

Reactive Extrusion of Recycled Polycarbonate Using Chain Extenders

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Received May 11, 2022; revised August 8, 2022; accepted September 20, 2022

Abstract—Electronic waste (e-waste) problem becomes more critical due to increasing demand on upgrading of electrical-electronic devices in accordance with the latest technology nowadays. Polycarbonate (PC) is one of the most used plastics to produce computers, mobile phones and televisions thus recovering of PC wastes to develop new value-adding products has been an important task. The challenge of the PC recycling is thermal degradation under multiple thermo-mechanical processing cycles, which deteriorates molecular weight and properties of the polymer. In this study, a multi-functional anhydride chain extender (ANHY) and a multi-functional epoxy chain extender (EPOX) were used to offset thermal degradation of recycled PC (rPC) during extrusion. The effect of chain extenders on the properties of resulting materials was evaluated by rheology, tensile test, thermogravimetric analysis (TGA), dynamic mechanical analysis (DMA) and differential scanning calorimetry (DSC). Both types of chain extenders enhanced the viscoelastic and mechanical properties of rPC, however ANHY had much higher chain extension reactivity than EPOX. ANHY increased complex viscosity by 58% and tensile modulus by 39%, whilst EPOX showed 26% increment in complex viscosity and 10% enhancement in tensile modulus on the base rPC. The formation of long chain branching in the samples containing chain extenders was observed from the rheology tests. TGA results also confirmed the efficiency of ANHY chain extender that shifted decomposition temperature of rPC to higher value than that of EPOX. The incorporation of chain extenders slightly increased glass transition temperature (T_g) of rPC in DMA and DSC test results.

Keywords: polycarbonate, recycling, chain extender, extrusion, rheology

DOI: 10.1134/S1990793123010281

INTRODUCTION

Polycarbonate (PC) is an engineering thermoplastic mainly synthesized by condensation reaction between bisphenol A (BPA) and either phosgene or diphenyl carbonate [1]. PC possesses many desirable properties such as optical transparency, outstanding mechanical properties, and ease of processing which make it ideal polymer for construction, automotive and, electronics applications [2]. The electrical-electronic, where PC is widely used to produce computers, mobile phones, compact discs, has become one of the largest industries in the world [3]. However, short service life and demand on upgrading of these products in accordance with the latest technology cause discarded electronics that can be considered as the fastest growing waste stream and a global problem recently [4, 5].

Today, plastics industry moves from its conventional linear economy to a more strategic and sustainable model, circular economy, which is being strongly promoted by governments [6]. The strict legislations are being enforced researchers and businesses to develop sustainable polymer technologies that offer

less plastic waste, greenhouse gas emissions, energy consumption and use of natural resources [7]. Such needs address to plastic recycling technology, which also appears to be one of the best solutions for recovering PC from electronic wastes (e-wastes) [8]. However, mechanical recycling of condensation polymers such as PC has some difficulties when exposed to high shear stress and temperature in the repeated injection molding and extrusion processing cycles [9]. Thermo-mechanical processing induces severe degradation to the PC that breaks down the polymer chains into shorter chains, leading to lower molecular weight [10, 11]. The decrease in molecular weight causes impairment of the many properties such as mechanical and rheological, therefore restricts extensive usage of recycled PC (rPC) into engineering applications [12].

Previous studies reported that thermal degradation of PC occurred as chain scission of carbonate ester linkage which was eventually resulted into phenolic compounds such as phenol and isopropenylphenol with hydroxyl end groups [13–15]. A solution to this problem can be addressed by melt processing of rPC with chemical compounds called chain extenders.

Table 1. Chemical structures of the chain extenders used in this study

Chain extender	Chemical structure
Joncryn ADR 4300	
Joncryn ADR 3400	

Recently, solid-state polymerization (SSP), one of the most used techniques to build back the molecular weight of polycondensates, has been replaced by chain extenders due to the long polymerization time and expensive equipment requirements of SSP [16, 17]. The principle of chain extension method is that the functional groups of chain extender react with hydroxyl ($-\text{OH}$), carboxyl ($-\text{COOH}$) and amine ($-\text{NH}_2$) terminal end groups of polycondensates during conventional extrusion process [6, 16]. The recoupling of the split polymer chains by di- or multifunctional chain extenders has been known as chain extension and this method allows tailored solutions to improve molecular weight and properties of degraded polycondensates [16].

The use of chain extenders to restore properties of recycled poly(ethylene terephthalate) (PET), poly(lactic acid) (PLA) and polyamide-6 (PA-6) has been widely investigated and examples of chain extenders used in earlier studies were diisocyanates [18, 19], dianhydrides [20–23], diepoxides [24], bisoxazolines [25, 26], phosphites [27, 28], multi-functional epoxy oligomer [29–36] and multi-functional anhydride oligomer [37]. However, there is scarce research on chain extension of PC in the literature. Several studies [38–40] explored reactive extrusion of recycled PET and PC blends using diisocyanate and multi-functional epoxy chain extenders. These studies showed that notable benefit of chain extender to improve molecular weight of recycled PET and compatibility of the blends. Nevertheless, they did not extend to thermal degradation of rPC. So far, a multi-functional epoxy chain extender (Joncryn ADR 4300) was used to restore properties of rPC such as CD

scraps [41]. The study revealed that incorporation of chain extender at 1 wt % concentration during extrusion led to 52% lower melt flow index of rPC. Although this study confirmed applicability of the method using small quantities of chain extender without the requirements of catalyst and vacuum, more research is required to shed light on chain extension mechanism and effect of different chain extenders on the properties of rPC. Type of chain extender is the first critical point here as it affects directly yield of reaction and therefore performance of the end-product. Since dianhydrides were suggested to be more reactive than epoxy functional chain extender with hydroxyl terminated chains of polyesters [28, 42, 43], no studies have been carried out using anhydride functional chain extenders to recouple hydroxyl chain ends of rPC. Thus, this research has attempted to investigate effects of a multi-functional anhydride chain extender (Joncryn ADR 3400) and a multi-functional epoxy chain extender (Joncryn ADR 4300) on the rheological, mechanical, and thermal properties of rPC.

EXPERIMENTAL

Materials

Polycarbonate used in the experimental study is Makrolon 2605 supplied from Covestro with a melt volume flow rate of $12 \text{ cm}^3/10 \text{ min}$ (300°C) and a density of 1200 kg/m^3 . Chain extenders, Joncryn ADR 4300 (EPOX) and Joncryn ADR 3400 (ANHY), are manufactured by BASF and presented in Table 1. They show remarkable differences in their structures (epoxy to anhydride), densities (1080 –to 600 kg/m^3), molecular weights (6800 –to $10\,000 \text{ g/mol}$), glass transition temperatures (54 – 133°C).

Sample Preparation

In the study, a co-rotating Baker Perkins APV MP19TC twin screw extruder with a screw diameter of 19 mm ($L/D = 28$) with five heating zones was used. To simulate thermo-mechanical degradation of PC during the extrusion, virgin PC granules were initially extruded once, and the resulting product was used as rPC. The extruder was operated using temperature profile of 240 – 255 – 260 – 265 – 265°C from hopper to the die at a rotation speed of 150 rpm . After extrusion, the extrudate in the form of filament was pulled from the die, cooled in a water bath, and finally passed through the pelletizing unit to form rPC granules. rPC granules and 1 wt % chain extender were then fed simultaneously from two separate feeders to extruder screws with following same procedure in the previous recycling process stage. Prior to extrusion, the polymer and chain extender were dried under vacuum at 90°C for 12 h. To prepare molded plates for characterization of the samples, a hydraulic hot press was used. The extruded polymer granules were molded at

260°C, under 50–300 MPa pressure range for 4 min, and then cooled to 30°C, at 300 MPa pressure for 2 min.

Characterization

Rheological tests were performed on Anton Paar Physica MCR 501 rotational rheometer with 25 mm diameter parallel plate flow geometry and 1-mm gap size at 260°C. The amplitude sweep tests were initially applied to determine linear viscoelastic region of the samples and strain of 0.3% was found to be suitable. Thereafter at this strain, frequency sweep tests were performed over a frequency range of 0.1–100 Hz.

Tensile test specimens were obtained by die cutting of the molded plates in accordance with ASTM D638-10 type I test specimen standard (narrow section 57-mm long × 13 mm wide × 3 mm thick). The tests were carried out by using an Instron 5564 tensile test machine with a cross-head speed of 50 mm/min at room temperature. At least 5 tests were performed for each sample group and average values with standard deviations were recorded.

Differential scanning calorimetry (DSC) measurements were carried out using a TA Instruments Q20 DSC under nitrogen atmosphere. The samples of approximately 4 mg were placed in aluminum standard pans and heating/cooling/heating cycles (25 → 250 → 25°C) at a rate of 10°C/min were applied. As the response from the first heating cycle is generally influenced by thermal processing history of the samples, the data in the second heating cycle was used [44, 45].

Thermogravimetric analysis (TGA) was conducted to investigate thermal decomposition of the samples by using a Hitachi Exstar SII TG/DTA 7300 thermal analyser. The samples of approximately 10 mg weight were heated from 50 to 600°C at a heating rate of 10°C/min under nitrogen flow rate of 50 mL/min.

Dynamic mechanical analysis (DMA) was performed on a TA Instruments Q800 DMA analyzer operating in the tension mode at a frequency of 1 Hz to determine temperature dependent dynamic mechanical properties of the samples. The test specimens were cut from the molded plates with dimensions of 57 mm long, 13 mm wide, 3 mm thick. The tests were done in the temperature range from 35 to 185°C with a heating rate of 3°C/min.

RESULTS AND DISCUSSION

Possible Reactions between rPC and Chain Extenders

As explained in the introduction section, chain extenders aimed to react phenolic hydroxyl end groups of rPC occurred by scission of carbonate linkages [13–15]. In Fig. 1a, the chain extension reaction involves maleic anhydride ring-opening of ANHY by nucleophilic attack of hydroxyl end groups of rPC on carbon atoms of the chain extender which eventually results in

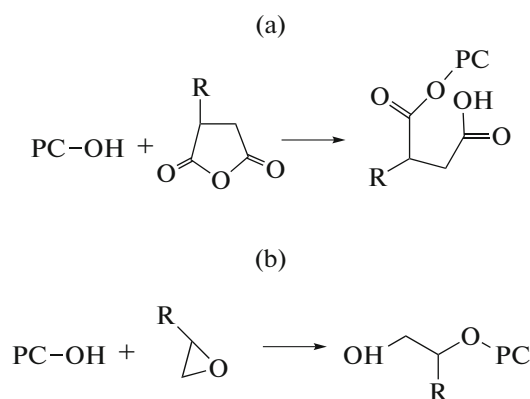


Fig. 1. Chain extension mechanisms of PC with (a) ANHY, (b) EPOX.

the formation of acidic compound [46]. On the other hand, the chain extension reaction appears as epoxy ring opening of EPOX by abstraction of hydrogen atom from hydroxyl group of rPC in Fig. 1b. It should be noted that both types of chain extenders have multiple reactive groups which enable to link with several polymer chains therefore branching in polymer structure may also occur [47].

Rheology

Rheology is a versatile tool to observe molecular changes in polymers and was used to investigate chain structure of PC such as chain scission, extension and branching in this study. Figure 2 presents storage modulus (G') of the samples as a function of frequency. Storage modulus reflects elasticity of a polymer and sensitive to structural changes therefore any changes with thermal degradation and incorporation of chain extenders can be traced from the samples [48]. For example, at 1 Hz frequency rPC/ANHY and rPC/EPOX indicated higher storage modulus values of 10640 and 7496 Pa respectively compared to rPC (5668 Pa) which could be attributed to formation of chain branching and enhancement of chain entanglement density. Due to multi-functional feature of chain extenders, several polymer chains might be recoupled chemically with a formation of branched structure and therefore enhanced melt elasticity. Compared to rPC/EPOX, the higher storage modulus of rPC/ANHY can be explained by chemical reactivity of anhydride with hydroxyl groups. This phenomenon is consistent with the results of earlier studies reported for anhydride chain extenders and hydroxyl chain end groups of PET [28, 42].

According to Fig. 3, the effect of thermo-mechanical degradation during recycling could be clearly observed from rPC. A complex viscosity of 1449 Pa s was measured for rPC compared to 3505 Pa s for virgin PC at a frequency of 1 Hz. The chain extenders com-

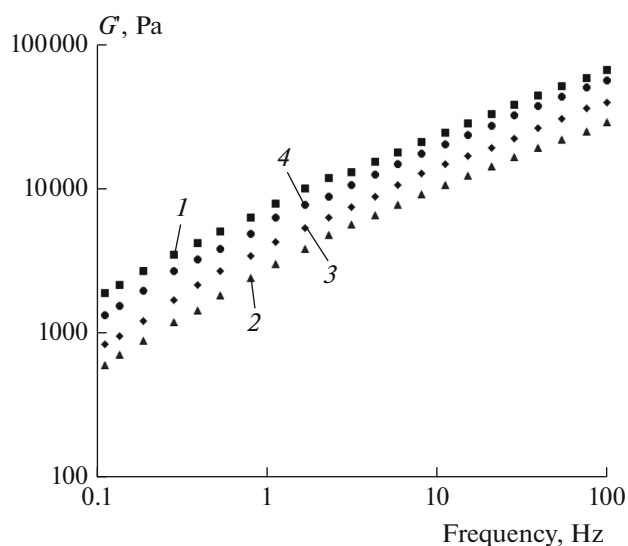


Fig. 2. Storage modulus (G') versus frequency: (1) Virgin PC; (2) rPC; (3) rPC/EPOX; (4) rPC/ANHY.

pensated degradation and increased the viscosity of rPC, which was more pronounced at lower frequencies. With regard to the type of chain extender, the complex viscosity values of rPC/ANHY (2291 Pa s at 1 Hz) and rPC/EPOX (1836 Pa s at 1 Hz) confirmed that multi-anhydride functional chain extender more effectively rebuilt the structure of rPC. From the figure, a striking Newtonian plateau can be observed for virgin PC and rPC, which could be attributed to linear chain structure of the polymer. Contrary to that, Newtonian plateau tended to gradually decrease for sam-

ples containing chain extender with a typical shear thinning behavior even at lower frequencies and this was more pronounced for rPC/ANHY [49]. Higher chain entanglements accompanied by chain extension and branching might be reason of such phenomenon [50, 51].

The data of loss angle (δ) as a function of frequency is shown in Fig. 4. In this figure, the curves of both rPC and virgin PC indicated plateau particularly at lower frequencies with higher loss angle closely 90° . Such behavior can be considered as a characteristic behavior of linear polymers, as samples have lower chain entanglement density and less rigid structure therefore the maximum loss angle value at 90° was reached and a distinct relaxation process with a plateau was occurred [52–54]. On the other hand, the chain extenders led to decrease phase angle of rPC/ANHY and rPC/EPOX along the frequency due to presence of long chain branching and higher molecular weight. This can be related to more elastic structure and higher chain entanglements of the samples modified with chain extenders therefore the polymer chains relaxed more slowly and loss angle linearly decreased with frequency without approaching the maximum [52, 53]. These results eventually evidenced the structural changes of rPC with incorporation of chain extenders.

Tensile Properties

It is known that the polymer structure significantly influences the mechanical properties therefore tensile test is an important tool to observe changes in the

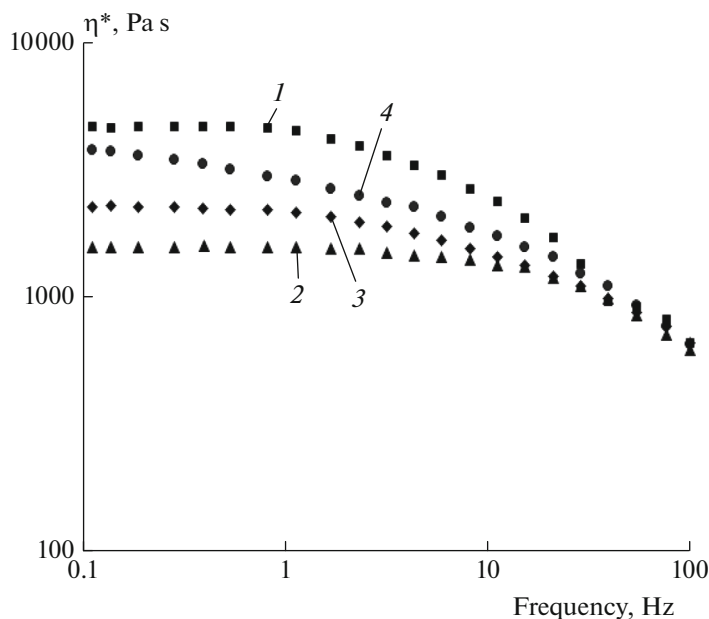


Fig. 3. Complex viscosity (η^*) versus frequency: (1) Virgin PC; (2) rPC; (3) rPC/EPOX; (4) rPC/ANHY.

structure of the samples due to thermal degradation and chain extension [55, 56]. Table 2 shows tensile modulus, yield strength, and yield strain data of the samples. As can be seen from Table 2, rPC showed the lowest tensile modulus and yield strength values. Such decrease can be attributed to thermal degradation and mechanical shear forces during recycling which resulted into cleavage of chains and lower molecular weight of the polymer [57]. With incorporation of chain extenders, mechanical properties of rPC were noticeably improved and clearly indicated recoupling of degraded chains with structure reinforcement. Anhydride functional chain extender was found to be more effective, increased tensile modulus of the rPC by 39%, whilst the epoxy functionalized type showed relatively small enhancement of 10%. Similar effect of chain extenders on the samples can also be observed from the following values of yield strength, a 32% increase for rPC/ANHY and a 12% increase for rPC/EPOX. Higher tensile modulus and yield strength data of rPC/ANHY sample can be explained by rigid structure of multi-anhydride functional chain extender and increased molecular weight, which limited chain mobility of the polymer [58].

As shown in Table 2, both types of chain extenders increased yield strain of rPC, where rPC/ANHY showed relatively higher strain values. This may be explained that enhancement of molecular weight and chain branching could lead to a higher degree of entanglement density and therefore samples become more resistant to mechanical deformation [52]. On the other hand, rPC presented least yield strain that can be attributed to reduction in ductility, mainly due to chain scission and lower molecular weight [6, 59]. Such results showed a reasonable agreement with rheology data and backed up our argument that multi-anhydride functional chain extender is more efficient than epoxy functionalized one.

Thermogravimetric Analysis (TGA)

TGA curves of the samples are shown in Fig. 5 and the onset temperature of degradation (T_{onset}) was evaluated as the degradation temperature at 5% weight loss [6, 15]. As can be inferred from Table 3, rPC indicated a lower T_{onset} than virgin PC due to thermo-mechani-

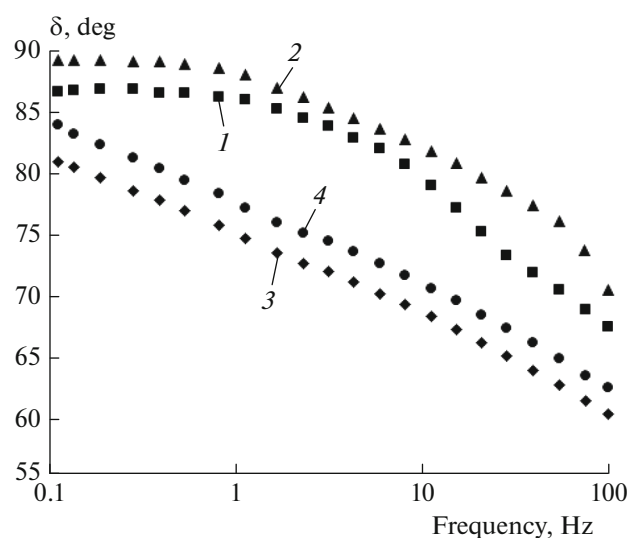


Fig. 4. Loss angle (δ) versus frequency: (1) Virgin PC; (2) rPC; (3) rPC/EPOX; (4) rPC/ANHY.

cal degradation during mechanical recycling. It has been reported that cleavage of polymer chains had profound effect on lower thermal stability of recycled polymers [37, 60]. In the case of PC, this can be attributed to chain scission of carbonate ester linkage that resulted into phenolic compounds with hydroxyl chain end groups. On the other hand, both two types of chain extenders shifted T_{onset} of rPC from 434°C to following values, 437°C for rPC/EPOX and 450°C for rPC/ANHY due to recoupling of the degraded chains. The efficiency of multi-anhydride functional chain extender to compensate thermal degradation of rPC can be explained by higher reactivity of the chain extender with the hydroxyl groups of rPC.

Differential Scanning Calorimetry (DSC)

Figure 6 presents DSC thermograms of the obtained samples. As a typical thermal behavior of amorphous PC, all samples indicated merely a second order transition in the thermograms that can be referred as glass transition temperature (T_g) and did not show crystallization peaks [2]. Like previous studies [59, 61, 62], small changes in T_g values of samples

Table 2. Tensile properties of the samples

Sample	Tensile modulus, MPa	Yield strength, MPa	Yield strain, %
Virgin PC	2418.7 ± 38.4	68.5 ± 1.4	6.5 ± 0.1
rPC	1706.1 ± 25.6	49.3 ± 0.5	4.7 ± 0.1
rPC/EPOX	1877 ± 22.9	55.4 ± 1.1	5.2 ± 0.2
rPC/ANHY	2363.2 ± 31.1	65.3 ± 0.9	6.1 ± 0.1

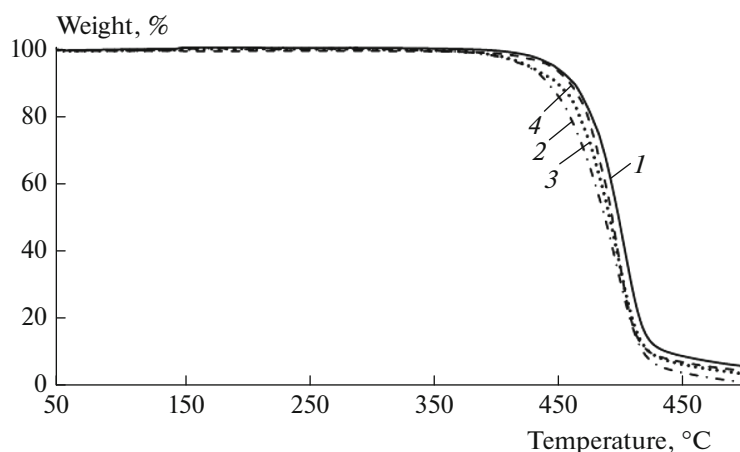


Fig. 5. TGA curves of the samples: (1) Virgin PC; (2) rPC; (3) rPC/EPOX; (4) rPC/ANHY.

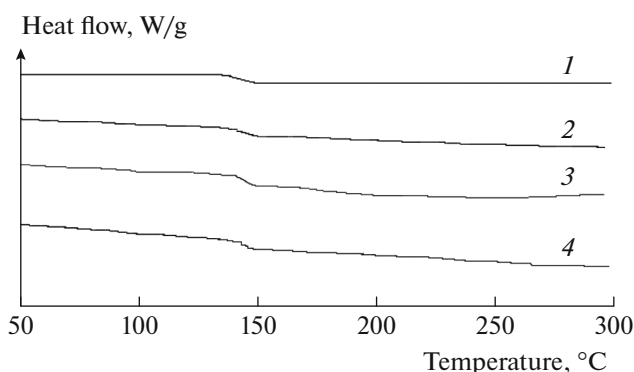


Fig. 6. DSC thermograms of the samples: (1) Virgin PC; (2) rPC; (3) rPC/EPOX; (4) rPC/ANHY.

were observed in Table 3. The chain extenders slightly increased T_g of rPC that can be attributed to lower chain mobility and free volume of the samples due to chain extension and higher molecular weight [37, 63].

In general, the results showed that the difference in T_g values of the samples can hardly be evaluated as significant. The glass transition temperature can be considered as independent of processing and extender type. One should be noted that samples modified with chain extenders did not show T_g of the chain extenders which were supposed to be at around 54 and 133°C in DSC thermograms. This indicated that the chain

extenders entirely reacted with chain end groups of rPC and consumed in the reaction.

Dynamic Mechanical Analysis (DMA)

DMA is a useful tool to investigate the changes in the structure of the materials over a wide temperature range. According to Fig. 7, the storage modulus (E') of the samples remarkably decreased with temperature, which can be explained by transition of the samples from glassy to rubbery state. The storage modulus curves were overlapped at elevated temperatures since the samples were in the rubbery state and the variations in elastic behavior become less prominent by further increase in temperature. Table 4 shows the values of storage modulus obtained at 35°C to better represent elastic properties of the samples. The chain extended samples indicated higher storage modulus than rPC, an increment of 31% for rPC/ANHY and a small increase of 11% for rPC/EPOX respectively. This can be ascribed to recoupling of degraded polymer chains with branching. The long chain branching structure was resulted into higher chain entanglements, which could enhance the rigidity of the samples [64].

Figure 8 presents loss factor ($\tan\delta$) as a function of temperature where the maximum at the peak of the $\tan(\delta)$ corresponds to T_g . DMA was used here as a complementary test for DSC analysis and T_g values of the samples indicated minor differences in Table 4. For rPC, a T_g of 148°C was observed, whereas rPC/EPOX and rPC/ANHY had slightly higher values 149 and 150°C respectively. Higher molecular weight and chain entanglement of the samples based on chain extenders might restrict segmental motion of polymer chains and therefore increased T_g . Although tendency of T_g was similar for both methods, T_g values extracted from \tan curves were slightly higher than

Table 3. Thermal properties of the samples

Sample	T_g , °C	T_{onset} , °C
Virgin PC	145	452
rPC	144	434
rPC/EPOX	145	437
rPC/ANHY	146	450

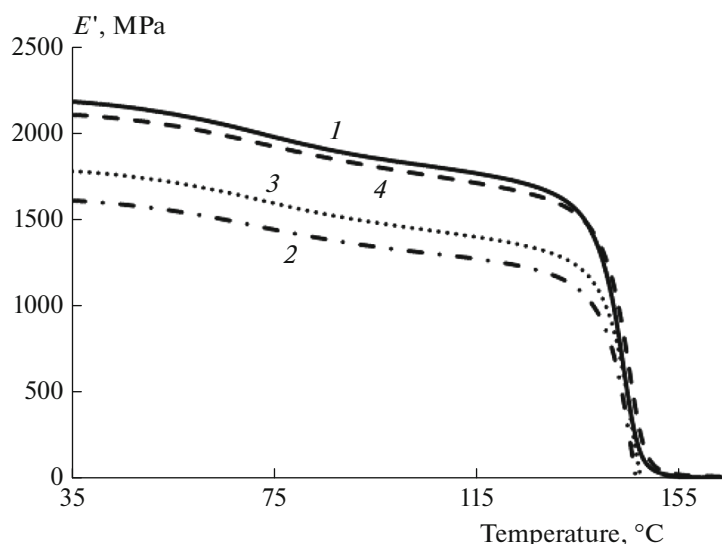


Fig. 7. Storage modulus (E') versus temperature: (1) Virgin PC; (2) rPC; (3) rPC/EPOX; (4) rPC/ANHY.

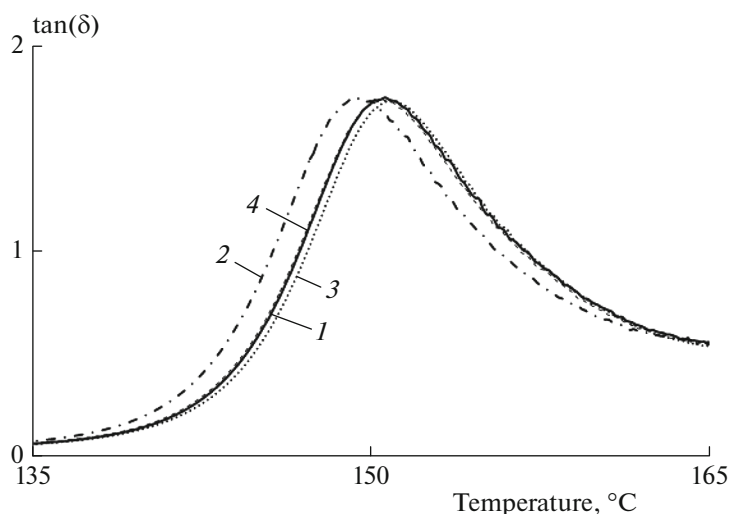


Fig. 8. Loss factor ($\tan \delta$) versus temperature: (1) Virgin PC; (2) rPC; (3) rPC/EPOX; (4) rPC/ANHY.

those obtained by DSC. Similar observations on T_g values were made by other researchers [6, 65] and the results were attributed to differences in test methods.

Table 4. Dynamic mechanical properties of the samples

Sample	T_g , °C	Tan(δ)max	E' at 35°C, MPa
Virgin PC	150	1.74	2179
rPC	148	1.73	1603
rPC/EPOX	149	1.74	1779
rPC/ANHY	150	1.74	2106

CONCLUSIONS

Recycling of PC remains a challenging area since PC undergoes significant thermo-mechanical degradation during re-extrusion. Thermal degradation of PC occurs as scission of carbonate linkages which results in phenolic hydroxyl chain end groups. To compensate this effect, chain extenders were melt-compounded with rPC using a twin-screw extruder. The earlier studies on chain extension of recycled PET revealed that anhydride-based chain extenders indicated more reactivity with hydroxyl end group of PET than epoxy based chain extenders. This conclusion was therefore adapted to the present study, using a

multi-functional anhydride chain extender with hydroxyl chain end groups of rPC for the first time. The hypothesis was also tested by using a multi-functional epoxy chain extender and compared the effects of chain extenders on the properties of the resulting products by rheology, tensile tests, TGA, DSC and DMA measurements. Both types of chain extenders enhanced the viscoelastic and mechanical properties of rPC due to recoupling of degraded chain end groups however, ANHY had much higher chain extension reactivity than EPOX. ANHY increased complex viscosity by 58% and tensile modulus by 39%, whilst EPOX showed 26% increment in complex viscosity and 10% enhancement in tensile modulus on the base rPC. These results evidenced that ANHY is a promising chain extender to adjust the molecular weight of rPC that enables its use in wider applications. The formation of branching occurring upon chain extension was observed as a change in flow behavior of the samples through rheology tests due to reaction of multiple functional groups of chain extenders with hydroxyl chain end groups of rPC. TGA results further supported the findings that ANHY is the most suitable chain extender to rebuild PC degraded structure as ANHY increased the onset temperature for thermal degradation of rPC to higher temperature than that of EPOX. The incorporation of chain extenders slightly increased T_g of rPC in DMA and DSC test results. The higher molecular weight and formation of long chain branching resulted into lower segmental motion of polymer chains and shifted T_g values of the samples.

ACKNOWLEDGMENTS

This work was supported by Kirsehir Ahi Evran University Scientific Research Project Coordination Unit, Project no. MMFA4.20.002. The author acknowledges to BASF, Germany for providing Joncryl ADR 3400 and 4300.

CONFLICT OF INTEREST

The author declares that she has no conflicts of interest.

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