent	Semi-transparent, tough
Trisilanolisooctyl-POSS		
2.5 wt%	Opaque	Translucent, tough
5 wt%	Opaque	Translucent, tough
10 wt%	Opaque	Somewhat translucent, tough, many bubbles
Octaphenyl-POSS		
2.5 wt%	Translucent, tiny particles visible in matrix	Nearly transparent, a few bubbles, somewhat brittle
5 wt%	White opaque	Translucent, many bubbles
10 wt%	White opaque, porous	Opaque, porous
Al-phenyl-POSS		
2.5 wt%	Brown translucent, small bubbles dispersed in matrix	Light brown, transparent, many bubbles
Al-isobutyl-POSS		
2.5 wt%	Orange semi-transparent, big bubbles dispersed in matrix	Nearly transparent, large amount of bubbles, very brittle film

[a] Film thickness is 0.2–0.3 mm.

Table 2
Extrusion observation and film appearance of POSS/HTLT Z-1050 composites

Samples	Extrusion observation	Optical clarity of films through compression molding[a]
HTLT Z-1050 control	Transparent	Transparent, tough
5 wt% trisilanolphenyl-POSS/HTLT Z-1050	Transparent	Transparent, tough

[a] Film thickness is 0.2–0.3 mm.

In Table 2, qualitative properties of the PC/polysulfone blend HTLT Z-1050, are presented. The incorporation of 5 wt% trisilanolphenyl-POSS into HTLT Z-1050 matrix results in high film transparency, similar to that obtained with PC itself. This again suggests a high level of compatibility between trisilanolphenyl-POSS and HTLT Z-1050.

3.2. Thermal and dynamic mechanical analysis

Trisilanolphenyl-POSS is white, crystalline powder at room temperature. The DSC scan of this pure POSS indicates the melting temperature at 208 °C. Thus during processing with PC, this POSS would be expected to be in a molten state, improving the mixing efficiency and minimizing its domain sizes within the nanocomposites. It is of interest to point out that no crystalline melting endothermic peaks were observed in the DSC curves of trisilanolphenyl-POSS/PC composites, and only a glass transition was identified for these composites with 2.5–10 wt% POSS composition; the composite T_g values were very close to that of the PC control, and decrease slightly with the increase of the POSS concentration (Fig. 1, Table 3). These observations clearly suggest that the POSS additive is well dispersed within the PC matrix leading to the suppression of crystallization of pure POSS. Another possibility here is that the crystallization of POSS in PC matrix does occur, however, the size of formed crystals is ultra-small, most likely in nanometer size and at very low overall content. Thus the DSC measurements might not be sensitive enough to show the melting peaks.

The trisilanolphenyl-POSS/PC composites were also evaluated by dynamic mechanical analysis (DMA; temperature scan at 1 Hz and 1% strain). The spectra of storage modulus E' and loss factor tan δ (Fig. 2) display only one α relaxation process corresponding to the glass transition of the composites with different POSS loadings, quite similar to the behavior of the PC control. The DMA glass transition temperatures (T_g) are listed in Table 3, which are consistent with the DSC results. A slightly higher T_g value of DMA than that of DSC in Table 3 is due to the frequency difference between these two measurements. We note the modulus $E^* = (E'^2 + E''^2)^{1/2}$ ($\sim$2 GPa) for the PC control is numerically consistent with the tensile modulus reported by the Dow Chemical Co. [18]. The plateau E' values (Table 3) before the glass transition slightly increase with the POSS loading and are a slightly larger than that of the PC control, suggesting that the POSS addition has little influence on the dynamic modulus. Nevertheless, the behavior of E' drop at glass transition demonstrates a small difference between the composites and PC control. In fact the peak values of tan δ in Fig. 2 show a decreasing trend from the PC control (1.94) to 2.5 wt% (1.85), 5 wt% (1.83) and 10 wt% (1.72) POSS content, consistent with a slight increase in the dynamic storage modulus with the increasing POSS concentration (Fig. 2 and Table 3). This behavior is consistent with the increase of the interfacial area between POSS and PC matrix due to the rise of the POSS concentration.

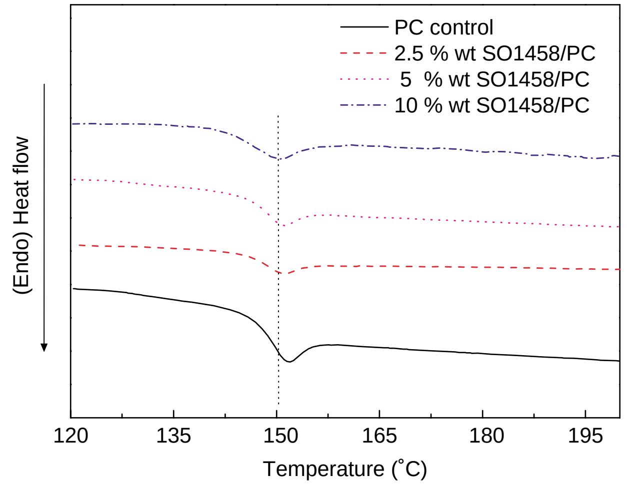


Fig. 1. DSC curves of trisilanolphenyl-POSS/polycarbonate composites.

Since trisilanolisooctyl-POSS (SO1455) is a viscous fluid, the T_g values for these composites are slightly lower than the unfilled PC (Fig. 3), as one might expect for non-molecular scale dispersions. This is particularly true at high trisilanolisooctyl-POSS loadings (ca. 10 wt%), as shown also in Table 3. This phenomenon is clearly related to the plasticization effect

Table 3
The results of dynamic mechanical analysis (DMA) and DSC for various POSS/polycarbonate composites

POSS/PC samples	DMA results			DSC T_g (°C)
	E' at 30 °C (GPa)	Peak values of tan δ at T_g	T_g (°C)	
Polycarbonate control	1.68	1.94	153.2	150.1
Trisilanolphenyl-POSS				
2.5 wt%	1.63	1.85	152.8	149.6
5 wt%	1.67	1.83	154.2	149.6
10 wt%	1.88	1.72	153.3	148.5
Trisilanolisooctyl-POSS				
2.5 wt%	1.48	2.17	152.4	148.0
5 wt%	1.61	1.95	151.8	148.2
10 wt%	1.36	2.05	152.0	145.3
Octaphenyl-POSS				
2.5 wt%	1.51	1.99	145.1	–
Al-phenyl-POSS				
2.5 wt%	1.78	1.87	145.5	–

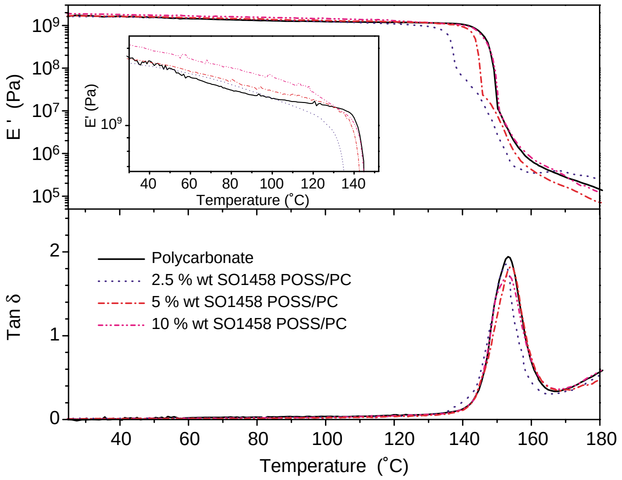


Fig. 2. Dynamic mechanical spectra (depicting E' and tan δ) of trisilanolphenyl-POSS(SO1458)/polycarbonate (PC) composites at different POSS concentrations. The inset is the zoom-in plot of E' data before the glass transition in selected temperature region.

due the addition of such liquid-like POSS. In Fig. 4 and Table 3, the DMA T_g and storage modulus show a small drop, in consistent with the DSC results. Correspondingly, the peak values of tan δ for these composites are slightly larger than that of the PC control (Fig. 4 and Table 3). Apparently the drop of the mechanical properties of such composites is due to the poor compatibility between filler and matrix as was indicated above.

As shown in Table 1, the PC composites with octaphenyl-POSS and Al-POSS demonstrate either opaque or colored films, and their overall properties are worse than the PC with trisilanol type POSS. Thus mechanical testing was carried out only for 2.5 wt% samples of these composites; the DMA results are shown in Fig. 5, and the data of 2.5 wt% trisilanol/PC composites are also shown as a comparison. The DMA T_g values of the PC composites with octaphenyl-POSS and Al-phenyl-POSS are considerably lower than that of the trisilanol/PC composites (also in Table 3). However, the storage modulus of the Al-phenyl-POSS/PC composite before the T_g transition is higher than the other POSS/PC composites and the PC control.

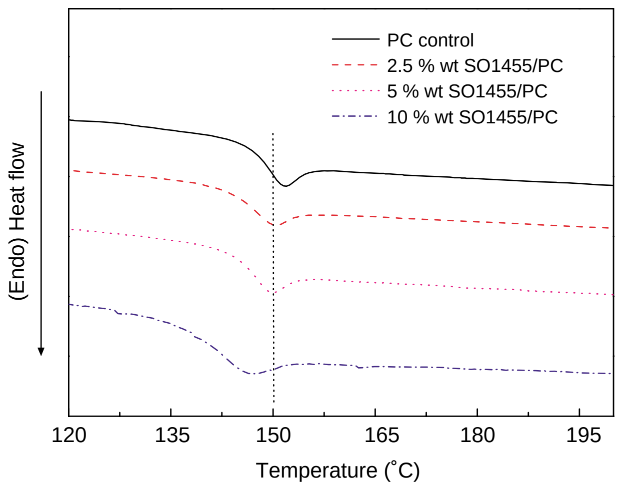


Fig. 3. DSC curves of trisilanolisooctyl-POSS/polycarbonate composites.

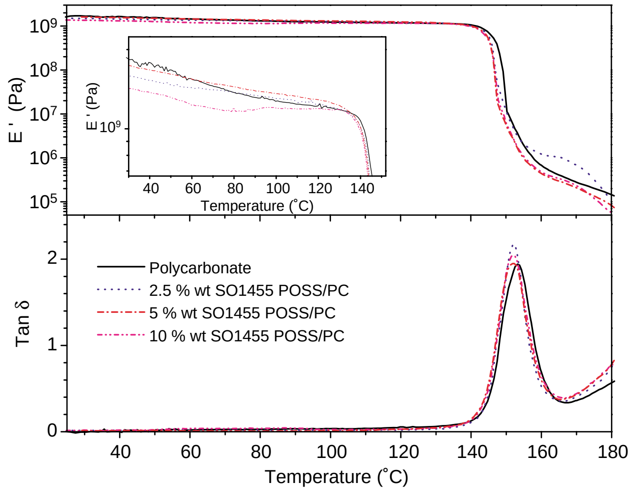


Fig. 4. Dynamic mechanical spectra (depicting E' and tan δ) of trisilanolisooctyl-POSS(SO1455)/polycarbonate (PC) composites at different POSS concentrations. The inset is the zoom-in plot of E' data before the glass transition in selected temperature region.

The DMA results were obtained for the trisilanolphenyl-POSS(SO1458)/HTLT Z-1050 composites and are presented in Fig. 6 and Table 4. We note that the T_g of this PC-polysulfone thermoplastic is significantly higher (∼84 °C) than that of the pure PC. The trisilanolphenyl-POSS again shows the good compatibility with this polymer, as discussed above. However, the incorporation of 5 wt% trisilanolphenyl-POSS in HTLT Z-1050 matrix leads to a slight drop of both the DMA T_g and the storage modulus (Table 4), suggesting that this POSS may not be the candidate for the reinforcement of this high T_g polymer. As shown in Fig. 7, the results of the thermal stability through thermogravimetric analysis demonstrate almost identical behaviors of thermal degradation before 540 °C for both samples. However, the residue amount 14% (after 600 °C) of unfilled HTLT Z-1050 is slightly lower than the value 18% of the trisilanolphenyl-POSS(SO1458)/HTLT Z-1050 composite, which is in good agreement with the 5 wt% added POSS (which would be converted into an inorganic silica at high

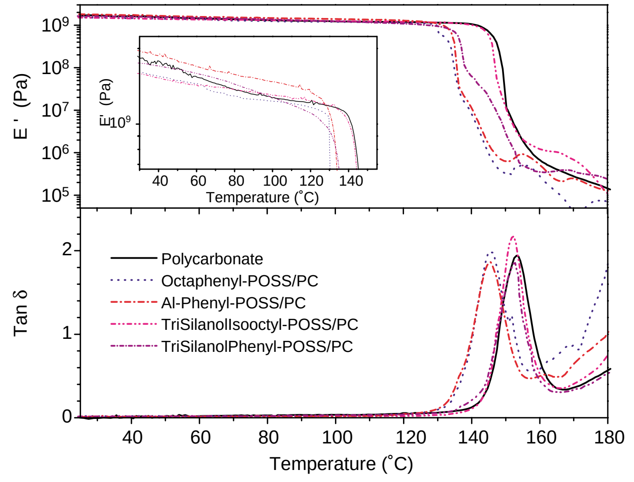


Fig. 5. Dynamic mechanical spectra (depicting E' and tan δ) of 2.5 wt% different POSS/polycarbonate (PC) composites. The inset is the zoom-in plot of E' data before the glass transition in selected temperature region.

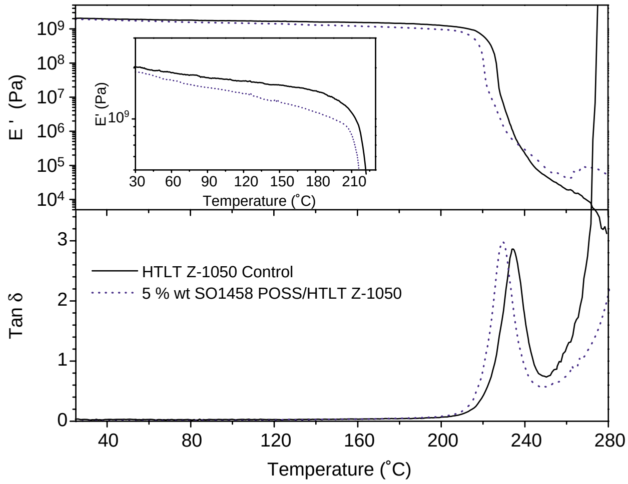


Fig. 6. Dynamic mechanical spectra (depicting E' and tan δ) of HTLT Z-1050 control and 5 wt% trisilanolphenyl-POSS(SO1458)/HTLT Z-1050 composite. The inset is the zoom-in plot of E' data before the glass transition in selected temperature region.

temperatures). Thus, the addition of SO1458 POSS has a very small effect on the thermal stability of HTLT Z-1050 sample.

3.3. Tensile properties

Since the trisilanolphenyl-POSS/PC composites show better performance than other POSS/PC combinations, these samples were further subjected to the tensile testing. Polycarbonate is a transparent, ductile amorphous engineering polymer. The addition of trisilanolphenyl-POSS gives the typical stress–strain curves shown in Fig. 8. Fracture strain and strength for PC decreases monotonically with increasing POSS loading levels. The fracture strain deceases significantly from 120% for the PC control to 40% for 10 wt% POSS/PC, together with a small drop in the fracture strength. These results are indicative of disruption of the polymer molecular structure, though the magnitude of strain loss is not as great as would be expected with such high loadings of traditional fillers. The statistical results of the tensile properties at room temperature are tabulated in Table 5. The modulus before 2% strain (661–674 MPa) and yield stress (51–52 MPa) and strain (~11.5%) of POSS/PC composites are very close to that of the PC control and are within the statistical deviation while fracture strain and strength apparently decreases. This indicates that the addition of up to 10 wt% trisilanolphenyl-POSS to PC matrix may still maintain the overall strength of virgin polymer with some loss of some ductility, although the 10 wt% POSS/PC composite may display a slightly enhanced modulus. The yielding behaviors of these samples are associated with the necking of the test specimens. The necking region of the PC control stays transparent, whereas the necking zones of the POSS/PC composites gradually become opaque with the increase of POSS concentration.

Table 4
The results of dynamic mechanical analysis (DMA) for trisilanolphenyl-POSS(SO1458)/HTLT Z-1050 composites

Samples	E' at 30 °C (GPa)	Peak values of tan δ at T_g	T_g (°C)
HTLT Z-1050 control	2.01	2.86	234.5
2.5 wt% trisilanolphenyl-POSS/HTLT Z-1050	1.89	2.99	229.3

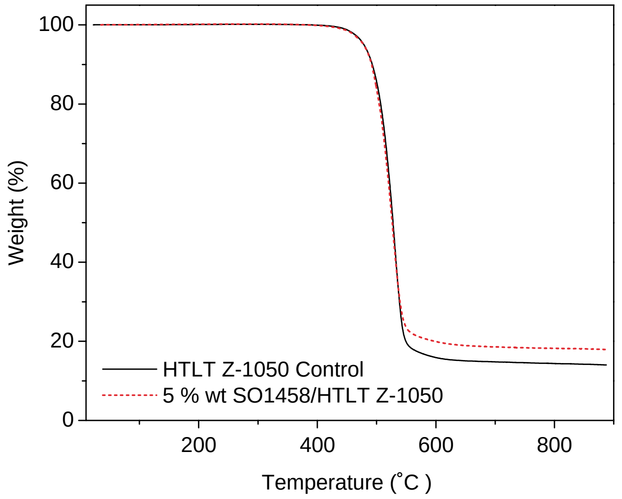


Fig. 7. Thermogravimetric analysis of the HTLT Z-1050 control and trisilanolphenyl-POSS(SO1458)/HTLT Z-1050 composites.

3.4. Morphology analysis

The microstructure of the trisilanolphenyl-POSS/PC composites was of further interest to be studied by the wide-angle X-ray diffraction (XRD) measurements. Fig. 9 shows the XRD profiles of these samples in increasing POSS concentration (b–d), which are quite similar to the amorphous PC control (a). However, a small diffraction peak at $2\theta=7.0°$ ($d=12.7$ Å) was observed in these POSS composites. Obviously this peak corresponds to the strong diffraction of the pure POSS(SO1458) powders (e), which is also a typical crystal peak of other POSS with similar structure [19,20]. Hence, this indicates that the separate POSS domains in crystalline phase are present in these POSS/PC composites. As a result, we also

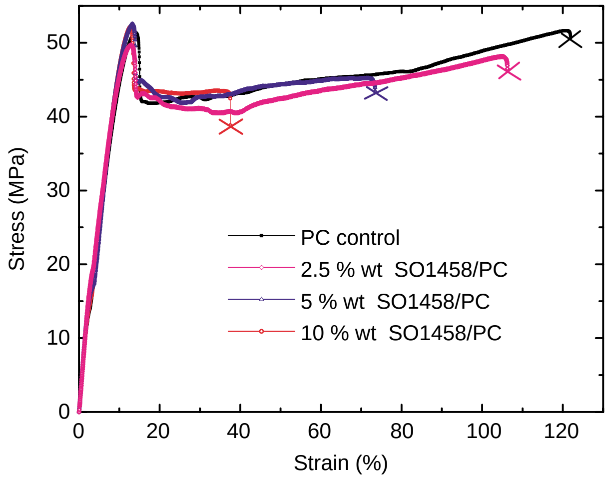


Fig. 8. Typical stress–strain plot of trisilanolphenyl-POSS (SO1458)/polycarbonate (PC) composites at different POSS concentrations.

Table 5
The tensile and optical properties of trisilanolphenyl-POSS/polycarbonate composites

Composition of POSS	Modulus[a] (MPa)	Yield stress (MPa)	Yield strain (%)	Fracture stress (MPa)	Fracture strain (%)
PC control	667 ± 12	52.0 ± 1.2	11.9 ± 0.3	51.2 ± 1.8	115.3 ± 16.8
2.5 wt%	661 ± 14	50.9 ± 2.3	11.4 ± 0.4	49.5 ± 2.5	115.0 ± 15.9
5 wt%	661 ± 18	51.9 ± 2.9	11.7 ± 0.7	43.1 ± 2.6	44.2 ± 22.2
10 wt%	674 ± 17	51.4 ± 1.2	11.3 ± 0.4	42.3 ± 1.1	29.2 ± 12.0

[a] Apparent modulus is obtained from data before 2% strain.

observed the increase of the peak intensity with increasing the POSS concentration. On the other hand, the POSS-polymer interactions (either polar or covalent) may also possibly play a role in the enhanced compatibility and give rise to the broadening of the peak of $2\theta = 7.0°$.

Fig. 10 shows the XRD profiles of the oriented samples taken from the stretched part of the tensile specimens (Scheme 2). Clearly the orientation has no effect on the diffraction profile of amorphous PC control. However, it is interesting to observe four additional peaks of $2\theta = 14.0°$, 16.9, 18.4 and 25.5 ($d = 6.3$, 5.2, 4.8 and 3.5 Å) for both parallel and perpendicular aligned 10 wt% SO1458/PC samples, which corresponds to four crystalline peaks on the pure POSS (SO1458) profile. The peak at 7.0° seems to be slightly enhanced as well due to the orientation. This again supports the presence of separate POSS domains in this POSS/PC composite. The stretching tends to align these POSS crystalline domains and makes some crystal forms more prominent on the XRD patterns. We also point out that both parallel and perpendicular alignment produce almost the same diffraction pattern, which is possibly due to the shearing bands at 45° for the tensile test of PC samples. More work is under way toward a better understanding of these results.

3.5. Izod impact tests of PC and PC/POSS

Four POSS/PC composites were selected for larger scale compounding, based on the lab-scale results presented above. These resins were injection molded then Izod impact tests were preformed on the molded samples. Each of the four polycarbonate-based samples were easily injection molded to produce clean, high luster test parts. Both of the POSS-containing samples developed a noticeable yellow tint during injection molding—this coloration was not present in the starting pellets, nor after drying prior to molding.

Impact testing of each material gave highly reproducible results (Table 6). Two different grades of PC were tested to ensure internal consistency—both grades performed equally, suggesting that neither product contained proprietary additives which would subtly alter interactions with POSS and ultimate performance. While all four PC samples showed the excellent un-notched impact properties expected, the PC/3 wt% PM1271 (partially polymerized trisilanolphenyl POSS) sample exhibited noticeably lower toughness when de-gating injection molded parts. This lower toughness was observed in notched izod impact testing, where its strength was an order of magnitude less than the un-filled resin. The PC/SO1458 (trisilanolphenyl POSS) showed no

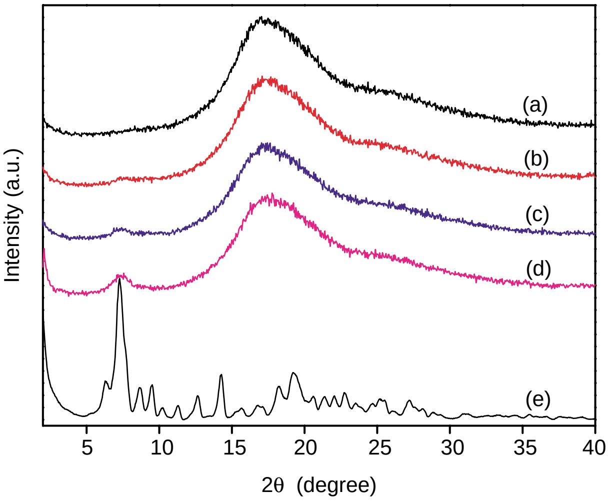


Fig. 9. XRD profiles of the trisilanolphenyl-POSS (SO1458)/polycarbonate (PC) composites at different POSS concentrations: (a) PC control; (b) 2.5 wt%; (c) 5 wt%; (d) 10 wt%; (e) POSS(SO1458) powder.

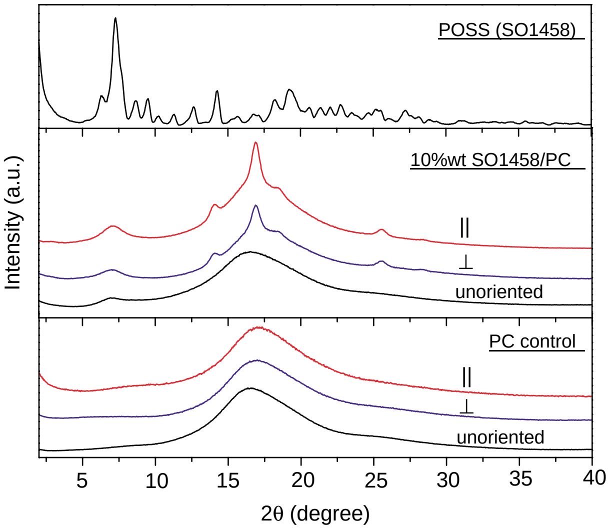


Fig. 10. XRD profiles of the oriented samples (from tensile tests) of PC control and 10 wt% trisilanolphenyl-POSS (SO1458)/polycarbonate (PC) composite. The powder diffraction profile of POSS (SO1458) is also shown as comparison.

Table 6
Data for the impact tests of POSS/PC composites

Material	Notched Izod (ft-lb/in.)	Un-notched Izod (ft-lb/in.)
Calibre PC	2.04 ± 0.16 (hinge)	> 5 (NB)
Makrolon PC	1.80 ± 0.06 (hinge)	> 5 (NB)
PC/3% PM1271	0.17 ± 0.03 (break)	> 5 (NB)
PC/3% SO1458	1.64 ± 0.10 (hinge)	> 5 (NB)

statistically significant difference in notched impact strength from un-filled PC. The most likely explanation for the decreased impact properties in the PC/PM1271 material is that this POSS material is less well dispersed in the polymeric matrix; poor distribution of POSS was previously observed to correlate with composite brittleness in PET/POSS composites [14].

4. Conclusions

POSS derivatives exhibit different compatibilities with polycarbonate, depending on the nano-scale filler's specific structure. Trisilanol POSS molecules generally provide better compatibility with PC than other POSS derivatives tested in this study. Octaphenyl-POSS/PC composites show reduced optical transparency, suggesting a lower level of compatibility, while Al-phenyl-POSS/PC and Al-isobutyl-POSS/PC produced colored films with dispersed bubbles, indicative of filler degradation. The PC composites with liquid trisilanolisooctyl-POSS exhibited slightly reduced T_g and storage modulus values due to plasticization effects. Trisilanolphenyl-POSS/PC composites possess the best overall performance among the POSS materials tested. The high compatibility between the trisilanolphenyl-POSS and polycarbonate matrix results in generation of transparent samples up to 5 wt% POSS content. Slightly enhanced mechanical properties including tensile and dynamic mechanical modulus are observed with the increase of trisilanolphenyl-POSS loading at the cost of decreasing ductility of the nanocomposites. The 3 wt% trisilanolphenyl-POSS/PC shows no statistically significant difference in notched impact strength from the PC control. Optimal loading of compatible, crystalline grades of POSS would appear to be approximately 5 wt%, balancing mechanical and optical properties of the system. These results suggest that the trisilanolphenyl-POSS/Polycarbonate composites show similar optical, thermal and mechanical properties to the polycarbonate control, although the XRD results indicate the presence of some ultra-fine POSS crystalline domains in PC matrix. Importantly, upon orientation of the PC/POSS nanocomposite, crystallization of POSS within the oriented material results—this observation is consistent with a growing number of observations which suggest that 'bottom-up' formation of structures incorporating multiple POSS cages result from orientation of these nanocomposites, and that the hybrid organic-inorganic inclusions may be at the heart of observed nano-scale reinforcement. The incorporation of trisilanolphenyl-POSS into another high T_g PC-polysulfone thermoplastics (HTLT Z-1050) leads to slight drop of T_g and storage modulus as well as similar thermal degradation behavior. The incorporation of POSS molecules into PC matrix potentially imparts new surface and optical properties (e.g. chemical resistance, anti-scratch property, flame retardant, tunable refractive index) which constitute the future exploratory research efforts toward the application of such POSS/PC nanocomposites.

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