

Synthesis and Properties of Random Copolymers of 2,2-Dimethyltrimethylene Carbonate and Ethylene Carbonate Catalyzed by Lanthanide Tris(2,6-di-*tert*-butyl-4-methylphenolate)

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ABSTRACT: Random copolymers of 2,2-dimethyltrimethylene carbonate and ethylene carbonate (EC) were synthesized with lanthanide tris(2,6-di-*tert*-butyl-4-methylphenolate)s [Ln(DBMP)₃; Ln = La, Nd, Sm, or Dy] as catalysts, among which La(DBMP)₃ showed the highest activity. Poly(2,2-dimethyltrimethylene carbonate-co-ethylene carbonate)s [poly(DTC-co-EC)]s with high molecular weights were prepared at room temperature and characterized with ¹H-NMR and size exclusion chromatography. The thermal behavior and crystalline properties of the poly(DTC-co-EC)s were analyzed with differential scanning calorimetry, thermogravimetric analysis, and X-ray diffraction. The crystallinity and melting temperatures of

the poly(DTC-co-EC)s both decreased with increasing EC content in the copolymers. The mechanical properties of these copolymers were also investigated with dynamic mechanical analysis and tensile strength measurements, which revealed that a reduction of the glass-transition temperature and great enhancement of the tensile properties could be achieved with higher EC contents. These improvements in the thermal and mechanical properties indicate potential applications in biomedical research for novel polycarbonates. © 2009 Wiley Periodicals, Inc. *J Appl Polym Sci* 115: 46–51, 2010

Key words: copolymerization; polycarbonates

INTRODUCTION

In the past 2 decades, there has been considerable interest in aliphatic polycarbonates for widespread applications in medicine and pharmacy because of their high biocompatibility, facile biodegradation, low toxicity, and excellent mechanical properties.^{1,2} The most convenient method for preparing polycarbonates with high molecular weights and narrow molecular weight distributions seems to be the ring-opening polymerization (ROP) of cyclic carbonates.^{1,3,4} Quite a few catalysts have been successfully developed to catalyze the ROP of six-membered cyclic carbonates, such as trimethylene carbonate (TMC) and 2,2-dimethyltrimethylene carbonate (DTC).^{5–11} Copolymers of these carbonates have widespread applications

because of their adjustable crystallization properties, mechanical behavior, hydrolysis processes, degradability, and so forth.^{12–15}

Poly(ethylene carbonate) (PEC) is a unique polycarbonate known to possess a surface degradation mechanism via superoxide anions produced by the adhesion of polymorphonuclear leucocytes and macrophages.^{16–18} However, it is difficult to obtain PEC via ROP because of the positive enthalpy of ethylene carbonate (EC) homopolymerization, and the polymerization of EC usually proceeds at a high temperature (> 100°C) with partial decarboxylation and the elimination of carbon dioxide, which mostly depends on the choice of the initiator.¹ One convenient method for preparing polymers containing EC units is the ring-opening copolymerization of EC with other cyclic monomers such as ϵ -caprolactone and L-lactide. Different kinds of initiators have been developed for EC copolymerization at reasonably low temperatures, resulting in copolymers without the loss of carbon dioxide.^{19–21} To the best of our knowledge, no one has reported the copolymerization of EC with other cyclic carbonates in the literature.

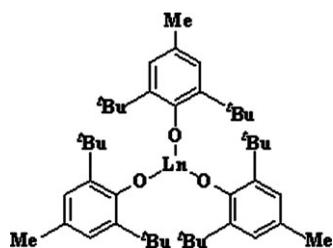
Recently, lanthanide complexes have attracted much interest as catalysts because of their well-defined structures. A number of lanthanide complexes have been found to be highly active for the ROP of cyclic esters and carbonates.^{11,22–27} Previously, our group

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Scheme 1 Chemical structure of $\text{Ln}(\text{DBMP})_3$ ($\text{Ln} = \text{La}, \text{Nd}, \text{Sm}, \text{or Dy}$).

reported the efficient activities of lanthanide aryl oxides for the homopolymerization and copolymerization of ϵ -caprolactone, TMC, and DTC.^{14,15,28–31} In this work, lanthanide tris(2,6-di-*tert*-butyl-4-methylphenolate)s [$\text{Ln}(\text{DBMP})_3$; $\text{Ln} = \text{La}, \text{Nd}, \text{Sm}, \text{or Dy}$; Scheme 1] were firstly employed as catalysts for the synthesis of poly(2,2-dimethyltrimethylene carbonate-*co*-ethylene carbonate) [poly(DTC-*co*-EC)]. Random copolymers with different EC contents were prepared through the control of the feed molar ratio of the comonomers. Thermal, crystalline, and mechanical properties of these copolymers were analyzed with differential scanning calorimetry (DSC), X-ray diffraction (XRD), thermogravimetric analysis (TGA), dynamic mechanical analysis (DMA), and tensile strength measurements in detail.

EXPERIMENTAL

Materials

DTC was prepared by the exchange reaction of 2,2-dimethylpropane-1,3-diol and diethyl carbonate and

was recrystallized before use.¹⁷ EC (Sinopec, Beijing, P.R., China; 99%) was recrystallized before use. $\text{Ln}(\text{DBMP})_3$ was prepared according to the methods used in our group.²⁸ Toluene was distilled over a blue benzophenone–sodium complex before use. Other reagents and solvents were purified in the usual way.

Copolymerization of DTC and EC

All copolymerizations were carried out in 20-mL ampules with the Schlenk technique. In a typical procedure, EC (0.264 g, 1.5 mmol), DTC (0.525 g, 3.5 mmol), and 4.8 mL of toluene were added to an ampule at room temperature (25°C). Then, 0.01 mmol of $\text{La}(\text{DBMP})_3$ in 0.2 mL of toluene was injected into the ampule with a syringe. After 20 h, the polymerization was terminated by ethanol with 5% HCl. The product was then precipitated in cold ethanol, filtered, and dried *in vacuo* at 60°C to a constant weight.

Characterization

The intrinsic viscosity (Shanghai Nirun Intelligent Technology Co. LTD Shanghai, P.R. China) of poly(DTC-*co*-EC) was measured with an Ubbelohde viscometer in CHCl_3 at $25.0 \pm 0.1^\circ\text{C}$. $^1\text{H-NMR}$ spectra were recorded on a Bruker Avance DMX 500-MHz spectrometer (Germany) in CDCl_3 with tetramethylsilane as an internal standard. Size exclusion chromatography (SEC) measurements were conducted on a Waters 208 apparatus (Milford, MA) with a Waters 2410 refractive-index detector in tetrahydro-

TABLE I
Copolymerizations of DTC and EC Catalyzed by $\text{Ln}(\text{DBMP})_3$

No.	Catalyst	DTC/EC in the feed (molar ratio)	Temperature (°C)	Yield (%)	EC content in the polymer (mol %) ^a	$[\eta]$ (dL/g)	M_n (10^4 g/mol) ^b	MWD (M_w/M_n) ^b
1	$\text{La}(\text{DBMP})_3$	100 : 0	25	94.4	0	0.87	10.2	1.58
2	$\text{La}(\text{DBMP})_3$	90 : 10	25	87.8	1.0	1.37	12.5	1.68
3	$\text{La}(\text{DBMP})_3$	70 : 30	25	75.7	3.3	1.89	23.6	1.47
4	$\text{La}(\text{DBMP})_3$	50 : 50	25	52.8	5.1	1.97	22.5	1.52
5	$\text{La}(\text{DBMP})_3$	30 : 70	25	28.4	7.5	1.81	—	—
6	$\text{La}(\text{DBMP})_3$	10 : 90	25	6.4	—	0.51	—	—
7	$\text{La}(\text{DBMP})_3$	0 : 100	25	—	—	—	—	—
8	$\text{Nd}(\text{DBMP})_3$	70 : 30	25	65.9	2.8	1.47	—	—
9	$\text{Sm}(\text{DBMP})_3$	70 : 30	25	61.3	2.6	1.14	—	—
10	$\text{Dy}(\text{DBMP})_3$	70 : 30	25	53.3	3.4	1.36	—	—
11	$\text{La}(\text{DBMP})_3$	70 : 30	−20	80.9	—	0.86	—	—
12	$\text{La}(\text{DBMP})_3$	70 : 30	100	28.0	—	0.31	—	—

The copolymerization was performed in toluene for 20 h with the following concentrations: $[\text{DTC}] + [\text{EC}] = 1.0$ mol/L and $[\text{Ln}(\text{DBMP})_3] = 2.0 \times 10^{-3}$ mol/L. η = intrinsic viscosity; M_n = number-average molecular weight; M_w = weight-average molecular weight; MWD = molecular weight distribution.

^a Measured from $^1\text{H-NMR}$ spectra, the EC content in the copolymers was calculated as follows: $\text{EC}(\%) = \frac{I_{\text{H}^c}}{I_{\text{H}^b} + I_{\text{H}^c} + I_{\text{H}^d}} \times 100\%$ where I_{H^b} , I_{H^c} , and I_{H^d} represent the intensities of H^b , H^c , and H^d , respectively, as shown in Figure 1.

^b Measured by SEC analysis.

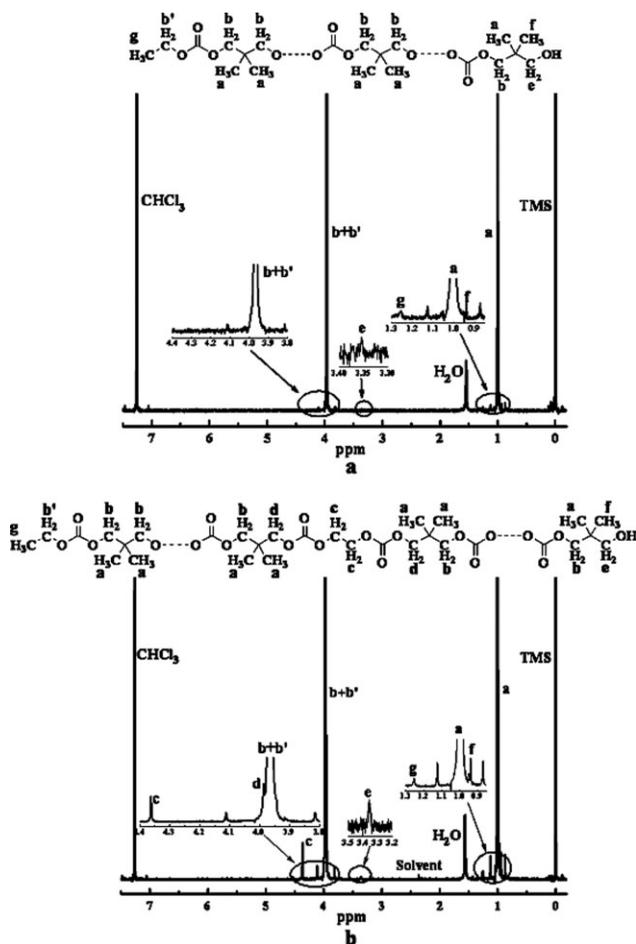


Figure 1 ¹H-NMR spectra of (a) PDTC (no. 1, Table I) and (b) poly(DTC-co-EC) (no. 2, Table I).

furan (1.5 mL/min) at 30.0°C, and commercial polystyrene standards were used for calibration.

Thermal and crystal analysis

DSC curves were determined on a PerkinElmer Pyris 1 apparatus (Waltham, MA). Each sample was heated from room temperature to 150°C at a heating rate of 20°C/min, cooled to room temperature at a cooling rate of 20°C/min, and then heated to 150°C again at a heating rate of 10°C/min. The thermostability of the copolymers was characterized with TGA on a PerkinElmer Pyris 6 apparatus with measurement temperatures ranging from 40 to 400°C at a heating rate of 20°C/min. The crystalline structure of poly(DTC-co-EC) was examined on a DMAX-RA X-ray diffractometer (BEDE, United Kingdom) with 12-kW Cu K α radiation ($\lambda = 1.5406 \text{ \AA}$). All samples were prepared into films (10 mm \times 10 mm \times 0.10 mm) by the solvent-casting method.

Mechanical analysis

To obtain homogeneous samples for mechanical analysis, the polymer powder was compression-molded at 140°C with a pressure of 1.5×10^7 Pa for 15 min and subsequently cooled to room temperature under this pressure. The samples for DMA were rectangular (30 mm \times 12 mm \times 1.5 mm). The measurements were carried out on a Netzsch DMA 242 analyzer (Germany) under a nitrogen atmosphere employing a sample holder of a single cantilever (16 mm) with a typical frequency of 1.0 Hz and a scanning rate of 3.0°C/min from -150 to $+150$ °C. The tensile strength measurements were tested on a Reger RGT-X010 universal testing machine (Shenzhen, P.R. China) at 25°C with a tensile rate of 2 mm/min. Each sample was prepared to be dumbbell-shaped according to ASTM D 638-02. For each data point, three samples were tested, and the average value was taken.

RESULTS AND DISCUSSION

Random copolymerization of DTC and EC

The random copolymerization of DTC and EC was first carried out with lanthanide aryl oxides [Ln(DBMP)₃; Ln = La, Nd, Sm, or Dy] as single-component catalysts, with the results presented in Table I. The copolymers prepared with La(DBMP)₃ exhibited higher yields and intrinsic viscosities than those prepared with the other lanthanide complexes (nos. 3 and 8–10, Table I) under the same conditions; this implies that La(DBMP)₃ had the highest catalytic activity. This result coincides with the activity

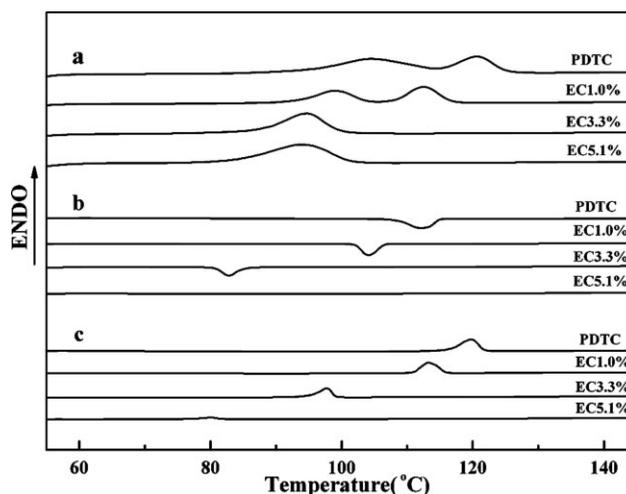


Figure 2 DSC curves of poly(DTC-co-EC): (a) first heating run, (b) cooling run, and (c) second heating run. The curves labeled PDTC, EC1.0%, EC3.3% and EC5.1% represent polymers 1–4 in Table I, respectively.

TABLE II
Thermal Data for Poly(DTC-co-EC)s with Different EC Contents by DSC and TGA

No.	EC content in the polymer (mol %)	M_n (10^4 g/mol) ^a	T_c (°C) ^b	ΔH_c (J/g) ^b	T_m (°C) ^c	ΔH_m (J/g) ^c	T_d (°C) ^d
1	0	10.2	112.2	-21.9	119.8	22.5	248.9
2	1.0	12.5	104.1	-21.5	113.2	22.1	266.5
3	3.3	23.6	83.9	-16.8	97.7	17.2	271.7
4	5.1	22.5	—	—	80.1	2.3	278.3

ΔH_c = enthalpy of crystallization; ΔH_m = enthalpy of melting; M_n = number-average molecular weight; T_c = crystallization temperature; T_d = initial decomposition temperature; T_m = melting temperature.

^a Measured by SEC analysis.

^b Measured from the cooling run of DSC [Fig. 2(b)].

^c Measured from the second heating run of DSC [Fig. 2(c)].

^d Measured by TGA (Fig. 3).

sequence of lanthanide complexes catalyzing ROP of cyclic carbonates in other reports.^{27,28} With an increasing amount of EC in the feed (nos. 1–7, Table I), the yields of the copolymerization decreased, whereas the EC contents in the copolymers (calculated with the intensity ratio of protons in ¹H-NMR) increased. However, the EC contents were reasonably low (only up to 7.5%; no. 5, Table I), and this confirms that it is difficult for the EC monomer to undergo ROP, as mentioned previously. The effect of the polymerization temperature on the copolymerization catalyzed by La(DBMP)₃ is also summarized in Table I (nos. 3, 11, and 12). It seems that a moderate temperature (25°C) is favorable for the copolymerization of DTC and EC with respect to both the yield and the intrinsic viscosity.

Typical ¹H-NMR spectra of poly(2,2-dimethyltrimethylene carbonate) (PDTC; no. 1, Table I) and poly(DTC-co-EC) (no. 2, Table I), along with the assignments of the detected signals, are illustrated in Figure 1. The small peak of H^d around 4.0 ppm is the characteristic signal of the last -CH₂- group in DTC segments next to the -OCOO- group of the EC unit. The intensity of H^d is approximately that of H^c assigned to the EC unit, and this implies that all EC units are singly embedded in the DTC segments and that no EC-EC unit exists; this proves that the product was a random copolymer. Besides the signals of the copolymer backbone, the singlet signals detected at 3.35 (H^e) and 0.94 ppm (H^f) show that the -C(CH₃)₂CH₂OH end group structures of the copolymer resulted from the DTC unit. The signal appearing at 1.25 ppm (H^g) is assigned to the end group of CH₃CH₂OCO-, which originated from ethanol. Therefore, both DTC and EC were inserted into the growing chains with acyl-oxygen bond cleavage, and the ethoxy group was incorporated into the growing chain during the termination stage, as previously reported by our group.^{28–31}

Usually, the polymerization of EC is accompanied by decarboxylation, which most likely involves metal carbonate species formation due to ring opening via C-O bond cleavage.^{1,21,32} The absence of a signal between 3.10 and 3.30 ppm indicates that the polymers obtained with Ln(DBMP)₃ are free of ether units developed from carbon dioxide elimination of EC or DTC, and this indicates that these lanthanide complexes are efficient catalysts for the synthesis of aliphatic polycarbonates at reasonably low temperatures.^{28,29}

The SEC measurements showed that poly(DTC-co-EC)s with high molecular weights as well as reasonably narrow molecular weight distributions were successfully prepared with La(DBMP)₃. The single peak of the SEC pattern revealed that there was only one kind of active species during the copolymerization and that the products were copolymers rather than blends of PDTC and PEC.

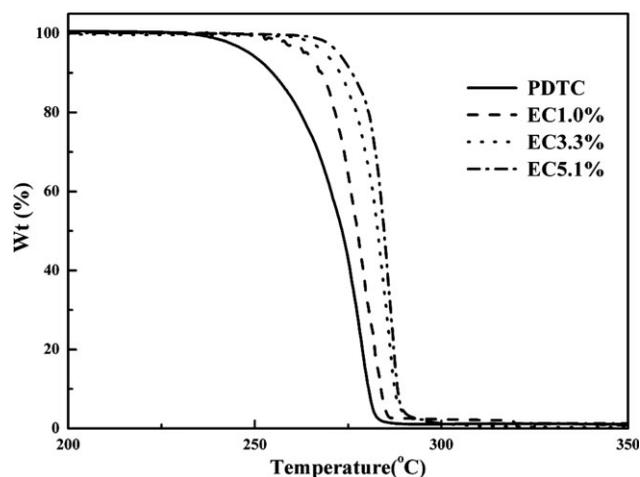


Figure 3 TGA curves of poly(DTC-co-EC). The curves labeled PDTC, EC1.0%, EC3.3%, and EC5.1% represent polymers 1–4 in Table I, respectively.

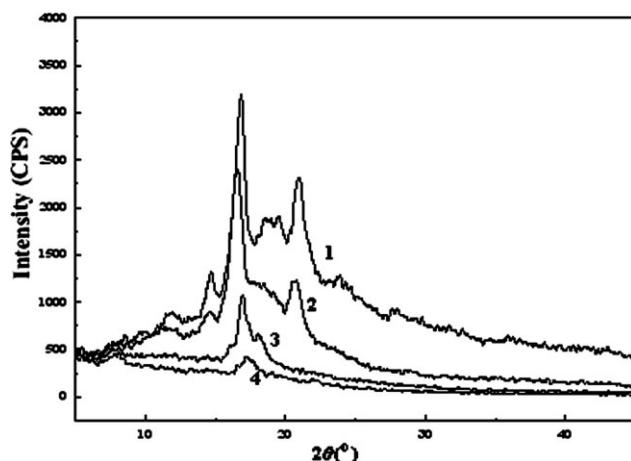


Figure 4 XRD curves of poly(DTC-co-EC). The curves labeled 1–4 represent polymers 1–4 in Table I, respectively.

Thermal and crystal behaviors of poly(DTC-co-EC)

Thermal and crystal behaviors of poly(DTC-co-EC) were investigated with DSC, TGA, and XRD. Figure 2 shows the DSC curves as a function of the EC contents in the copolymer, and all the melting and crystallization data are listed in Table II. With increasing EC contents, both the temperatures and enthalpies of the copolymer melting and crystallization transitions decreased, and no transition was detected in the cooling run of the copolymer with an EC content of 5.1% (no. 4, Table II). In addition, two endothermic peaks appeared in the first heating run [Fig. 2(a)] of PDTC and the copolymer with an EC content of 1.0%, indicating that there were two types of crystalline forms in these polymers, as reported by Meille et al.³³ As for the copolymers with higher EC contents (EC3.3% and EC5.1%), only one endothermic peak was detected in the first heating run, and this suggested that there was only one crystalline form in the copolymers with relatively high EC contents.

Figure 3 displays TGA curves of poly(DTC-co-EC)s with different EC contents. All the polymers presented reasonably good thermostability because no significant weight loss ($\sim 0.5\%$) occurred until

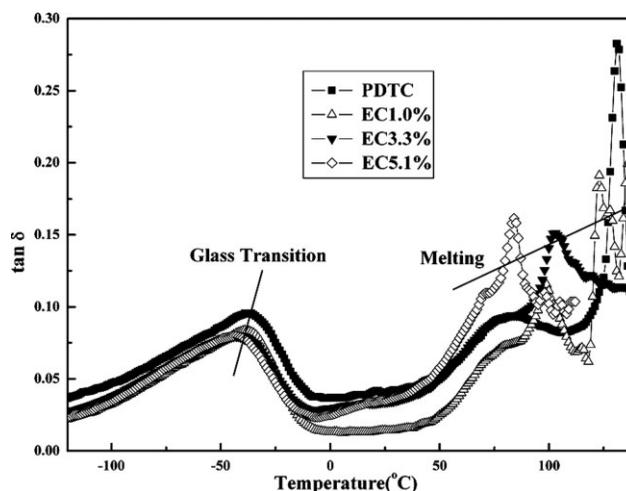


Figure 5 Tan δ of poly(DTC-co-EC). The curves labeled PDTC, EC1.0%, EC3.3%, and EC5.1% represent polymers 1–4 in Table I, respectively.

230°C. With increasing EC contents, the copolymers exhibited higher initial decomposition temperatures (Table II) with enhanced thermostability. All these polycarbonates decomposed quickly beyond the temperature of 280°C and lost almost their entire weight ($> 99\%$) around 300°C.

Figure 4 shows the XRD results for poly(DTC-co-EC)s with different EC contents. With increasing EC contents, the intensity of each main diffraction peak of PDTC ($2\theta = 15.4, 17.7, 19.6,$ and 20.5°) decreased a lot, and this showed that there was lower crystallization in the copolymers, which matched the results of DSC analysis quite well.

Mechanical behavior of poly(DTC-co-EC)

The mechanical behavior of poly(DTC-co-EC) was analyzed with DMA and tensile strength measurements. Figure 5 shows phase angle tan δ /temperature curves for poly(DTC-co-EC) measured by DMA. Two main peaks corresponding to the glass-transition and melting temperatures of the polymers can be observed in each curve, as summarized in Table III.²¹ Both the glass-transition and melting

TABLE III
Thermal and Mechanical Properties of Poly(DTC-co-EC)s with Different EC Contents

No.	EC content in the polymer (mol %)	T_g ($^\circ\text{C}$) ^a	T_m ($^\circ\text{C}$) ^a	Tensile strength (MPa) ^b	Tensile modulus (MPa) ^b	Elongation at break (%) ^b
1	0	-37.2	131.3	23.0 ± 1.7	1.9 ± 0.4	148.8 ± 3.2
2	1.0	-38.7	122.8	138.7 ± 3.6	11.4 ± 1.3	180.0 ± 4.1
3	3.3	-40.5	102.5	224.6 ± 3.4	17.8 ± 1.1	197.7 ± 3.7
4	5.1	-43.7	84.0	203.9 ± 3.1	16.3 ± 1.0	307.6 ± 3.4

T_g = the glass-transition temperature; T_m = melting temperature.

^a Measured by DMA (Fig. 5).

^b Measured from tensile strength measurements.

temperatures of the copolymers declined with increasing EC contents in the polymer, and this reconfirmed that the rigidity and regularity of the PDTC segments were lowered by the incorporation of EC units.

Mechanical properties such as the tensile strength, tensile modulus, and elongation at break of these polymers were also measured, and they are listed in Table II. All these properties were enhanced significantly with increasing EC incorporation. The copolymer with an EC content of 5.1% exhibited both high tensile strength and excellent elongation at break, and this suggests that this polymer may be widely applicable for materials in biomedicine and tissue engineering.

CONCLUSIONS

In this study, random copolymerizations of DTC and EC were successfully carried out with Ln(DBMP)₃ complexes as catalysts. La(DBMP)₃ exhibited the highest catalytic activity. Random copolymers of DTC and EC with high molecular weights and reasonably narrow molecular weight distributions were prepared. The mechanical properties of these copolymers were significantly enhanced with increasing EC contents.

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