Thermally Stable Aliphatic Polycarbonate Containing Bulky Carbazole Pendants

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ABSTRACT: To improve the thermal and mechanical properties of poly(propylene carbonate) (PPC), the terpolymers were synthesized by the terpolymerization of CO_2 with PO and a third monomer, N-(2,3-epoxylpropyl)carbazole (NEC) using supported zinc glutarate as catalyst. The catalytic activity, molecular weight, carbonate unit content, as well as the thermal and mechanical properties were investigated extensively. The experimental results showed that the catalytic activity, molecular weight, and carbonate unit content decreased with the incorporation of NEC. The introduction of NEC increased the glass transition temperature from 38.0 to 44.1°C. Moreover, the thermal decomposition temperature ($T_{g\text{-}5\%}$) of the terpolymer (278°C) was much higher than that of pure PPC (238°C). Accordingly, the mechanical properties proved to be enhanced greatly as evidenced by tensile tests due to the incorporation of bulky carbazole moieties. These improvements in thermal and mechanical properties are of very importance for the process of PPC. © 2008 Wiley Periodicals, Inc. J Appl Polym Sci 108: 3626–3631, 2008

Key words: aliphatic polycarbonates; terpolymerization; thermal stability; mechanical properties

INTRODUCTION

Carbon dioxide is considered the major greenhouse gas responsible for global warming, but it is also an inexpensive and abundant one-carbon resource, which storage on the earth is more than the sum of petroleum and coal. In this respect, the chemical fixation of CO_2 has become a very attractive subject. One of the most promising approaches is in the production of aliphatic polycarbonates from the copolymerization of CO_2 with oxiranes. As made materials have found uses in many applications, such as ceramic binders, evaporation mold, and adhesives.

Since the pioneering work of Inoue et al. in 1969,[1] researchers have strived to develop more efficient catalytic systems for the polycarbonate production. Thus, a lot of research work reviewed by Rokicki and Kuran,[2] Beckman,[3] Darensbourg and Holtcamp,[4] Sugimoto and Inoue,[5] and Coates and Moore[6] has demonstrated that CO_2 can be copolymerized with a number of epoxides. As described therein, in most cases propylene oxide and cyclohexene oxide were selected as epoxide resource. Although some improvements have been made on the activity of catalysts in the copolymerization of CO_2 with cyclohexene oxide, the resulting product, poly(cyclohexene carbonate), is less competitive in the restrictive world of commodity polymers because of its high cost of cyclohexene oxide and catalysts used. It is well known that synthetic commodity polymers are in general hardly degradable, causing ecological problems. Therefore, there is a strong need for polymeric materials which can degrade without producing any harmful stuffs. In this respect, poly(propylene carbonate) (PPC) derived from propylene oxide (PO) and CO_2 as a cheap and technically available comonomer appears to well meet above-said requirements.

As disclosed in literature,[7–14] alternating PPC was synthesized effectively from CO_2 and PO using zinc glutarate (ZnGA). However, the glass transition temperature (T_g) of PPC is rather low due to its flexible carbonate linkage in the backbone. The practical application of PPC has been limited by the poor thermal stability. To improve the thermal stability, many approaches including physical and chemical ones have been carried out. For example, many organic and inorganic components have been compounded with PPC to fabricate blends and composites.[15–17] PPC was also chemically end-capped by maleic anhydride, benzoyl chloride, ethyl silicate, acetic anhydride, and phosphorus oxychloride.[18] Moreover, block copolymerization of carbon dioxide with cyclohexene oxide and 4-vinyl-1-cyclohexene-

1,2-epoxide in based PPC by yttrium-metal coordination catalyst was also reported.[19]

It is well known that side chains have significant influence on the thermal properties of polymers. Eckl et al. reported that the T_g of polymethacrylates increased from about 100 to 190°C by introducing bulky adamant substitutes onto the side chains.[20] N-(2,3-epoxylpropyl)carbazole is a well known commercial monomer, which can be used for the preparation of photoconductive polymer.[21–23] The resulted polymer has relatively high T_g and thermal stabilities due to the presence of bulky and rigid carbazyl side groups.[24] In this case, the T_g of PPC can be increased by introducing a third rigid monomer.

We have previously reported the terpolymerization of CO_2 with PO and [(2-naphthyloxy)methyl] oxirane (NMO). The synthesized terpolymer showed limited thermal and mechanical properties compared with neat PPC. In this sense, we report herewith the terpolymerization of CO_2 with PO and N-(2,3-epoxyl-propyl)carbazole. The thermal and mechanical properties of the resulted terpolymers are fully characterized. The results demonstrate that the method is an effective way to modify PPC.

EXPERIMENTAL

Materials

Carbon dioxide of a purity of 99.99% was commercially obtained without further purification. Propylene oxide (PO) of 95.0% purity was pretreated by potassium hydroxide and refluxed over calcium hydride for 24 h. After the treatment, the water content of PO is lower (3 ppm). It was then distilled under dry nitrogen gas and stored over 4 Å molecular sieves prior to use. Carbazole is chemical pure reagent that was washed with a 5% solution of hydrochloric acid and recrystallized from ethyl alcohol, m.p. 110–111°C. Glutaric acid (GA) of 98.0% purity, zinc oxide of 99.0% purity and perfluorinated compound were used without further treatment. Epichlorohydrin, potassium carbonate, powdered potassium hydroxide, tetrabutylammonium bromide, and solvents such as toluene, methanol, acetone, chloroform, were of analytical reagent grade and used as received.

Preparation of catalyst

Supported zinc glutarate (s-ZnGA) was synthesized according to previous work.[14] To a two-necked flask equipped with mechanical stirrer, a Dean-Stark trap and a reflux condenser with a drying tube were added ZnO (100 mmol) and toluene (150 mL). Then, to the ZnO/toluene suspension was added GA (98 mmol) and perfluorinated compound (1.96 mmol). After the addition was complete, the slurry mixture was stirred vigorously at 55°C for 8 h. Upon cooling down, the reaction mixture was filtered off and washed with acetone several times, followed dried overnight in a vacuum oven at 80°C. The supported zinc glutarate obtained was a fine powder in white color with a high yield greater than 99.5%. The surface area was determined using Brunauer-Emett-Teller (BET) method. A NOVA1000 instrument used for the measurements was produced by Quantachrome Company of USA. The surface area of supported catalyst (82.9 m²/g) was much larger than that of unsupported catalyst (5.6 m²/g).

Synthesis of N-(2,3-epoxylpropyl)carbazole

N-(2,3-epoxylpropyl)carbazole (NEC) was prepared as described in the literature.[25] To an acetone solution (100 mL) in a two-necked flask equipped with an upright condenser and mechanical stirrer were added Carbazole (0.1794 mol, 30.0 g), potassium carbonate (1.2020 mol, 166.1 g), powdered potassium hydroxide (0.1311 mol, 7.4 g), epichlorohydrin (1.2020 mol, 94 mL), and tetrabutylammonium bromide (0.0179 mol, 5.8 g). The reaction mixture was stirred vigorously and heated in a water bath at 52°C for 1 h. Then, potassium carbonate (0.6003 mol, 83.0 g) and powdered potassium hydroxide (0.1311 mol, 7.4 g) were added to the mixture, and the reaction was continued for another 3 h. Subsequently, inorganic substances were separated by filtration and rinsed with acetone. The solvent was removed from the filtered solution to give a yellowish solid that was dissolved and recrystallized from ethanol to afford the white pure product (35.5 g, yield 89%). ^{1}H NMR (400 MHz, $CDCl_3$): 2.56 (d d, J = 2.6 and 4.8 Hz, 1H, CH—O—CHH), 2.79 (t, J = 4.0 Hz, 1H, CH—O—CHH), 3.33 (m, 1H, Ar—O—CH$_2$—CH —O—CH$_2$), 4.39 (d d, J = 4.8 and 15.9 Hz, 1H, Ar—N—CHH—CH—O—CH$_2$), 4.61 (d d, J = 3.4 and 15.8 Hz, 1H, Ar—N—CHH—CH —O—CH$_2$), 7.25 (m, 2H, Ar—H), 7.46 (m, 4H, Ar—H), 8.09 (d, J = 7.7 Hz, 2H, Ar—H). Pure N-(2,3-epoxylpropyl)carbazole was dried for 5 h under vacuum at 80°C and stored in a drier prior to use.

General polymerization procedure

The synthesis of PPC and PPCN were carried out in a 500 mL autoclave equipped with a mechanical stirrer using PO as both reactant and solvent.

The predried s-ZnGA was introduced into the autoclave as quickly as possible. The autoclave was capped with its head, and the entire assembly was connected to the reaction system equipped with a vacuum line. The autoclave with catalyst inside was further dried for 24 h under vacuum at 100°C and then was cooled to 30°C. Subsequently, the autoclave was purged carefully with carbon dioxide and evac-
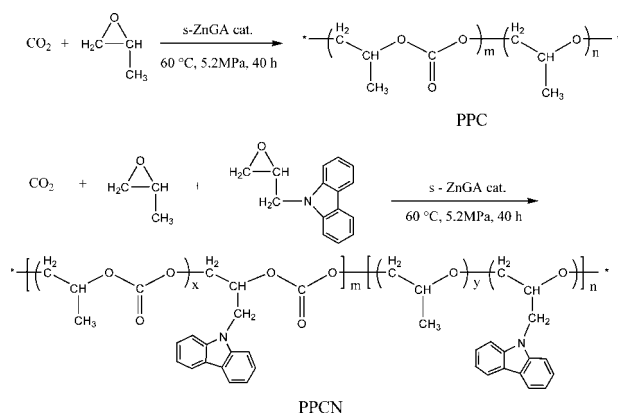
Scheme 1 Syntheses of PPC (top) and PPCN (bottom).

uated alternatively for three times, followed by adding purified quantitative PO with a large syringe. The autoclave was then pressurized to 5.2 MPa via a CO_2 cylinder. The copolymerization was performed at 60°C under stirring for 40 h and afterwards the autoclave was cooled to room temperature and the pressure was released. The resulting viscous mixture was unloaded and dissolved in a sufficient volume of chloroform containing 5% solution of hydrochloric acid to decompose the catalyst. The transparent organic layer was washed to the neutral reaction and slowly added to excess vigorously stirred MeOH to precipitate the polymer. The precipitated polymer was additionally washed with MeOH several times and dried at 80°C under vacuum to a constant weight. The as-made copolymer was weighed to calculate the yield of the copolymer.

PPCN was synthesized in the similar procedure to that of PPC, except that the required N-(2,3-epoxyl-propyl)carbazole was quickly added into the autoclave when the autoclave was cooled to 30°C.

Measurements

^{1}H NMR spectra were recorded on a Bruker DRX-400 NMR spectrometer using tetramethylsilane as an internal standard and D-chloroform (CDCl$_3$) as solvent. Molecular weight distribution (M_w and M_n) of a polymer product was measured using a gel permeation chromatography (GPC) system (Waters 515 HPLC Pump, Waters 2414 detector) with a set of three columns (Waters Styragel 500 A, 10,000 A, and 100,000 A). The GPC system was calibrated by a series of polystyrene standards with polydispersities of 1.02, which were supplied from Shodex Inc. THF (HPLC grade) was used as an eluent.

Differential scanning calorimetry (DSC) measurements were carried out under nitrogen flow on a Netzsch calorimeter (Model 204) from −30 to 100°C at a heating rate of 10°C/min. Thermogravimetric

analysis (TGA) measurements were performed in a PerkinElmer TGS-2 under a protective nitrogen atmosphere. The temperature ranged from 30 to 400°C with a heating rate of 20°C/min.

Tensile tests were performed at 23°C and relative humidity of 50% ± 5% using an automatic tensile tester (SANS-CMT) according to the ASTM E-104 standard. The cross-head speed was 10 mm/min. Five specimens of each sample were tested, and the average results were reported.

RESULTS AND DISCUSSION

Structural characterization

The synthesis of PPC and PPCN are depicted in Scheme 1. Figure 1 shows the ^{1}H NMR spectra of the resulting methanol-insoluble polymers. The ^{1}H NMR peaks (Fig. 1, top) indicate the existence of carbonate linkages and small fraction of random incor-
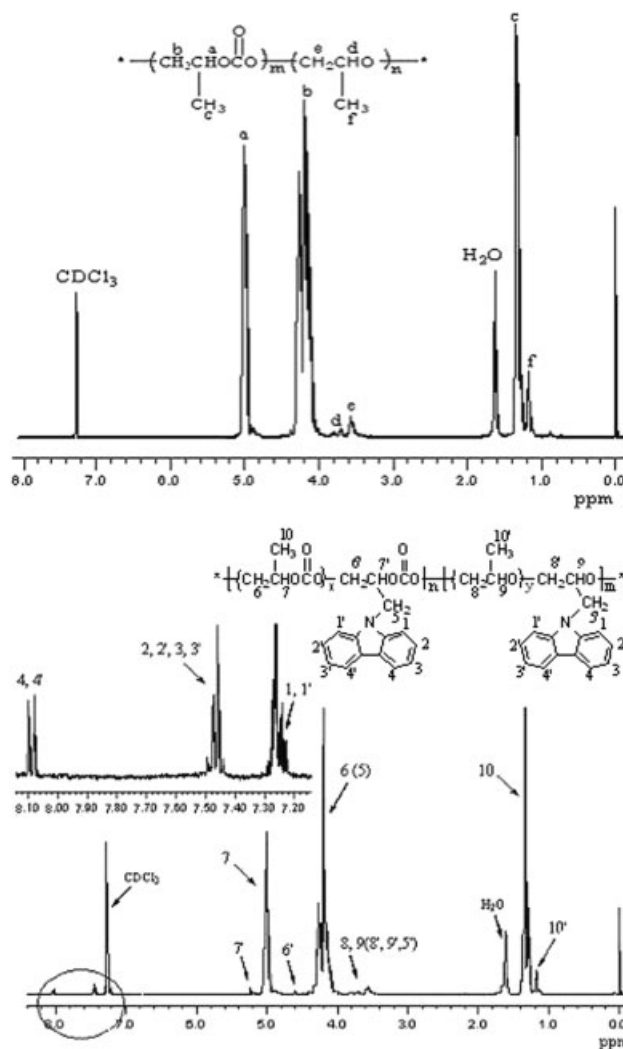

Figure 1 ^{1}H NMR spectra of the PPC polymer (top) and the PPCN terpolymer (bottom).

TABLE I
The Terpolymerization Results of CO_2 with PO and N-(2,3-Epoxylpropyl)Carbazole with s-ZnGA Catalyst[a]

NEC feed content (%)[b]	Yield[c]	M_n/M_w/MWD[d]	Composition of the polymer (molar fraction in %)[e]			
			f_{CMEC} (%)	f_{PC} (%)	f_{PE} (%)	f_{NEC} (%)
0	110	113 K/405 K/3.6		98.0	2.0	
0.8	92	98 K/304 K/3.1	0.2	96.1	3.5	0.4
2.0	84	92 K/258 K/2.8	0.4	94.4	4.6	1.0
4.3	60	74 K/280 K/3.8	0.9	92.8	5.0	2.2

[a] Polymerization was conducted at 60°C for 40 h under 5.2 MPa CO_2 pressure using 0.5 g of catalyst and 70 mL of PO.
[b] The molar fraction of the loaded NEC with respect to the total moles of NEC and PO.
[c] As g of polymer/g of cat.
[d] Determined by gel-permeation chromatography (GPC) and calibrated with polystyrene standards in tetrahydrofuran.
[e] Determined by ^{1}H NMR spectroscopy.

porated ether units in PPC backbone, suggesting that the resulting polymer from CO_2 and PO is almost alternating one. Figure 1 (bottom) shows a representative ^{1}H NMR spectrum for the PPCN terpolymer with a N-(2,3-epoxylpropyl)carbazole feed content of 2 mol % with respect to the total moles of N-(2,3-epoxylpropyl)carbazole and PO: ^{1}H NMR (δ, CDCl$_3$), 1.3 [3H, CH_3(10)], 4.2 [2H, CH_2CH(6)], 4.6 [2H, CH_2CH (6′)], 5.0 [1H, CH$_2$$CH$(7)], 5.3 [1H, CH$_2$$CH$ (7′)], 7.2 [2H, Ar—H (1,1′)], 7.5 [4H, Ar—H (2, 2′, 3, 3′)], 8.1 [2H, Ar—H(4,4′)], 1.2 [3H, CH_3 (10′)], 3.4–4.4 [protons in all ether linkages (5,5′, 8,8′ 9,9′)]. Compared with the ^{1}H NMR spectrum of PPC, the observed emergent peaks at 5.3, 4.6, and 7.2–8.1 ppm demonstrated that N-(2,3-epoxylpropyl)carbazole was successfully incorporated into the terpolymer backbone. These signals are assigned as the methine and methylene of [(9-carbazyl)methyl]ethylene carbonate units and aromatic protons in N-(2,3-epoxylpropyl)carbazole units.

Based on the ^{1}H NMR spectra, the compositions, including [(9-carbazyl)methyl]ethylene carbonate molar fraction (f_{CMEC}), propylene carbonate molar fraction (f_{PC}), PO ether molar fraction (f_{PE}) and the overall incorporated N-(2,3-epoxylpropyl)carbazole (both ester and ether units) molar fraction (f_{NEC}) of the resulted terpolymer can be estimated via the relative integration (A) of the correlated protons of the terpolymer according to the following equations.

$$f_{CMEC} = A_{5.3}/[A_{5.3} + A_{5.0} + (A_{1.2}/3) + (A_{7.2} - A_{5.3})/2] \times 100\% \quad (1)$$

$$f_{PC} = A_{5.0}/[A_{5.3} + A_{5.0} + (A_{1.2}/3) + (A_{7.2} - A_{5.3})/2] \times 100\% \quad (2)$$

$$f_{PE} = (A_{1.2}/3)/[A_{5.3} + A_{5.0} + (A_{1.2}/3) + (A_{7.2} - A_{5.3})/2] \times 100\% \quad (3)$$

$$f_{NEC} = (A_{7.2}/2)/[A_{5.3} + A_{5.0} + (A_{1.2}/3) + (A_{7.2} - A_{5.3})/2] \times 100\% \quad (4)$$

For PPC, the values of $A_{5.3}$ and $A_{7.2}$ are zero due to the absence of N-(2,3-epoxylpropyl)carbazole units in the PPC backbone.

The calculated results are summarized in Table I. It can be seen that PO, N-(2,3-epoxylpropyl)carbazole and CO_2 was successfully copolymerized using s-ZnGA as the catalyst. Thus PPCN terpolymers have number average molecular weight (M_n) ranging from 74,000 to 98,000 g/mol and molecular weight distribution (MWD) values ranging from 2.8 to 3.8. The MWD tends to become broader with decreasing M_n, and the catalytic activity decreased from 110 to 60 g polymer/g of cat. with increasing N-(2,3-epoxylpropyl)carbazole feed content from 0 to 4.3 mol % with respect to the total moles of N-(2,3-epoxylpropyl)carbazole and PO. The molar fractions of the overall N-(2,3-epoxylpropyl)carbazole content in the resulted terpolymers are all smaller than the corresponding N-(2,3-epoxylpropyl)carbazole feed content. From Figure 1, it can also be observed that N-(2,3-epoxylpropyl)carbazole in erpolymer existed in both carbonate linkages and ether linkages. But the CMEC content changed in a very limited range from 0.2 to 0.9 mol %. It is apparent that the total carbonate content decreased with increasing N-(2,3-epoxylpropyl)carbazole feed molar fraction. This implies that N-(2,3-epoxylpropyl)carbazole exhibited less reactivity than PO in the terpolymerization. Presumably, this can be explained by the fact that (9-carbazyl)methyl is a weaker electron donor compared with the methyl of PO. Strong electron-donor substituent at oxirane ring can result in an enhanced reactivity of the oxirane in the copolymerization.[2] On the other hand, the bulky substituent of N-(2,3-epoxylpropyl)carbazole is another reason to coordinate difficultly with metal at active site, which is necessary for oxiranes to insertion into metal-oxygen bond.[26]

The bulky aromatic rings of N-(2,3-epoxylpropyl)carbazole monomer can prevent the movements of the resulted terpolymer chains. Therefore, the physical properties such as Tg and thermal stability of PPC can be tailored by adjusting the N-(2,3-epoxylpropyl)carbazole content.
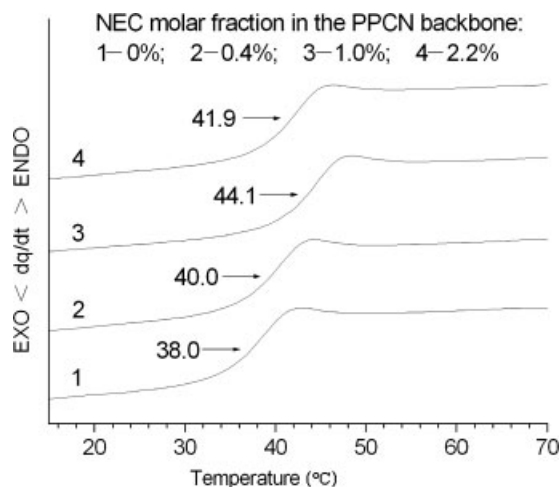
Figure 2 DSC thermograms for the PPCN terpolymers with different *N*-(2,3-epoxylpropyl)carbazole contents.

Improved thermal properties of PPCN

Figure 2 shows the DSC thermograms of PPCN terpolymers. It can be seen that the PPCNs exhibited higher T_g than PPC, although PPC has higher molecular weight than those of PPCN terpolymers. By adjusting the molar fraction of *N*-(2,3-epoxylpropyl)-carbazole in PPCN, the PPCN terpolymers with varying properties can be obtained as shown in Table II. Figure 3 shows the energy minimized *N*-(2,3-epoxylpropyl)carbazole molecular chain with bulky carbazyl groups. It can be imaged that the bulky carbazyl groups can restrict the rotation of the molecular chain, leading to an increase in T_g. The PPCN with 1.0 mol % *N*-(2,3-epoxylpropyl)carbazole moiety terpolymers showed the highest T_g of 44.1°C. Beyond this value, the T_g of the PPCN terpolymer decreased with further increasing *N*-(2,3-epoxylpropyl)carbazole content. This is due to the decrease in the molecular weight of the PPCN terpolymers.

Figure 4 shows the weight loss curves of the PPCN terpolymers. The 5% weight loss temperature ($T_{-5\%}$) appeared to increase dramatically with the incorporation of *N*-(2,3-epoxylpropyl)carbazole into the PPCN terpolymer. The $T_{-5\%}$ of the PPCN with 1.0 mol % *N*-(2,3-epoxylpropyl)carbazole was 40°C

TABLE II
Thermal and mechanical properties of the PPCN terpolymers

NEC content (%)[a]	T_g (°C)	$T_{-5\%}$ (°C)	Tensile strength (MPa)	Elongation at break (%)
0	38.0	238	31.7	544.1
0.4	40.0	270	33.7	26.1
1.0	44.1	278	38.2	15.7
2.2	41.9	265	34.9	6.1

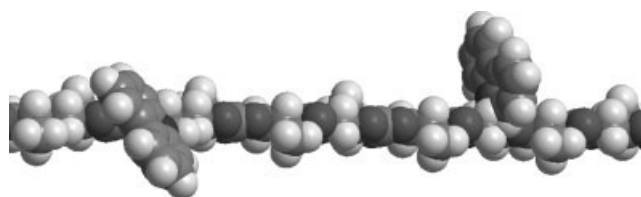[a] NEC molar fraction in the PPCN backbone.

Figure 3 Energy minimized space filling structure of PPCN segment with bulky carbazyl groups.

higher than that of PPC. The $T_{-5\%}$ of end-capped PPCs was about 240°C in nitrogen at a heating rate of 10°C min^{-1}.[18] The highly thermal stability made the PPCN terpolymer could be readily melt-processed at 150–180°C without decomposition. According to previous work,[27] the thermal decomposition of PPC obeys two kinds of mechanism, the main chain scission reaction and the unzipping reaction. While the unzipping reaction involves the backbiting of the terminal hydroxyl groups at the carbon of carbonate linkage leading to the formation of cyclic propylene carbonate. When *N*-(2,3-epoxylpropyl)carbazole units was introduced into PPC backbone, thus the formed bulky (9-carbazyl)methyl side groups can suppress the backbiting process and diminish the unzipping reaction. This effect is obviously favorable for increasing the thermal decomposition temperature. However, when *N*-(2,3-epoxylpropyl)carbazole content exceeded 1.0 mol %, the decrease in molecular weight became more obvious. Low molecular weight polymer has more hydroxyl terminal groups that can then accelerate the unzipping reaction, leading to low thermal decomposition temperature.

Mechanical properties

The mechanical properties of PPCN terpolymers were characterized in terms of tensile strength (σ_a) and elongation at break (ε). As shown in Table II, it can be seen that the tensile strength of the PPCN ter-
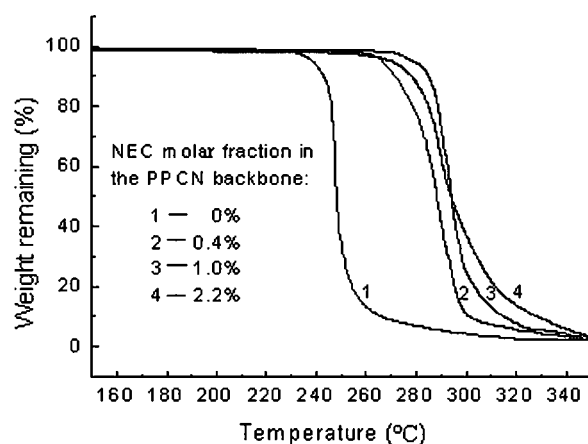

Figure 4 TGA curves for the PPCN terpolymers with different *N*-(2,3-epoxylpropyl)carbazole contents.

polymer with 1.0 mol % *N*-(2,3-epoxylpropyl)carbazole moiety increased to 38.2 MPa which was 6.5 MPa higher than that of PPC, together with an elongation at break of 15.7%. This indicates that the incorporation of *N*-(2,3-epoxylpropyl)carbazole led to the PPCN terpolymers more brittle due to the presence of bulky and rigid carbazyl groups.

CONCLUSIONS

N-(2,3-epoxylpropyl)carbazole monomer were successfully copolymerized with carbon dioxide and PO using supported zinc glutarate as catalyst. The resulted PPCN terpolymers exhibited greatly enhanced thermal and mechanical properties compared with PPC owing to the incorporation of the bulky carbazole pendants. But the addition of *N*-(2,3-epoxylpropyl)carbazole resulted in the decrease in the molecular weight of the PPCN terpolymers. The optimal properties of the terpolymers with T_g of 44.1°C and 5% weight loss temperature of 278°C, as well as high tensile strength of 38.2 MPa, were achievied at a *N*-(2,3-epoxylpropyl)carbazole content of 1.0 mol % in the terpolymer. This demonstrated that the bulky and rigid side chains were favorable to enhance the physical properties of the terpolymers.

Finally, very small amount of *N*-(2,3-epoxylpropyl)carbazole moiety in the synthesized terpolymer can greatly increase the thermal properties of PPC, resulting in a thermally stable terpolymer.

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