



Direct Conversion of Waste PET to Regenerated Plastics Using Flame Retardants as Depolymerization Catalysts

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Abstract

Polyethylene terephthalate (PET) has been extensively used in the packaging industry, particularly for the production of plastic bottles. PET wastes can be converted into monomeric and oligomeric substances through depolymerization and purification processes. Currently, the chemical recycling of waste plastics is one of the most viable environmental strategies, and the direct repolymerization of depolymerized chemicals without any purification is gaining significant attention because of its simple and cost-effective recycling characteristics. In this study, expandable graphite (EPG) and zeolites were used as glycolysis catalysts for PET depolymerization, and then the reaction product was directly used to polymerize polyurethane foams (PUFs). Both EPG and the zeolites could depolymerize PET, achieving PET conversion as high as 90% with bis(2-hydroxyethyl terephthalate) yields of > 50%. PUFs were also effectively produced from both depolymerized batches, and all PUF samples had a limiting oxygen index of > 31%, indicating high flame retardancy. Thus, EPG and zeolites can play dual roles as glycolysis catalysts and flame retardants. Additionally, this study proposes an efficient PET recycling route.

Keywords Waste PET · Zeolite · Expandable graphite · Glycolysis · Remanufactured polymer

Introduction

Over the past 30 years, plastic consumption has quadrupled worldwide, primarily because of the expansion of emergent markets. From 2000 to 2019, the global production of plastics doubled to 460 million tons. Plastics account for 3.4% of global greenhouse gas emissions. Moreover, 353 million tons of plastic waste were generated from 2000 to 2019. Plastics with lifetimes of less than 5 years contribute nearly two-thirds of plastic wastes, with packaging accounting for 40%, consumer products 12%, and clothing and textiles 11%. Additionally, 9% of plastic waste is recycled, 19% is incinerated, and 50% is deposited in landfills. In certain developing

nations, most wastes evade waste management systems and end up in uncontrolled dumps, open pits, or terrestrial or aquatic environments [1–3].

Polyethylene terephthalate (PET) is one of the most versatile plastics used in a wide variety of applications, particularly as a packaging material, and the generation of PET wastes has increased in tandem with the increased use of PET [3–5]. According to reports, the degradation of PET wastes takes several hundreds of years [6–8]. PET waste recycling technologies that are efficient and cost-effective are desired because conventional methods, such as landfilling and incineration, necessitate large amounts of space and raise environmental concerns. The melting, re-extrusion, or respinning of PET waste was first commercialized as a mechanical recycling technology in the 1970s. However, the thermal and hydrolytic degradation that occurs during re-pelletizing and re-extrusion causes recycled PET to be of poor quality. Over the past decade, alternative chemical recycling technologies, such as hydrolysis, glycolysis, aminolysis, ammonolysis, and methanolysis, have been extensively investigated [9–12]. Glycolysis is used to depolymerize PET into various monomeric and oligomeric bis(2-hydroxyethyl terephthalate) (BHET) polyols in the presence of ethylene glycol and

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a catalyst. Afterward, separation and purification procedures, such as distillation and extraction, are used to obtain monomeric BHET with a high degree of purity. However, these sequential separation and purification procedures are expensive and energy-intensive, which occasionally degrades the commercial quality of glycolized products [11, 13–16]. The depolymerized products contain numerous polyols, including BHET monomers, BHET oligomers, and glycols, and can therefore be used as polyols in the polymerization of polyurethane. During the polymerization process, these recycled polyols serve as flexible segments of polyurethane by irreversibly reacting with isocyanate [17–22]. The direct reuse of depolymerized PET would eliminate the need for an energy-intensive purification process, thereby suggesting an energy-efficient PET recycling process.

Expandable graphite (EPG) is a form of intercalated graphite that appears as 100–500 µm-sized particulates or flakes. When EPG is heated to temperatures of 140–300 °C, it expands and elongates to form a worm-like structure. This produces particles that are significantly longer and larger than the original particles. EPG can serve as an intumescent flame retardant when merged with various flammable polymers [23–27]. Additionally, the intercalated layer of EPG contains defective sites and metal ions that could serve as reactive sites for PET glycolysis reactions. Conversely, zeolites are microporous crystalline aluminosilicates that are primarily used as catalysts and absorbents. Zeolites with high Al substitutions have a large ion-exchange capacity and have been extensively used as catalysts, adsorbents, and detergent builders because of their high charge density [28–30]. As flame retardants, zeolites reduce total smoke production, carbon dioxide evolution, and polymer matrix degradation; they also catalyze the transformation of amorphous carbon residues into thermally stable graphitic structures [31–34]. Similar to EPG, zeolite frameworks contain reaction metal ions that can function as Lewis or Brønsted acid sites for glycolysis reactions.

Considering that EPG and zeolites can function as both potential catalysts for PET glycolysis and flame retardants, we investigated the depolymerization and direct repolymerization of PET into PUF in the presence of EPG and zeolites. First, the efficacy of PET depolymerization was investigated using an EPG and zeolite-based catalyst system. Second, the reaction products were used as polyols in the polyurethane foam (PUF) polymerization process. Third, the structural properties of the PUF were analyzed using scanning electron microscopy (SEM), Fourier-transform infrared spectroscopy (FT-IR), X-ray diffraction (XRD), and gel permeation chromatography (GPC). Finally, the thermal stability and flame retardancy of the synthesized PUF samples were evaluated using derivative thermogravimetric (DTG) analysis, micro-combustion calorimetry (MCC), and the limiting oxygen index (LOI).

Herein, the efficacy of the proposed PET recycling strategies is discussed.

Experimental

Materials

Aluminum hydroxide (Al(OH)₃), cerium dioxide (CeO₂; ≥ 99.0%), n-pentane (≥ 99%), kaolin, silicon dioxide (SiO₂; ~ 99%), calcium carbonate (CaCO₃; ≥ 99.0%), 1,4-diazabicyclo[2.2.2]octane (DABCO 33-LV), zeolite 13X, monoethylene glycol (≥ 99%), tetrahydrofuran (THF; ≥ 99.9%), and PET were purchased from Sigma-Aldrich. The zeolite was supplied by Alfa Aesar. Tricresyl phosphate (TCP) was obtained from Duksan Chemical Co., Ltd. (Korea). The silicon surfactant (DC 193) was supplied by Dow Inc. (USA). Potassium octanoate (K-15) was supplied by Air Products (USA), and 4,4-diphenylmethane diisocyanate (MDI) was supplied by BASF SE (Germany). EPG 80 mesh (expansion rate: 280–300 mL/g; fixed carbon: 94.63%) was supplied by SamJung C&G (Korea). Polyether polyol (PPG CS-743) was purchased from Kumho Chemical (Korea). All the chemicals were used without any additional purification.

PET Depolymerization

Two types of polyols were produced for the depolymerization reactions. EPG 80 was used as a glycolysis catalyst for PET depolymerization. Next, zeolites 4A and 13X were used together as a dual catalytic system in a separate experiment. Table 1 presents the detailed recipes. PET glycolysis was performed at 195 °C. The PET conversion and BHET yield were recorded after 2 h of reaction with respect to the reaction time. The PET conversion was calculated using the following equations.

$$\text{PET conversion(\%)} = \frac{W_{\text{PET,initial}} - W_{\text{PET,final}}}{W_{\text{PET,initial}}} \times 100, \quad (1)$$

Table 1 Recipes used for PET depolymerization

EPG-based batch	EPG-based batch (g)	Zeolite-based batch (g)
PET	3	3
Monoethylene glycol	15	15
EPG 80	0.6	
Zeolite 4A		0.75
Zeolite 13X		0.75

$$\text{BHET ;yield(\%)} = \frac{W_{\text{BHET}}}{W_{\text{PET,initial}}} \times 100. \quad (2)$$

Repolymerization of the Depolymerized Plastics

The repolymerization reaction was performed using a recipe reported in the literature [21]. First, the DABCO K-15 and 33-LV catalysts were used, and the ratio of potassium octanoate to diethylene glycol in the K-15 catalyst was set as 1:3.44. The hydroxyl and acid values of the reaction products were measured and calculated as follows:

$$\text{Acidvalue} \left(\frac{\text{mg}}{\text{g}} \right) = \frac{56.1(V_0 - V_1)T}{m}. \quad (3)$$

The OH values were measured using the NF T 52–113 standard method [35, 36]:

$$\text{Hydroxyl value} \left(\frac{\text{mg}}{\text{g}} \right) = \frac{56.1(V_0 - V_1)T}{m} + \text{Acid value}, \quad (4)$$

where V_1 is the volume (mL) of the NaOH solution used for the test sample [37], V_0 is the volume (mL) of the NaOH solution used for the blank sample, m (g) is the mass of the sample, and T is the normality of the NaOH solution (mol/L).

The concentration was measured using the NCO weight percentage.

$$\text{NCO value} = \frac{\text{Weight of NCO}}{\text{Weight of isocyanate}} \times \text{No. of NCO} \times 100, \quad (5)$$

where “No. of NCO” represents the number of functional groups per mole.

The equivalent weight of the isocyanates was calculated as follows:

$$\text{Equivalent weigh to fisocyanate} = \frac{4,200}{\text{NCOvalue}} F_{\text{equivalent}}. \quad (6)$$

Similarly, the equivalent weight of the polyols was calculated as follows:

$$\text{Equivalent weight of polyol} = \frac{56,100}{\text{OH value}} g_{\text{equivalent}}. \quad (7)$$

where the hydroxyl value was obtained using titration methods with a standard 0.5 *N* NaOH solution [35, 36].

Finally, the isocyanate index was obtained using the following equation.

$$\text{isocyanate index} = \frac{\frac{\text{Mass of MDI}}{\text{Equivalent weight of the isocyanate}}}{\frac{\text{Mass of polyol}}{\text{Equivalent weight of the polyol}}}, \quad (8)$$

Considering the stoichiometry of the polymerization reaction, isocyanate index 300 was adopted for the polymerization of the modified cyclic molecular structure [38–40].

Characterization

After applying platinum coatings, the repolymerized PUFs were examined with the glycolized polyols using SEM (XL30S FEG, Philips) and a Schottky field-emission electron gun at an acceleration voltage of 10 kV. The diffraction patterns were investigated by performing XRD analysis (Ultima IV, Rigaku) with a horizontal goniometer, which performed θ – 2θ scans (X-ray source: Cu $K\alpha$ radiation, $\lambda = 1.5418 \text{ \AA}$; $2\theta = 1.5^\circ$ – 160° ; scan speed: $3^\circ/\text{min}$; step size: 0.02° ; power: 40 kV and 40 mA). An FT-IR spectrometer (ALPHA-II, Bruker, Billerica, US) with an attenuated total reflectance (ATR) module was used to verify the completion of the polymerization reactions. The IR spectra were recorded by conducting an FT-IR run with the OPUS software. The conditions for the FT-IR measurements were a scan range of 350 – 6000 cm^{-1} , 24 scans, and a resolution of 4 cm^{-1} . GPC (Waters e2695 Alliance) was performed with a differential refractometer connected to Styragel HR-0.5, HR-1, HR-4E, and HR-4 ($7.8 \times 300 \text{ mm}$) columns using THF as an eluent. The thermal degradation behaviors of four samples using depolymerized products (were investigated using thermogravimetric analysis (EVO II TG8120 series, Rigaku) in a nitrogen atmosphere to analyze the thermal stability of the remanufactured polyurethane. MCC was performed using the Federal Aviation Administration (FAA) microcalorimeter based on the pyrolysis combustion flow calorimetry of ASTM D7309 (Fire Testing Technology Ltd., United Kingdom). MCC has been closely correlated with fire test data (cone calorimeter, OSU), flammability results (LOI, UL94), and combustion tests (bomb calorimeter), and it is regarded as an efficient tool for assessing flammability.

Results and Discussion

Table 1 lists the recipes for performing PET glycolysis at 195°C using EPG 80 and zeolite as single catalysts. The glycolysis was conducted for 2 h, and the corresponding PET conversion and BHET yield were measured (Fig. 1). As shown in Fig. 1(a), the PET conversion for the EPG-based reaction batch reached 94.46% after 1 h and 96.80% after 2 h. This indicates that EPG can serve as a glycolysis catalyst, with its catalytic activity originating from its structural defects. Metallic ions present in the intercalated

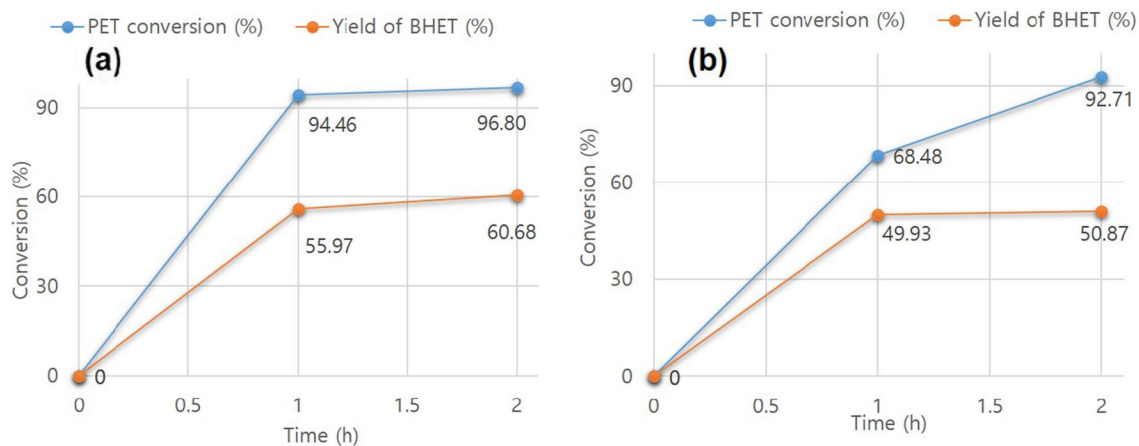


Fig. 1 PET conversion and BHET yield of the **a** EPG systems and **b** zeolite systems

layer of EPG may facilitate the glycolysis reaction. Conversely, a batch of glycolysis system containing zeolites 4A and 13X had a slightly low PET conversion of 68.48% after 1 h, and then the PET conversion increased to 92.71% after extending the reaction time to 2 h (Fig. 1(b)). Although both zeolites 4A and 13X contain Al species that serve as Lewis acid sites in their frameworks, their PET conversion was less effective than that of EPG. This may indicate that the PET chains could not access the microporous regions of the zeolite because of their large size, while the external surface of the zeolite only functioned as an active site for the glycolysis reaction. Similar trends were observed for the BHET yield, where the EPG-based catalyst system outperformed the zeolite-based catalyst system by producing higher yields of 55.97% and 60.68% after 1 h and 2 h of reaction, respectively. None of the two catalyst systems could achieve a BHET yield of > 90%, unlike metal and ionic-based catalyst systems. Such outcomes are attributable to the nature of bulky solid catalysts, which offer a low frequency of catalytic reactions. Thus, neither EPG nor zeolite was an effective catalyst for producing monomeric BHET. However, the products of both reactions contained numerous polyols, such as BHET dimers, trimers, tetramers, pentamers, and hexamers. Such polyols can be used as the main components for PUF polymerization.

PUF repolymerization was performed using the depolymerized products. Table 2 describes the ingredients and nomenclature of the samples. Figure 2 displays the SEM images of the PUF specimens. All the samples exhibited similar anisotropic cell morphologies with cavity structures, as shown in Fig. 2. The cell size and topology were not altered by the PUF recipe, but they were presumably affected by the blowing agent type, blowing conditions, and gelation rate, which were the same for all the samples. The cell size of all the samples was $\sim 300 \mu\text{m}$, and the SEM analysis did not detect any discernible changes in physical dimensions.

Table 2 Recipes for repolymerization using the depolymerized products

	PUF_E1	PUF_E2	PUF_Z1	PUF_Z2
MDI	87.50 g	87.50 g	87.50 g	87.50 g
CS-743	13.94 g	13.00 g	13.22 g	13.58 g
Glycolyzed polyol (with EPG at 195 °C)	2.11 g	2.11 g	–	–
Glycolyzed polyol (with zeolite at 195 °C)	–	–	2.68 g	2.68 g
TCP	3.75 g	3.75 g	3.75 g	3.75 g
Expandable graphite (EPG 80 mesh)	4.81 g	4.81 g	4.99 g	4.99 g
CaCO ₃	7.50 g	7.50 g	7.50 g	7.50 g
Al(OH) ₃	3.75 g	3.75 g	3.75 g	3.75 g
CeO ₂	–	2.49 g	–	–
Kaolin	–	2.49 g	–	–
Zeolite 4A	3.75 g	–	–	–
Zeolite 13X	3.75 g	–	–	–
SiO ₂	–	6.42 g	–	6.65 g

As shown in Fig. 3(a), all the samples exhibited PUF peaks corresponding to the amide group $-\text{NH}$ ($\sim 3300 \text{ cm}^{-1}$), carbonyl urethane group $-\text{C}=\text{O}$ ($\sim 1700 \text{ cm}^{-1}$), carbamate group $-\text{CN}$ ($\sim 1600 \text{ cm}^{-1}$), and the distinct C–N stretching vibration ($\sim 1405 \text{ cm}^{-1}$) of the isocyanurate ring structure in the IR-ATR mode. Thus, the PUF samples were effectively repolymerized using the depolymerized products. However, the peak at $2020\text{--}2400 \text{ cm}^{-1}$ indicates the presence of free isocyanate groups in samples containing unreacted isocyanate [38–40]. It was challenging to precisely identify the amount of polyol that participated in the polymerization because there were various polyol structures in the depolymerization batch; thus, excess isocyanate was used. Figure 3(b) displays the XRD results for the PUF samples. EPG 80, zeolite 4A, and zeolite 13X were used as both glycolysis

Fig. 2 SEM images of the PUF samples. **a** PUF_E1, **b** PUF_E2, **c** PUF_Z1, and **d** PUF_Z2

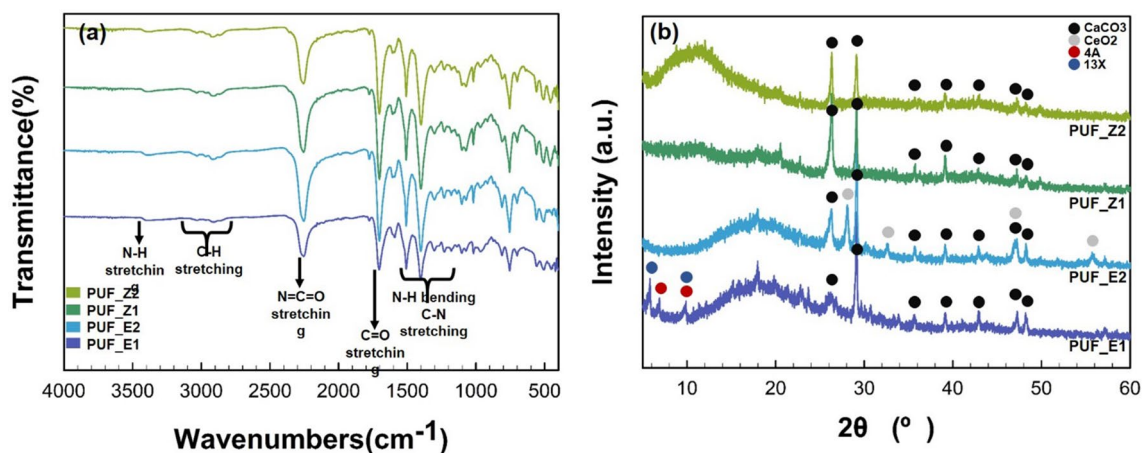
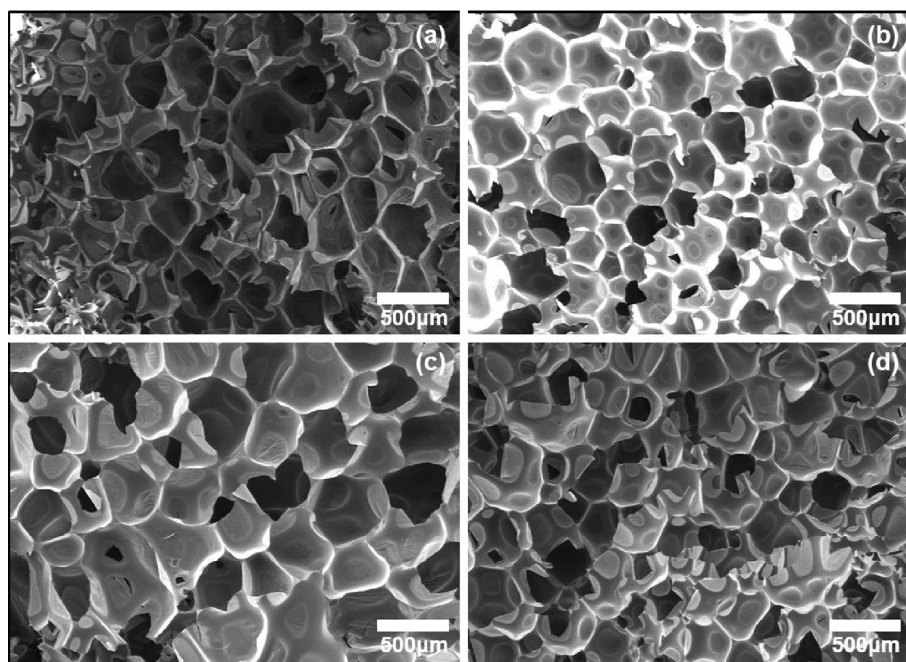


Fig. 3 **a** FT-IR and **b** XRD data for the PUF samples

catalysts and flame retardants. Other flame retardant additives, such as CaCO_3 and $\text{Al}(\text{OH})_3$, were also included, as shown in Table 2. Figure 3(b) shows the XRD data for these inorganic materials. PUF_E1 clearly exhibits visible zeolite, EPG 80, and CaCO_3 peaks, and the other three samples also reveal the presence of EPG 80 in their structures. The distinct diffraction peak of EPG 80 indicates that the EPG crystal structure was well preserved after polymerization. Contrarily, the zeolite diffraction peak in the depolymerized batches for PUF_Z1 and PUF_Z2 was weakly detected at 25° because of low loading (< 1 wt%) in the PUF. The XRD diffraction patterns were comparable, but there was a shift in the broad spectra at 10° for PUF_Z2 and 18° – 20° for PUF_E1 and PUF_E2 due to the packing of polymer

chains, indicating that the nature of the polyols used for the repolymerization may have influenced the packing of the polymer chains. The connection between the packing of the polymer chains and the properties of the polyols has not yet been fully understood; therefore, additional research is required.

The GPC data for the lower weight average molecular weight (Mw) range of small polyurethane particles overlap with those of glycolized products; thus, PUFs would take more than 24 h to slightly dissolve in THF. Therefore, the recycled polyol samples were analyzed via GPC to perform a targeted analysis on the glycolized products, which were converted from PET plastic waste. Figure 4 and Table 3 present the corresponding GPC data. The polydispersity

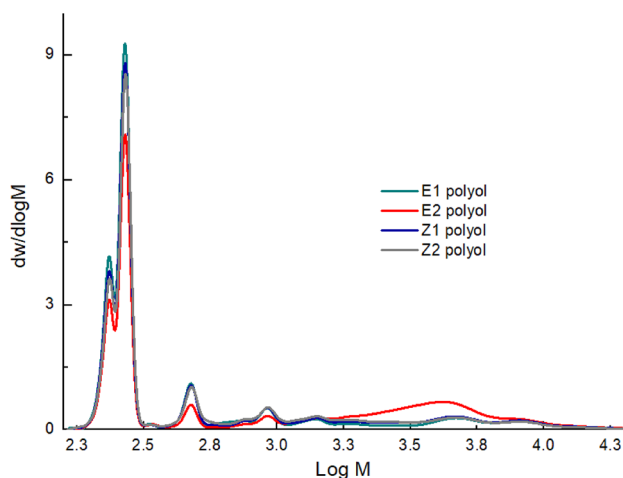


Fig. 4 GPC chromatogram of the polyol samples

indices of all the peaks were greater than 1, and the molecular weight did not vary significantly. PET depolymerization yields BHET monomers and oligomers when the molecular formula and weight of BHET are $C_{12}H_{14}O_6$ and 254.24 g/mol, respectively. The number average molecular weight (M_n) ranged between 342 and 430 g/mol, whereas the M_w of the polyol samples ranged between 1260 and 2045 g/mol. Thus, BHET dimers, trimers, and tetramers would be the predominant oligomers in this depolymerization reaction, as M_n regards polymeric particles equally and M_w weighs the long chains. Since the BHET yield of the E1 and E2 polyols was 60.68% and that of the Z1 and Z2 polyols was 50.87%, the increased polydispersity index was expected. This indicates that in the case of EPG depolymerized polyols, 39.32% of BHET oligomers comprise dimers, trimers, and tetramers in addition to 60.68% of the monomer Table 4.

Figure 5 depicts the DTG results and heat release rate (HRR). Mass losses observed during DTG analysis were sensitive during microcalorimetry because of the presence of additives, such as organic and inorganic flame retardants, emulsifiers, cross-linkers, unreacted PET, the BHET monomer and oligomers, and the side-reaction products of BHET. Interestingly, the PUF samples prepared from the zeolite-based depolymerized batch had lower HRC, HRR, and total heat release (THR) values than those prepared from the EPG-based batches. This indicates that PUF_Z1

and PUF_Z2 exhibited higher thermal stability than PUF_E1 and PUF_E2 even though all the samples had identical flame retardant loading amounts. The high thermal stability is due to the PUF_Z1 and PUF_Z2 polyols containing less monomeric BHET than the PUF_E1 and PUF_E2 polyols because the depolymerization reaction was less active in the zeolite-based batch.

The LOI test performed on the samples revealed that PUF_Z1 and PUF_Z2 had greater flame-retarding properties than PUF_E1 and PUF_E2. Additionally, high LOI values of 32.48% and the lowest THR of 12.86 kJ/g were achieved for the remanufactured PUF using zeolites as a glycolysis catalyst. The PUF_E1 and PUF_E2 samples afforded a greater BHET yield than the zeolite-based counter samples. Thus, the synthesized PUF samples contained relatively few BHET oligomers and low aromatic group contents. The carbon atoms in benzene have sp^2 hybrid orbitals and form s -bonds with two adjacent carbon atoms and one hydrogen atom. Each carbon atom possesses a p -orbital that can form p -bonds. BHET oligomers would demonstrate enhanced flame-retarding properties because they contain connected aromatic structures with a high rate of energy dissipation. Thus, lowering either the reaction temperature or time in EPG catalyst systems would be advantageous for enhancing flame retardancy because these conditions can produce depolymerized polyols with high oligomeric BHET contents. Moreover, the depolymerized PUF exhibited comparable thermal stability and flame retardancy to PUFs fabricated using conventional polyols. Thus, the proposed upcycling method employing EPG and zeolites as dual-functional materials would be an effective strategy for managing PET waste.

Table 4 Thermal stability and flame retardancy of the PUF samples

	PUF_E1	PUF_E2	PUF_Z1	PUF_Z2
HRC (J/g·K)	122.6	120.4	108.6	110.6
Peak HRR (W/g)	114.0	111.9	105.7	111.2
THR (kJ/g)	14.68	14.48	12.86	13.04
LOI (conc.%)	31.56	31.43	32.48	32.03

Table 3 Average molecular weight and molecular weight distribution

	Number average molecular weight (M_n , g/mol)	Weight average molecular weight (M_w , g/mol)	Polydispersity index
E1 polyol	342	1260	3.681932
E2 polyol	439	2045	4.659212
Z1 polyol	359	1391	3.877853
Z2 polyol	365	1317	3.611856

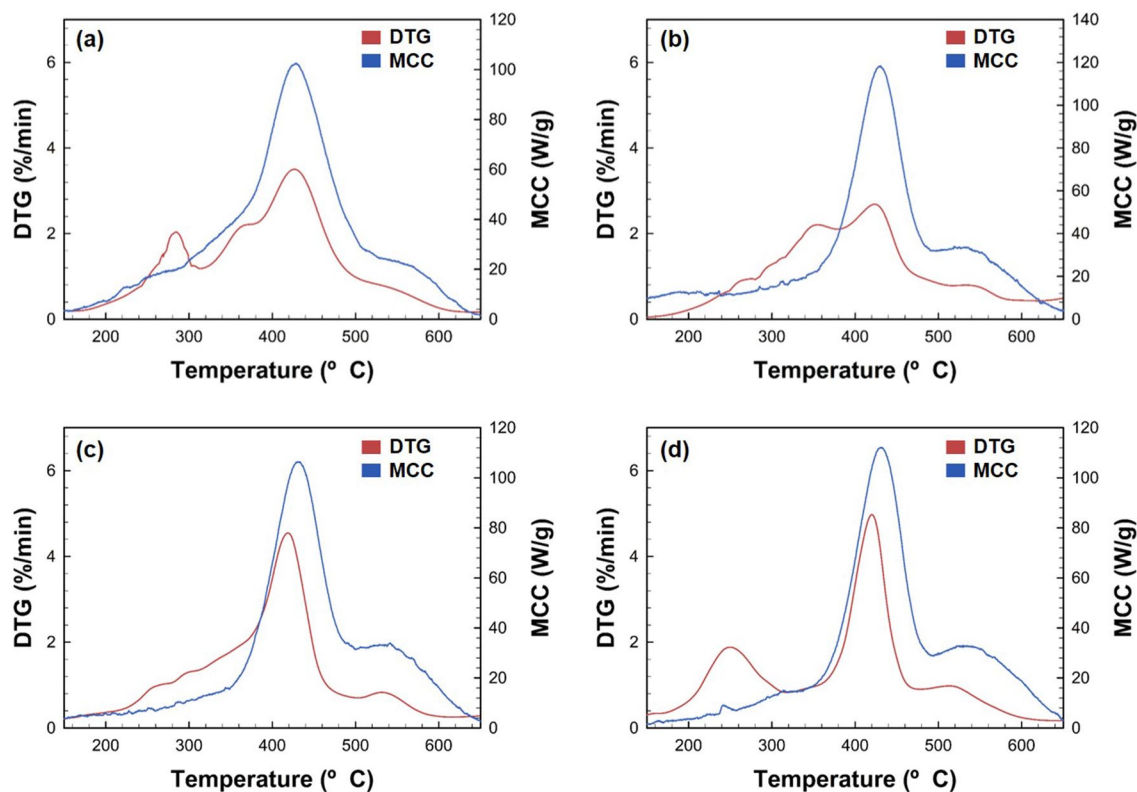


Fig. 5 DTG analysis and HRR of PUF_E1, PUF_E2, PUF_Z1, and PUF_Z2

Conclusion

In this study, EPG and zeolites were used as depolymerization catalysts for waste plastic recycling and remanufacturing. These materials also served as flame retardants during the subsequent process of remanufacturing the flame-retarding insulation foam. It is evident that EPG and zeolites both play a role as catalysts in the depolymerization of PET and that the depolymerized polyols can be used to successfully polymerize remanufactured PUFs. Although both EPG and the zeolites afforded a low BHET yield, they achieved a PET conversion of > 90%. Therefore, EPG and zeolites would be appropriate for use as catalysts in the polymerization of high-value plastics because they produce high contents of BHET oligomer derivatives. However, they would not be suitable for the production of pure monomeric chemicals.

Author Contributions PS analyzed and interpreted the data. SMG analyzed and interpreted the data, especially the kinetics. PS made major contributions to the writing of the manuscript, and SMG supervised the project. All authors have read and approved the final manuscript.

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Availability of Data and Materials The datasets used and/or analyzed are available from the corresponding author upon reasonable request.

Declarations

Conflict of Interest The authors declare that they have no competing interests.

Ethical Approval Not applicable.

Consent to Participate Not applicable.

Consent to Publish Not applicable.

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