Flame Retardancy of a Polycarbonate–Polydimethylsiloxane Block Copolymer: The Effect of the Dimethylsiloxane Block Size

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ABSTRACT: This report describes the flame retardancy of a polycarbonate (PC)–polydimethylsiloxane (PDMS) block copolymer with a dimethylsiloxane (DMS) block size of 15–350 units, and the effects of the block size and amount of DMS on the flame retardancy are studied. PC–PDMS block copolymers with DMS units of 40–130 had high limiting oxygen index values with 1.0 wt % PDMS. The PDMS block size influenced the PDMS dispersibility in PC, and a moderate PDMS dispersion (ca. 50 nm) caused high flame retardancy for PC. These PC–PDMS block copolymers could form a lot of fine bubbles in the role of good thermal insulators through the reaction of PC and PDMS in combustion. Furthermore, the silica particles from PDMS remained mostly on the surface of the char, so the amount of char with high oxidation resistance increased. © 2006 Wiley Periodicals, Inc. J Appl Polym Sci 100: 565–575, 2006

Key words: block copolymers; degradation; flame retardance; polycarbonates

INTRODUCTION

Polycarbonate (PC) has excellent mechanical properties and flame retardancy and is used for various applications. For electron and electric applications, because high flame retardancy is required, flame-retardant technologies for PC have been developed.[1–7] Flame-retardant PC through the addition or copolymerization of silicone compounds has been investigated.[8–12] Kambour et al.[8] reported the behavior of several families of polydimethylsiloxane (PDMS) block copolymers with respect to the limiting oxygen index (LOI), and PC–PDMS block copolymers with a dimethylsiloxane (DMS) block size of 2–40 units were found to increase the LOI. When the molar ratio of bisphenol A (BPA) to DMS in the PC–PDMS block copolymers was equal, the LOI value was maximum. The behavior was correlated with a rise in the yield of pyrolytic char and an improvement in the char oxidation resistance. Recently, flame-retardant technologies through the blending of silicone compounds were developed. Iji and Serizawa[12] reported that a special silicone with a branched chain structure and with an aromatic group in the chain was greatly effective in retarding the combustion of PC.[12]

This report describes the flame retardancy of PC–PDMS block copolymers with a DMS block size of 15–350 units, and the effects of the block size and amount of DMS on the flame retardancy are studied to consider the flame-retardant mechanism.

EXPERIMENTAL

Materials

Commercial BPA, *p-tert*-butylphenol (PTBP), triethylamine (TEA), sodium hydroxide, and dichloromethane were used without further purification. Reactive PDMS compounds with a DMS block size of 15–350 units were supplied by Dow Corning Toray Silicone Co., Ltd. The PC used in this study was Toughlon FN1900A (Idemitsu Kosan Co., Ltd., Japan). This was a BPA-type PC with a viscosity-average molecular weight (M_v) of 19,000 and a melt flow index of 20 g/10 min (at 300°C and 1.2 kg).

Preparation of the polycarbonate oligomer (PCO)

BPA (60 kg) was dissolved in 5 wt % aqueous sodium hydroxide (400 L). This solution, dichloromethane, and phosgene were introduced through an orifice plate at 25°C at flow rates of 138, 69, and 10.7 kg/h, respectively, into a tubular reactor with an inner di-

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TABLE I
Characteristics of PC–PDMS Block Copolymers

| Kind | Block copolymer | | |
	DMS units (n)	M_v	PDMS content (wt %)
PC	0	19,000	0
PC–PDMS-15	15	19,300	2.8
PC–PDMS-40	40	19,200	3.0
PC–PDMS-70	70	19,000	3.0
PC–PDMS-100	100	19,300	3.0
PC–PDMS-130	130	19,400	2.8
PC–PDMS-150	150	19,500	2.8
PC–PDMS-350	350	18,900	2.6

ameter of 10 mm and a length of 10 m. This procedure was continued for 3 h. After the reaction solution was left for a certain time, the organic phase was separated to obtain a PCO solution with an oligomer concentration of 317 g/L. The degree of polymerization of the resultant PCO was 3–4.

Preparation of the PC–PDMS block copolymer

A solution prepared through the dissolution of reactive PDMS (185 g) in dichloromethane (2 L), 2.6 wt % aqueous sodium hydroxide (1 L), and TEA (5.7 cc) was added to a PCO solution (10 L). The mixed solution was stirred at 500 rpm at room temperature for 1 h. BPA (600 g), 5.2 wt % aqueous sodium hydroxide (5 L), and a solution prepared through the dissolution of PTBP (81 g) in dichloromethane (8 L) were further added and stirred at 500 rpm for 1 h at 23°C. After this, dichloromethane (5 L) was added, and the solution was washed successively with water (5 L), 0.01M aqueous sodium hydroxide (5 L), 0.1M aqueous hydrochloric acid (5 L), and water (5 L). Then, the solvent was evaporated to obtain the PC–PDMS block copolymer in a flake form. The resulting PC–PDMS block copolymer was dried for 12 h at 110°C and pelletized by an extruder at 280°C. Table I lists the characteristics of the resulting PC–PDMS block copolymers. Reactive PDMS with a different DMS unit size was used with the same procedure to change the PDMS chain length in the PC–PDMS block copolymer. Furthermore, the amount of PDMS was prepared by the melt kneading and extrusion of a mixture of the PC–PDMS block copolymer and PC (Toughlon FN1900A).

Molding

The resulting pellets were dried at 120°C for 5 h and then injection-molded at an injection temperature of 280°C into test pieces for the measurement of the flame retardancy.

Limited oxygen index (LOI)

LOI measurements were performed according to the JIS K7201 method (15–20-mm ignition flame) with a candle-type flammability tester (Toyo Seiki Seisaku-Sho, Japan). Sample bars 3 mm thick were made by injection molding and were conditioned for 1 week at 23°C and 50% relative humidity.

Flame-retardant test by a cone calorimeter

The measurements were performed at an incident heat flux of 50 kW/m^2 with a cone heater (Toyo Seiki Seisaku-Sho). The sample plates (100 mm × 100 mm), 3 mm thick, were made by injection molding and were conditioned for 1 week at 23°C and 50% relative humidity. The tests were conducted twice for each sample.

Scanning electron microscopy

After the LOI test, the morphology of the char was observed by scanning electron microscopy (JSM-6100, JEOL, Ltd., Japan) at an acceleration voltage of 15 kV. The char sample was sputter-coated with gold with an ion sputter (JEC-1100, JEOL) to enhance conductivity.

Thermogravimetric analysis (TGA)

A PerkinElmer (Wellesley, MA) TGA-7 thermal analyzer was used to determine the weight loss during the thermal degradation of the PC–PDMS block polymers. Experiments were carried out on 10-mg samples under nitrogen at a furnace heating rate of 20°C/min up to 700°C. In combustion, oxygen was consumed in the gas phase, so degradation and char formation in the solid phase were generated under a non-oxygen atmosphere.

Haze

The haze was performed according to JIS K7205 with a haze meter (HZ-1, Suga Test Instruments Co., Ltd., Japan). The sample bars, 3 mm thick, were made by injection molding.

Si surface concentration

Si surface concentrations of the samples were measured by electron spectroscopy for chemical analysis (ESCA; JPS-90MC, JEOL). The conditions were Mg Kα rays (10 kV and 10 mA), an energy sweep range of 30 eV, and a step width of 0.1 eV. The analysis through the depth used the inclination method and the argon etching method. The plates (10 mm × 10 mm), 3 mm thick, were made by injection molding and were con-
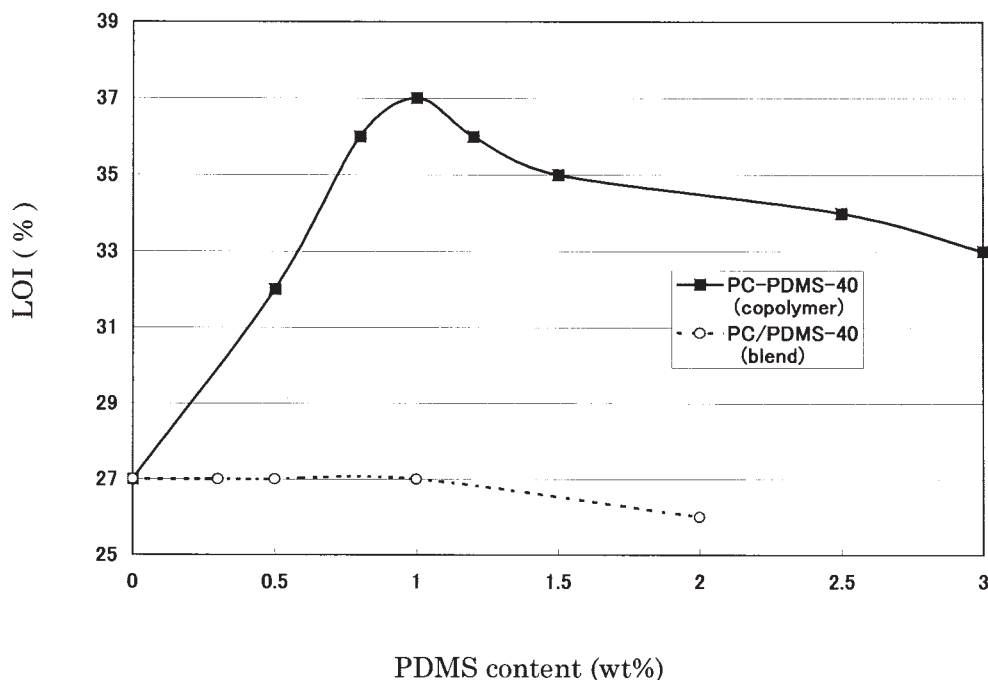
Figure 1 LOI values as a the function of the PDMS concentration.

ditioned for 1 week at 23°C and 50% relative humidity.

RESULTS AND DISCUSSION

Flame retardancy of the PC–PDMS block copolymer

The flame retardancy of the PC–PDMS block copolymer with a DMS block size of 40 units (PC–PDMS-40) was tested. The molar ratio of BPA to DMS in PC–PDMS-40 was almost the same. The PDMS copolymerization concentration (0–3.0 wt %) was prepared by the melt kneading and extrusion of a mixture of PC–PDMS-40 and PC. PC blended with the same structural reactive PDMS compound (PC/PDMS-40) was measured for comparison. The LOI values as a function of the PDMS copolymerization concentration are shown in Figure 1. PC–PDMS-40 caused an LOI increase, and the LOI value was maximum (37) at 1.0 wt % PDMS. When the PDMS copolymerization concentration increased more than 1 wt %, the flame retardancy decreased to LOI = 33 at 3.0 wt % PDMS. On the other hand, the simple addition of the same structural PDMS to PC (PC/PDMS-40) did not causes an LOI increase, so the flame retardancy of PC–PDMS-40 did not contribute PDMS itself, and PDMS copolymerization with PC is an important factor for flame retardancy. When a specimen of PC–PDMS-40 was in combustion, a lot of fine bubbles and char were formed, and the combustive behavior of PC–PDMS-40 (copolymer) was different from that of PC/PDMS-40

(blend). Photographs of TGA samples at 500°C and LOI test specimens after combustion for PC–PDMS-40 (copolymer: PDMS concentration = 1.0 wt %) and PC/PDMS-40 (blend: PDMS concentration = 2.0 wt %) are shown in Figure 2(a–d). PC–PDMS-40 (copolymer) generated a lot of bubbles and promoted charring. The combustive behavior of PC/PDMS-40 (blend) was almost the same as that of PC, which generated few bubbles and was slow to form char in combustion. It is known that BPA and DMS react at high temperatures as shown in Scheme 1.[13]

Therefore, because the branched structure of the PC–PDMS block copolymer was formed at the initial degradation temperature, it is thought that the melt tension increased and that bubbles swollen by degradation gases were generated. The morphologies of these chars were observed with scanning electron microscopy (Fig. 3). The char of PC–PDMS-40 (copolymer) looked like a closed cell foam that intercepted flammable gas from the inside. The char may have been formed from the conditions of bubbles appearing under the initial degradation. Kambour[14] reported a similar char morphology of a PC–PDMS block copolymer. Although the PDMS concentration of his PC–PDMS block copolymer was 18 wt % and much more than that of our sample, the PC–PDMS block copolymers had similar flame-retardant behaviors; that is, the char prevented more volatile fuel production and served as a thermal insulator preventing the temperature from rising. Next, the heat release rate (HRR) plots for PC–PDMS-40 (copolymer: PDMS concentration = 1.0 wt %) and PC/PDMS-40 (blend: PDMS concentration =2.0
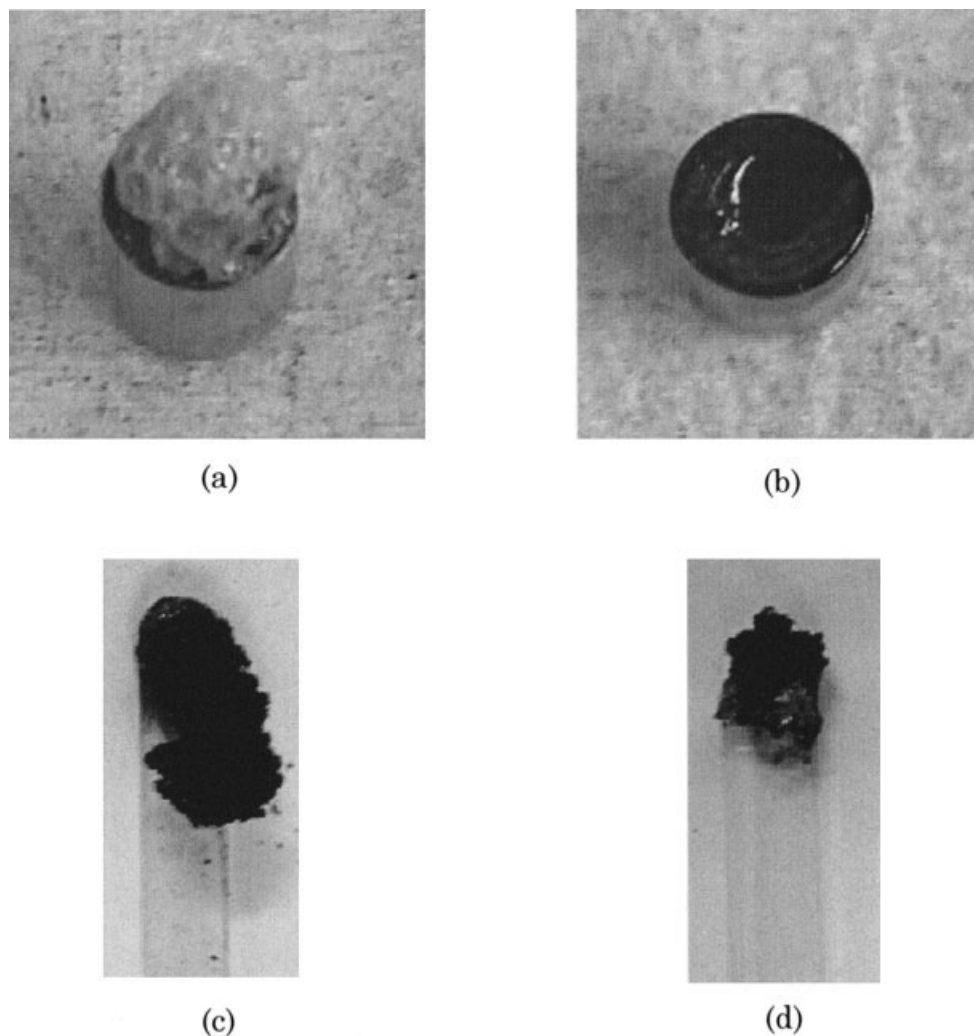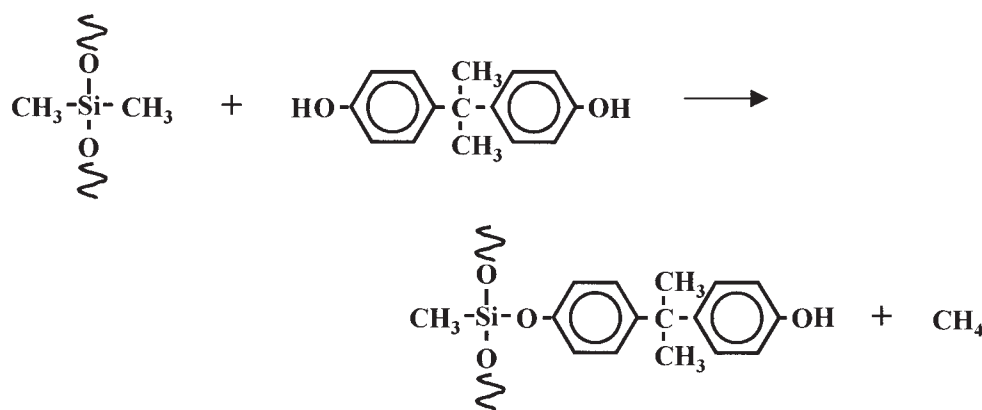
Figure 2 Photographs of TGA samples at 500°C [(a) PC–PDMS-40 (copolymer, PDMS concentration = 1.0 wt %) and (b) PC/PDMS-40 (blend, PDMS concentration = 2.0 wt %)] and photographs of LOI test specimens after combustion [(c) PC–PDMS-40 (copolymer, PDMS concentration = 1.0 wt %) and (d) PC/PDMS-40 (blend, PDMS concentration = 2.0 wt %)].

wt %) by cone calorimetry are shown in Figure 4. The peak HRR of PC–PDMS-40 (copolymer) was lower and broader than that of PC/PDMS-40 (blend), and the flame retardancy increased. Furthermore, PC–PDMS-40 (copolymer) had another peak HRR after 1000 s when the char layer on the surface collapsed. It is thought that
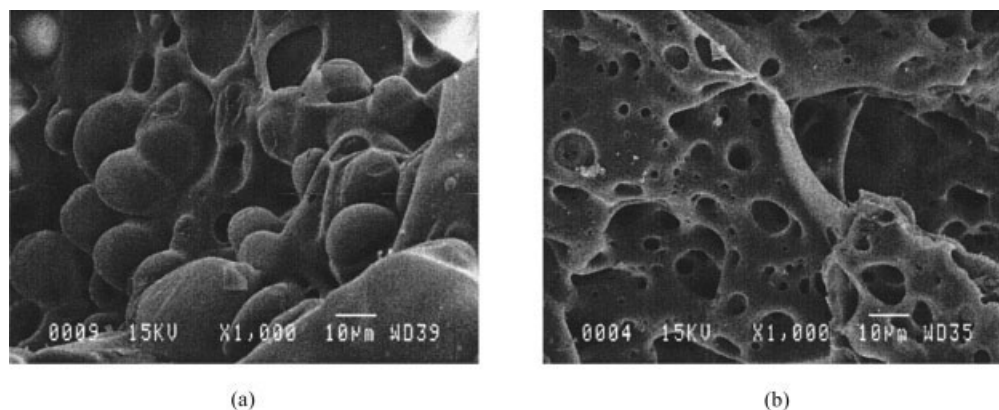

Scheme 1

Figure 3 Morphologies of char by scanning electron microscopy: (a) PC–PDMS-40 (copolymer, PDMS concentration = 1.0 wt %) and (b) PC/PDMS-40 (blend, PDMS concentration = 2.0 wt %).

PC–PDMS-40 (copolymer) had more heat-stable char, and this feature of the char was the factor increasing the flame retardancy.

Figure 5 shows the rates of weight loss versus the temperature in TGA to investigate the behavior of flame retardancy. The numbers in parentheses show the amounts (%) of residue at 650°C. The maximum rates of PC–PDMS-40 (copolymer) were lower than those of PC and PC/PDMS-40 (blend), and the initial degradation temperature decreased as the PDMS concentration increased. Furthermore, the amounts of residue for PC–PDMS-40 increased. It is suggested that PC and DMS reacted by the copolymerization of PDMS with PC, and the bubbles and the char formed by the reaction inhibited the polymer degradation. On the other hand, the

degradation behavior of PC/PDMS-40 (blend) was almost the same as that of PC. This reason is considered in the next section with the effect of the DMS block size. The high flame retardancy of PC–PDMS-40 was caused by the initial bubble formation and the increase in the thermally stable char. The decrease in the flame retardancy at more than 1.0 wt % PDMS in PC–PDMS-40 was caused by the flame spreading easily on the surface, as we previously reported for PC containing organic metal salts.[15]

Effect of the PDMS block size in the PC–PDMS block copolymer on the flame retardancy

The effect of the DMS block size in the PC–PDMS block copolymer on the flame retardancy was investi-
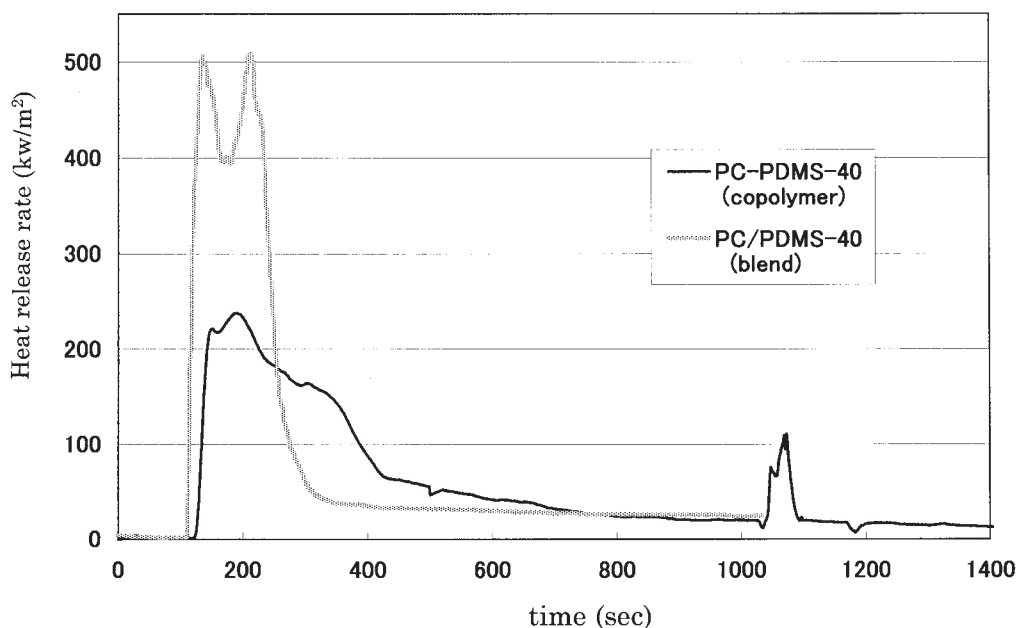

Figure 4 HRR plots for PC–PDMS-40 (copolymer, PDMS concentration = 1.0 wt %) and PC/PDMS-40 (blend, PDMS concentration = 2.0 wt %).
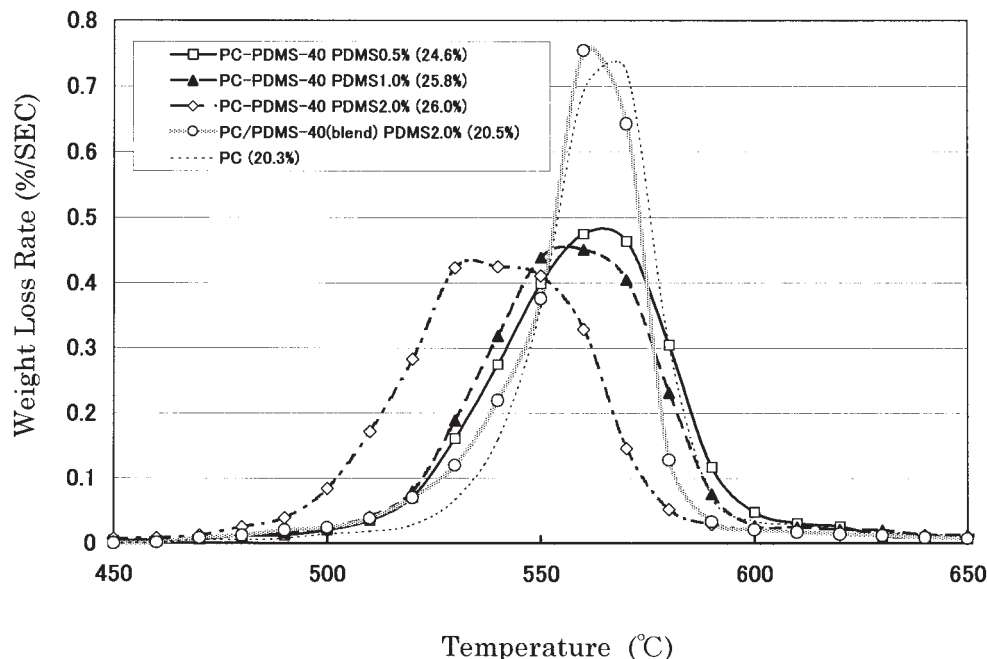
Figure 5 Weight-loss rate and amount of residue for PC–PDMS-40, PC/PDMS-40, and PC versus the temperature by TGA (the numbers in parentheses show the amounts of the residue).

gated. The DMS units ranged from 15 to 350. The LOI values as a function of the PDMS copolymerization concentration are shown in Figure 6. The PC–PDMS block copolymers with 40–350 DMS units had the maximum LOI value with 1.5 wt % PDMS, and PC–PDMS-70 (70 DMS units) and PC–PDMS-100 (100 DMS units) had the highest maximum LOI. Moreover,

as the PDMS block size increased, the maximum LOI value decreased, and PC–PDMS-350 (350 DMS units) did not show high flame retardancy any more. On the other hand, PC–PDMS-15 (15 DMS units) did not reach the maximum LOI at 2.5 wt % PDMS, and the LOI increase was lower. Kambour et al.[8] evaluated PC–PDMS block copolymers with short PDMS block
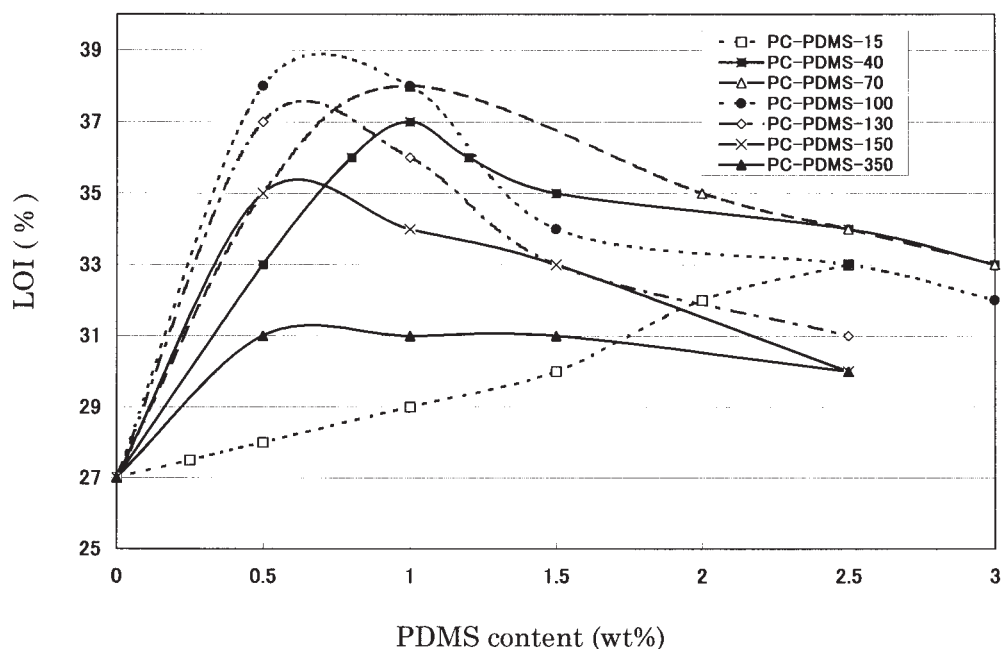

Figure 6 LOI as a function of the PDMS concentration for PC–PDMS block copolymers with a PDMS block size of 15–350.
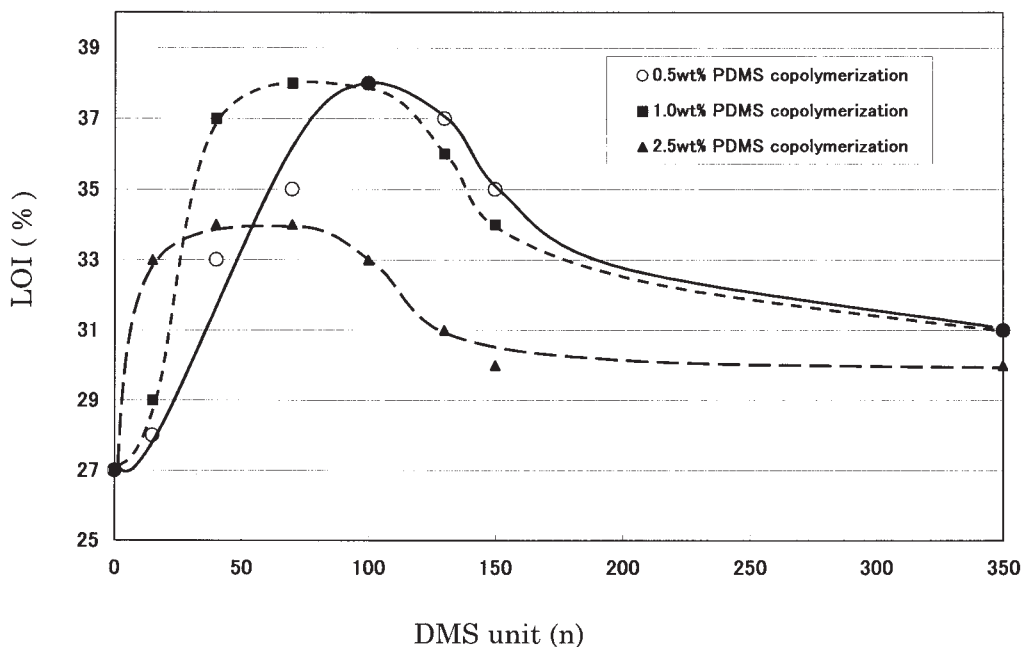
Figure 7 Relationship between the LOI and PDMS block size for 0.5, 1.0, and 2.5 wt % PDMS.

sizes (2–40 DMS units), and the LOI value of the PC–PDMS block copolymer with 11 DMS units was maximum (43) at 12 wt % PDMS. PC–PDMS-15 reached the maximum LOI when the PDMS concentration increased. Figure 7 shows the relationship between the LOIs and PDMS block size for 0.5, 1.0, and 2.5 wt % PDMS. The maximum LOI shifted to a short block size as the PDMS concentration increased, and the PC–PDMS block copolymer with a shorter PDMS block needed a greater PDMS concentration to increase the flame retardancy. The PDMS block size influenced the behavior of the flame retardancy for the PDMS concentration, and the flame retardancy of PC could increase at a small PDMS copolymerization concentration (0.5–1.0 wt %) when the PDMS block size was 70–130.

Figure 8 shows the rates of weight loss versus the temperature in TGA to investigate the difference in the flame retardancy of the tested PC–PDMS block copolymers (PDMS concentration = 1.0 wt %). The maximum rates of the PC–PDMS block copolymers tended to be lower when the LOI values were higher, and PC–PDMS-100 had the lowest. When the PDMS block size was lower, the degradation temperature of the PC–PDMS block copolymer tended to be lower, so the reaction of BPA and DMS in the thermal degradation could increase. However, PC–PDMS-15 with a shorter PDMS block did not form a lot of fine bubbles as PC–PDMS-40 did at the initial degradation temperature, and it is suggested that the flow properties (e.g., melt tension) of degraded PC–PDMS-15 were different from those of PC–PDMS-40. Next, the amounts of the residue as a function of the PDMS copolymerization concentration for each PC–PDMS block copolymer are shown in Figure

9. PC/PDMS-40 (blend: PDMS concentration = 1.0 wt %) was measured for comparison. The amounts of the residue for each PC–PDMS block copolymer increased rapidly until about 1.0 wt % PDMS. The amounts of the residue for PC–PDMS-100, PC–PDMS-150, and PC–PDMS-350 were almost saturated over 1.0 wt % PDMS, and those for PC–PDMS-15 and PC–PDMS-40 increased slowly. PC–PDMS-100, which had a high maximum LOI value, increased the amount of the residue at the low PDMS concentration. The amounts of the residue for PC–PDMS-15 and PC–PDMS-350, which had low LOI values, were less than those for the other PC–PDMS block copolymers. That is, the PDMS block size affected the amount of the residue. On the other hand, the amount of the residue for PC/PDMS-40 (blend) was almost the same as that of PC, and the residue generated from PC and PDMS increased by PDMS copolymerization with PC. As van Krevelen[16] reported that the LOI increases with the amount of the residue, the increase in the amount of the residue is one factor for the flame retardancy in PC–PDMS block copolymers. However, estimating from van Krevelen's correlation between the amount of the residue and the LOI value, we find that PC–PDMS block copolymers have high LOI values with respect to the amounts of the residue[16] (in the correlation, an LOI of 38 equals a residue amount of 50.) Therefore, the flame retardancy of the char from the PC–PDMS block copolymers was evaluated by LOI when the char, which was produced with an LOI tester under 40% oxygen, began to glow. This was also an evaluation of the char oxidation resistance in combustion. Figure 10 shows the LOIs of char as a function of the PDMS concentration for each PC–PDMS block copolymer. PC/
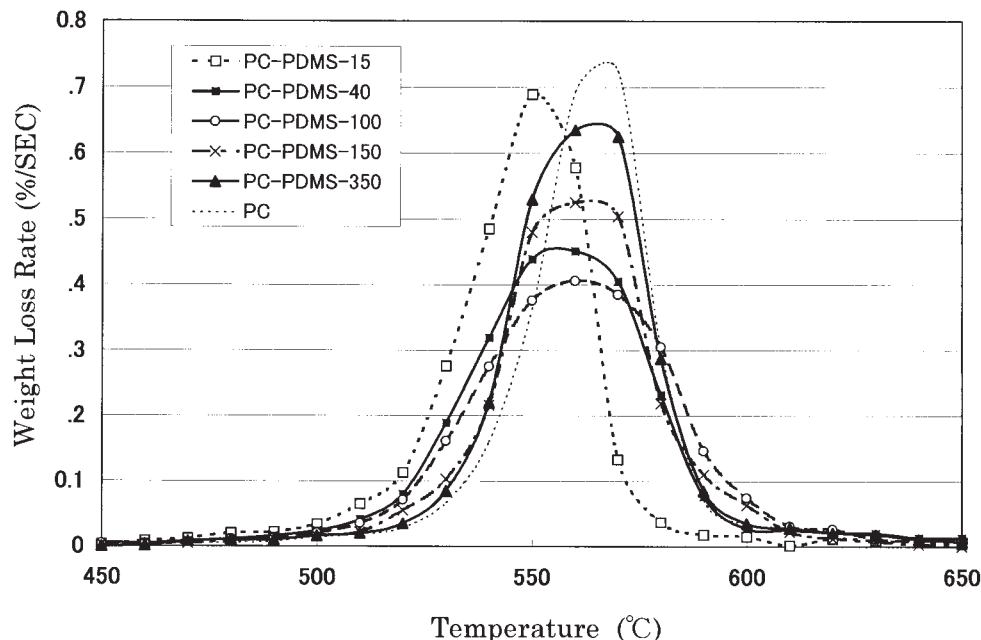
Figure 8 Weight-loss rate for PC–PDMS block copolymers (PDMS concentration = 1.0 wt %) and PC versus the temperature by TGA.

PDMS-40 (blend: PDMS concentration = 1.0 wt %) was measured for comparison. The LOI of char from ordinary PC was 48, and the addition of PDMS (PC/PDMS-40) did not cause the LOI to increase. Except for PC–PDMS-15, the LOIs of chars from PC–PDMS block copolymers increased rapidly until about 1.0 wt % PDMS and were saturated with more than 1.0 wt %. The maximum LOI of chars from PC–PDMS-40 and

PC–PDMS-100 was high (58). The increase in the LOI of the char was caused by fine silica particles, which did not glow anymore on the surface of char, and so it is thought that the chars of PC–PDMS-40 and PC–PDMS-100 included a lot of silica particles, which prevented the oxidation of char, as reported by Kambour.[14] Because a lot of silica remained in the char of these PC–PDMS block copolymers, the amount of the
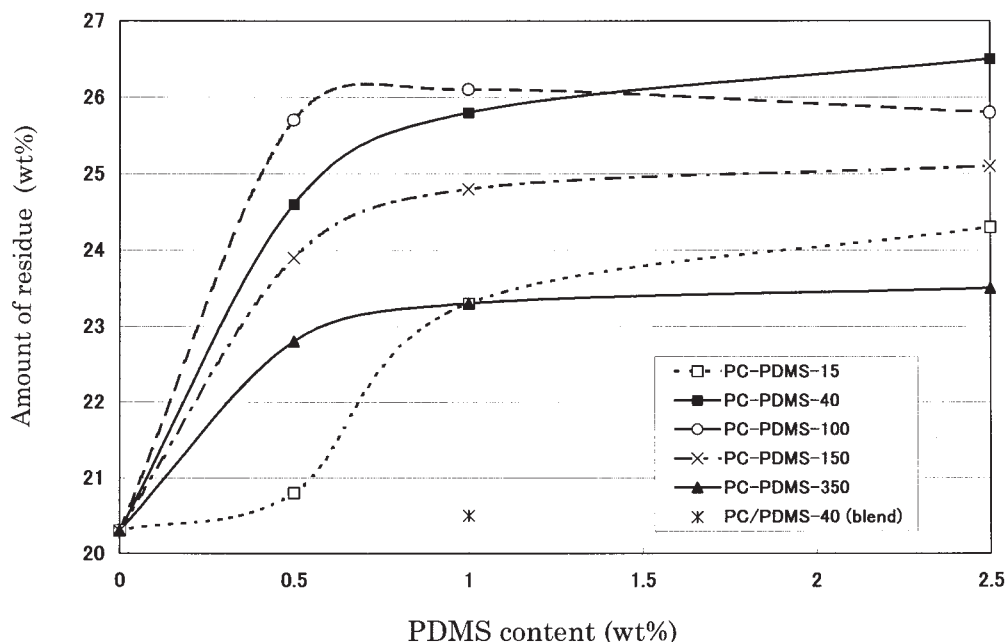

Figure 9 Amount of the residue as a function of the PDMS concentration for PC–PDMS block copolymers with a PDMS block size of 15–350 and for PC/PDMS-40 (blend).
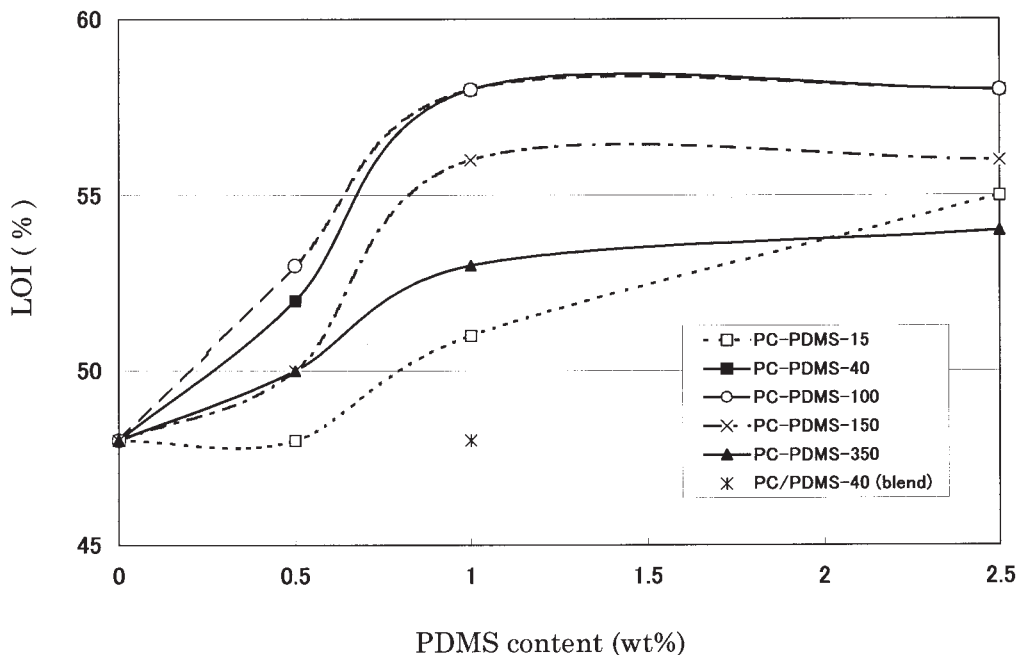
Figure 10 LOI of char as a function of the PDMS copolymerization concentration for PC–PDMS block copolymers with a PDMS block size of 15–350 and for PC/PDMS-40 (blend).

residue could increase. On the other hand, the LOI of char from PC–PDMS-15 did not increase until 0.5 wt % PDMS and then increased slowly. This behavior means that the silica particles remained with difficulty on the surface of char when the PDMS block size was smaller. This is a reason that the PC–PDMS block copolymer with shorter PDMS needed a lot of PDMS

to increase the flame retardancy (Fig. 7). Moreover, the addition of the PDMS compound (PC/PDMS-40) had little effect on the degradation behavior for PC, and hardly any silica remained on the surface of the char. The increase in the char oxidation resistance is another factor for the flame retardancy of the PC–PDMS block copolymers.
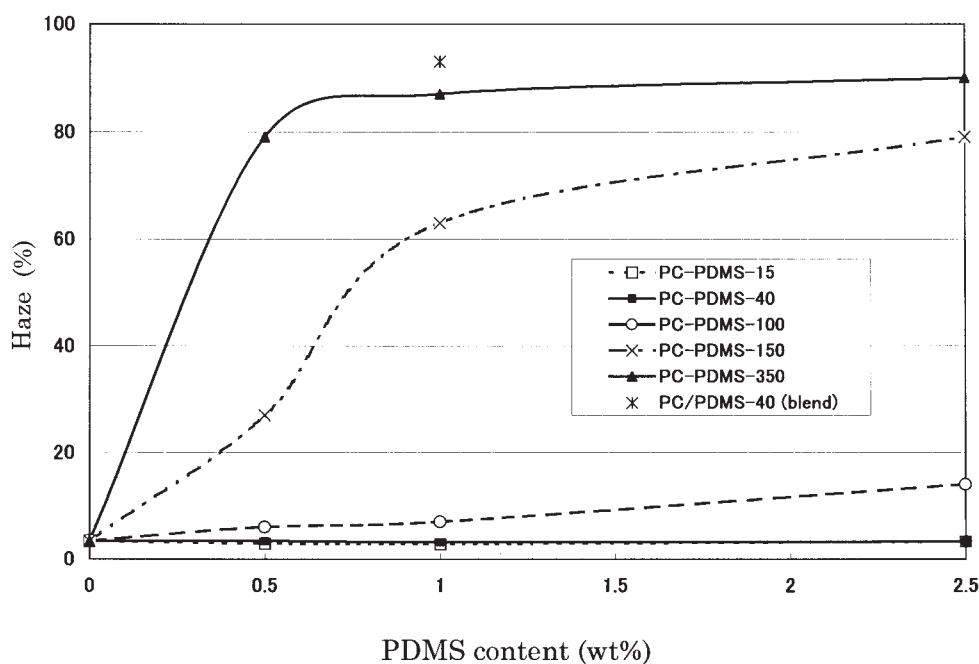

Figure 11 Haze as a function of the PDMS copolymerization concentration for PC–PDMS block copolymers with a PDMS block size of 15–350 and for PC/PDMS-40 (blend).
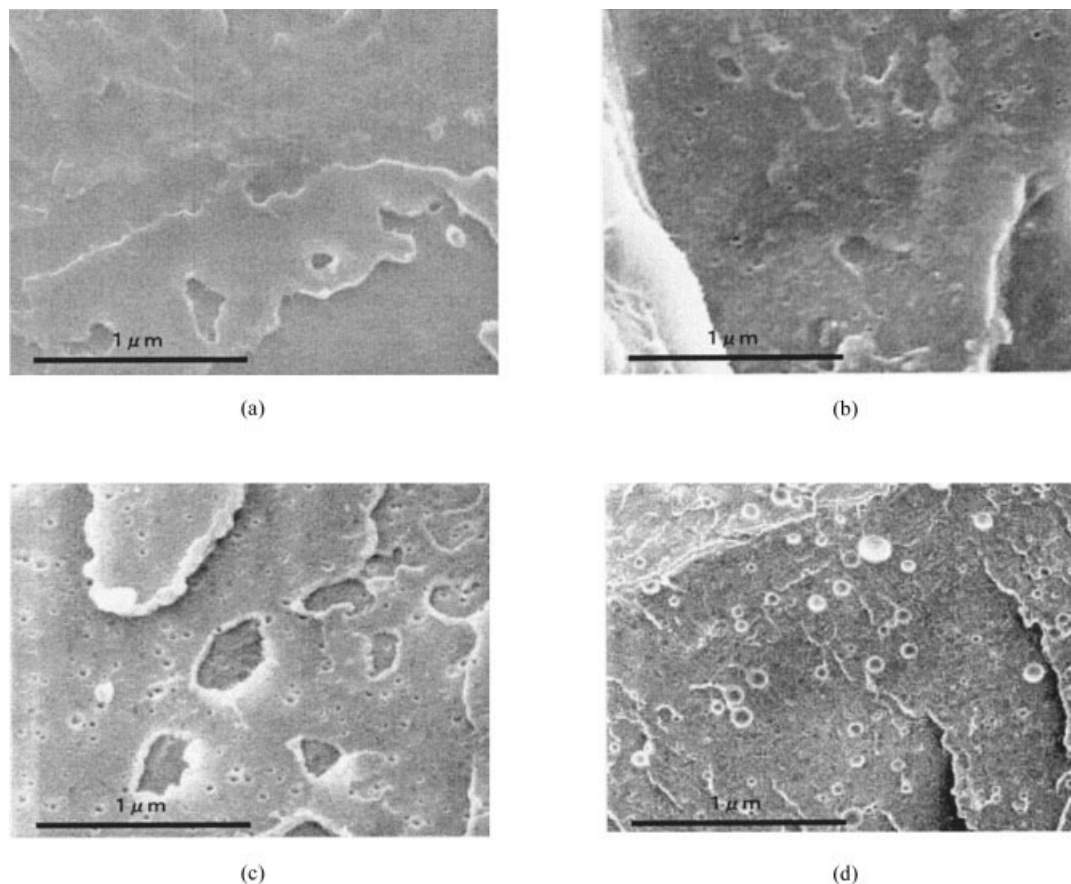
Figure 12 Morphologies by scanning electron microscopy: (a) PC–PDMS-40, (b) PC–PDMS-100, (c) PC–PDMS-350, and (d) PC/PDMS-40 (blend) with 2.5 wt % PDMS.

The dispersibility of PDMS was evaluated to investigate this different behavior of the PDMS block polymer. The dispersibility is thought to influence the amount of the residue of silica and the reaction of BPA and DMS. Figure 11 shows the values of the haze as a function of the PDMS copolymerization concentration for each PC–PDMS block copolymer. PC/PDMS-40 (blend: PDMS concentration = 1.0 wt %) was measured for comparison. Because PC and PDMS had different indices of refraction, PDMS aggregation increased the value of the haze. When the DMS units were 100 or fewer, the haze did not increase, and the PDMS dispersion was excellent. The haze of PC–PDMS-150 increased as the PDMS concentration increased, and PC–PDMS-350 and PC/PDMS-40 (blend) were opaque with a low PDMS concentration. The copolymerization of PDMS with PC caused a good dispersion of PDMS, and the increase in the PDMS block size promoted the formation of large PDMS aggregations because the compatibility of PC with PDMS was low. Figure 12 shows the morphologies of PC–PDMS-40, PC–PDMS-100, PC–PDMS-350, and PC/PDMS-40 (blend) by scanning electron microscopy. The PDMS concentration of these samples was 2.5 wt %. The PDMS domains seemed like black points

or spheres, and PDMS domains in PC–PDMS-40 were not observed. It is clear that PDMS had better dispersion when the PDMS block size was smaller and PDMS was copolymerized with PC. The PDMS domain size of PC–PDMS-100 was approximately 50 nm. Furthermore, the concentrations of PDMS in the near surface of the injection-molded plate for PC–PDMS block copolymers and PC/PDMS-40 (blend) with 1.0 wt % were evaluated by ESCA. The Si concentrations, given with respect to the C concentrations, are shown in Figure 13. When the PDMS dispersibility in PC was low (e.g., PC/PDMS-40 and PC–PDMS-350), the concentration of PDMS on the surface increased, and that of PC/PDMS-40 was very high. On the other hand, the concentration of PDMS on the surface of PC–PDMS-15 was low, and so it is thought that PDMS in PC–PDMS-15 was dispersed extremely finely.

On the basis of such results, PDMS on the surface did not cause an increase in the flame retardancy and therefore did not remain and form silica particles during thermal degradation. When the PDMS dispersibility was low (e.g., PC–PDMS-350 and PC/PDMS-40), the inside PDMS in the melt state easily migrated to the surface and condensed. Then, the PDMS on the surface could decompose and evaporate early by
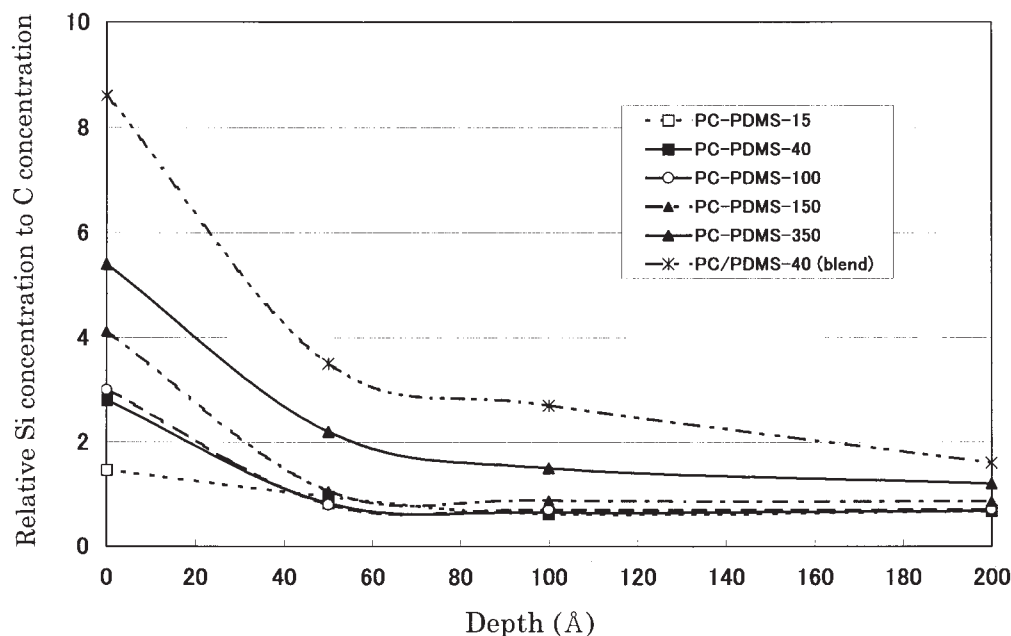
Figure 13 Si concentration with respect to the C concentration in the near surface of injection-molded plates for PC–PDMS block copolymers with a PDMS block size of 15–350 and for PC/PDMS-40 (blend) with 1.0 wt % PDMS.

flame before PC degraded. On the other hand, when the PDMS block size was smaller (e.g., PC–PDMS-15), the extreme reaction of PC and PDMS decomposed the polymer too much, and the melt tension could decrease reversely, so a lot of bubbles serving as a thermal insulator were not formed. Moreover, the decomposition products and silica, which were thought to become small, evaporated and did not remain mostly in the resin or char. Therefore, when the PDMS block size was 40–130, PDMS was dispersed moderately in PC, and the flame retardancy increased. The good PDMS dispersion (ca. 50 nm) was effective in the reaction with PC, and the decomposition products and silica from the PC–PDMS block copolymer with a medium PDMS block size were not hard to evaporate and remained mostly in the char. The decrease in the flame retardancy at a high PDMS concentration (excess 1%) was related to the lower temperature of the initial degradation in the PC–PDMS block copolymer because the amount and thermal stability of the char did not change after the maximum LOI. It is thought that the initial degradation temperature decreased with an increase in the reaction of PC and PDMS.

CONCLUSIONS

PC–PDMS block copolymers with 40–130 DMS units had high LOI values with a 1.0 wt % PDMS copolymerization concentration. The PDMS block size influenced the PDMS dispersibility in PC, and a moderate PDMS dispersion (ca. 50 nm) caused high flame retardancy. The PC–PDMS block copolymers with moderate PDMS dispersions could form a lot of fine bubbles,

which served as good thermal insulators, through the reaction of PC and PDMS in combustion. Furthermore, the silica particles from PDMS remained mostly on the surface of the char, so the amount of the char with high oxidation resistance increased. The char had a preferable structure that prevented volatile fuel productions and served as a thermal insulator.

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