Synthesis of Novel Thermostable Polyamideimides from Bis(2-aminoethyl)terephthalamide and Dianhydrides

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ABSTRACT: Bis(2-aminoethyl)terephthalamide, an amide-containing diamine, was obtained from the aminolysis of waste poly(ethylene terephthalate) bottles. This diamine reacts with various aromatic dianhydrides to form novel polyamideimides (PAIs). The formation of amic acid or ammonium carboxylate salt intermediates depends strongly on the substituents of the dianhydrides. The electron-withdrawing substituents promote the creation of an ammonium carboxylate salt, whereas the electron donors assist with the amic acid intermediate formation. These salts and amic acids were further converted into polyimides by thermal treatment. The structures of the intermediates and PAIs were characterized by Fourier transform infrared, 1H nuclear magnetic resonance (NMR), and 13C NMR spectroscopies, and their thermal properties were determined by differential scanning calorimetry and thermogravimetry. X-ray diffraction patterns and inherent viscosity values of these PAIs were also reported. By using these chemical transformations, waste poly(ethylene terephthalate) bottles were converted into high-performance PAIs. These PAIs can be used as membrane-modifying agents for industrial separation applications.

1. INTRODUCTION

Polyimide (PI) is one of the high-performance polymers. It exhibits excellent thermal, electrical, and mechanical properties, and consequently, it has been widely used in aerospace, electronics, and other industries.1–9 PI can be copolymerized with a polyamide. The commercial polyamideimide (PAI) with the brand name of TorlonVR 4000TF has the superior properties of both polyamides and PIs, such as high mechanical strength, thermal resistance, and wide chemical resistance; hence, there are many developments for its application, including gas separation, nanofiltration, osmotic power generation, pervaporation, and so forth.10 PAI (Torlon) can also be used as the modification agent for cellulose acetate membranes11 or polytetrafluoroethylene (PTFE) fibrous porous membranes.12 The prepared PAI-reinforced membranes have potential in industrial separation applications. In addition, PAI was used in composites, such as Nylon-12/PAI composite coatings.

Some key methods are used to prepare PAI: (i) condensation of an imide-containing diacid with a diamine,13–21 (ii) condensation of an amide-containing diamine with a dianhydride,22–24 and (iii) thermal imidization of a polyamide–amic acid (AA).25

Bis(2-aminoethyl)terephthalamide (BAET) is a diamine containing a terephthalamide moiety. This diamine was prepared by the aminolysis of waste poly(ethylene terephthalate) (PET) bottles with ethylenediamine.26 This is a short and cost-effective approach to prepare an amide-containing diamine compared to the two-step22 or four-step23 methods from monomers.

In this study, by the transformation of BAET into a novel PI, we can use waste PET bottles as a source of materials and hence contribute to the chemical methods of recycling waste materials. Therefore, the outcome of this study is the finding of a new environment-friendly approach to recycle PET into novel monomers/oligomers that are potential raw materials for preparing other new polymers (PIs) to attain useful products with special performance and properties.

2. RESULTS AND DISCUSSION

The Fourier transform infrared (FTIR) spectrum of BAET (Figure 1a) is identical to the published data.26 This diamine was used for PI preparation.

When an input molar ratio of BAET-pyromellitic dianhydride (PMDA) of 1:1 or 2:1 was used in dried dimethyl acetamide (DMAc), N-methylpyrrolidone (NMP), dimethyl sulfoxide (DMSO), or glacial acetic acid, the same intermediate was isolated and characterized by FTIR and
NMR spectroscopy techniques. The common absorption bands of AA could not be found in the FTIR spectrum of the intermediate (Figure 1b), but only the existence of an ammonium carboxylate (AC) salt was observed. For example, the comparison of the FTIR spectra of BAET (Figure 1a) with AC-BAET-PMDA (Figure 1b) showed that the weak band at 1577 cm\(^{-1}\) in BAET became stronger. This band is characteristic for the asymmetric CO\(_2\)-amide stretching band of BAET at 1632 cm\(^{-1}\). In addition, the C=O amide stretching band of BAET at 1632 cm\(^{-1}\) shifted to a lower wavenumber at 1623 cm\(^{-1}\) because of the existence of the NH\(_4\)\(^+\) asymmetric deformation band that used to appear in the range of 1625–1560 cm\(^{-1}\).

The structure of the AC salt (Scheme 1) obtained from BAET and PMDA was also confirmed by \(^{1}\)H NMR and \(^{13}\)C NMR spectroscopy techniques. The common absorption bands of AA could not be found in the FTIR spectrum of the intermediate (Figure 1b), but only the existence of an ammonium carboxylate (AC) salt was observed. For example, the comparison of the FTIR spectra of BAET (Figure 1a) with AC-BAET-PMDA (Figure 1b) showed that the weak band at 1577 cm\(^{-1}\) in BAET became stronger. This band is characteristic for the asymmetric CO\(_2\)-amide stretching band of BAET at 1632 cm\(^{-1}\). In addition, the C=O amide stretching band of BAET at 1632 cm\(^{-1}\) shifted to a lower wavenumber at 1623 cm\(^{-1}\) because of the existence of the NH\(_4\)\(^+\) asymmetric deformation band that used to appear in the range of 1625–1560 cm\(^{-1}\).

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Figure 2. DSC curves of AC-BAET-PMDA: (a) heating from RT to 320 °C at a rate of 10 °C/min and holding for 5 min at 320 °C; (b) cooling to RT at 50 °C/min; and (c) heating from RT to 320 °C at 10 °C/min.

Figure 3. TG and dTG curves of AC-BAET-PMDA heating from RT to 800 °C at a rate of 10 °C/min.

Figure 4. TG and dTG curves of PI-BAET-PMDA heating from RT to 800 °C at a rate of 10 °C/min.
step was 30 min. The FTIR spectrum of the obtained PI-BAET-PMDA (Figure 1c) exhibits absorptions at 1775, 1712 cm$^{-1}$ (C=O of N-aliphatic imide), 1396 cm$^{-1}$ (aliphatic C−N), 1156, and 727 cm$^{-1}$ (imide ring deformation). The terephthalamide moiety is observed at 3405 cm$^{-1}$ (stretching of amide N−H), 1640 cm$^{-1}$ (stretching of amide C==O), and 1540 cm$^{-1}$ (deformation of amide N−H).

The TG and dTG curves (Figure 4) show that the obtained PI is stable from RT to 400 °C with a maximum degradation temperature $T_d^\text{PI}$ at 514 °C. At 800 °C, the residue is 10.1%. This $T_d^\text{PI}$ is not different from the one of the second decomposition step of AC-BAET-PMDA (Figure 3). This $T_d^\text{PI}$ value is much higher than that at 410 °C of the N-aliphatic amide prepared by Mallakpour et al.29 from N,N'- (pyromellitoyl)-bis-l-leucine diacid chloride and aromatic diamine.

The presence of short aliphatic ethylene in diamine does not significantly reduce the thermal stability of PI-BAET-PMDA, but the existence of an imide moiety in the chain increased its thermal stability compared to the corresponding polyamide. The decomposition peak of poly(ethylene terephthalamide) was reported at 441 or 430 °C.30

The PAI preparation procedure and characterization, as described above, were also applied similarly to prepare PIs from BAET and 3,3',4,4'-benzophenonetetraacrylic dianhydride (BTDA) or 4,4'-oxydiphthalic anhydride (OPDA). The $^1$H NMR spectrum (Figure S3) of the intermediate isolated from the reaction of BAET with BTDA in DMSO showed complex structures that were identified as a mixture of AA and AC. The methylene protons appear as two pairs of peaks at (i) 3.51 and 3.01 ppm of AC and (ii) 3.48 and 3.40 ppm of AA. The signals of the methylene protons of AA are very close (the difference is only 0.08 ppm), as discussed above. In addition, the integrals of AC and AA protons are almost the same; therefore, this is an AC/AA mixture of 1:1.

Contrary to PMDA and BTDA, the reaction of BAET with OPDA produces a mixture of AA and AC intermediates and PI, as indicated by the bands at 1713 and 1776 cm$^{-1}$ in the FTIR spectrum. In the 3–4 ppm range of the $^1$H NMR spectrum (Figure S6) of the intermediate, two pairs of peaks were also observed; however, the signals of AA or PI methylenes were much stronger than that of AC. Quantitative determination by taking the integrals of these peaks is not accurate because they are overlapped by the H$_2$O signal, even after D$_2$O treatment (Figure S7). Hence, the intermediate isolated from the BAET–OPDA reaction contains principally AA, PI, and a minor quantity of AC. This AA intermediate easily eliminates water to transform into PI.

The formation of different intermediates from BAET and dianhydrides is explained by the reaction mechanism and electrophilic effects of substituents. In the first step of the reaction, an electron pair of the amine, acting as a Lewis base, attacks a carbonyl carbon in the anhydride group, acting as a Lewis acid center. The electron-withdrawing substituents in the dianhydride will enhance the acidity of this carbonyl carbon center and vice versa. As a consequence, the acidity of the carbonyl carbon in anhydrides decreases according to the order PMDA (two C=O as electron-withdrawing substituents) > BTDA (one C=O substituent) > OPDA (O-ether as an electron-donating substituent). In addition, PMDA is more soluble than BAET in any solvent; therefore, PMDA reacts at first with trace water or moisture to form carboxylic acid, and then, this acid reacts with BAET to form an AC salt. Because of its low acidity, OPDA is less sensitive to water and can react with BAET to form an AA intermediate.

The intermediates isolated from the reactions of BAET with BTDA or OPDA were also further transformed into PAIs by thermal treatment. The existence of the imide and amide structures of the final PAI products was confirmed by the corresponding pairs of peaks at 1775/1712 and 1640/1540 cm$^{-1}$ in the FTIR spectra (Figure 5).

From the TG curves (Figure 6) and Table 1, the thermal stability of PAIs prepared from BAET and dianhydrides was

![Figure 5](https://dx.doi.org/10.1021/acsomega.0c00799)

**Figure 5.** FTIR spectra of (a) PI-BAET-PMDA, (b) PI-BAET-BTDA, and (c) PI-BAET-OPDA.

![Figure 6](https://dx.doi.org/10.1021/acsomega.0c00799)

**Figure 6.** TG curves of (a) PI-BAET-PMDA, (b) PI-BAET-BTDA, and (c) PI-BAET-OPDA.

| PI               | Intermediates | Temperature at 10% weight loss (°C) | Maximum decomposition temperature of PI (°C) |
|------------------|---------------|-------------------------------------|-------------------------------------------|
| PI-BAET-PMDA     | AC            | 466                                 | 514                                       |
| PI-BAET-BTDA     | AC + AA       | 425                                 | 450                                       |
| PI-BAET-OPDA     | AA + PI       | 409                                 | 445                                       |

*Decomposition temperature at which a 10% weight loss was recorded by TG at a heating rate of 10 °C·min$^{-1}$ in nitrogen.*
rings yield more char residues at 800 °C compared to PI-BAET-PMDA.

The morphology of the obtained PAIs was also studied by wide-angle X-ray diffraction (XRD) on powder specimens (Figure 7).

Figure 7. Wide-angle XRD patterns of (a) PI-BAET-PMDA, (b) PI-BAET-BTDA, and (c) PI-BAET-OPDA.

The broad peaks centered at 2θ = 13.5° on each XRD pattern indicated that they were almost amorphous and attributed to the average intersegmental distance of polymer chains. The average d-spacing was calculated based on Bragg’s law: 

\[ n\lambda = 2d \sin \theta \]

where \( d \) is the dimension spacing, \( \theta \) is the diffraction angle, \( \lambda \) is the X-ray wavelength (1.54184 Å), and \( n \) is an integral number (1, 2, 3, ...). Therefore, the d-spacing values of our PAI samples were calculated as 6.6 Å.

Another series of PAIs was prepared from an aromatic amide-containing diamine, \( N,N' \)-bis(4'-amino-4-biphenylene)-isophthalamide, and PMDA, BTDA, or OPDA by the bulk thermal imidization of AAs. The XRD patterns of these PAIs showed broad peaks at 2θ = 20° or d = 4.4 Å. These aromatic PAIs have a slightly tighter chain packing compared to our N-aliphatic imide PI-BAET-PMDA shows a high thermal stability compared to the presence of a benzene ring (PMDA); however, these carbonyl (BTDA) and ether (OPDA) groups in the dihydrides enhance their stability to moisture compared to the strong inter- and intramolecular interactions of the flat symmetrical pyromellitimide rings.

The crystallinity of PI-BAET-PMDA was derived from the strong inter- and intramolecular interactions of the flat symmetrical pyromellitimide rings. In contrast, the crystallinity of PI-PMDA-OPDA was acquired from the PI ring formation in solution. The lower crystallinity of PI-PMDA-BTDA is ascribed to the PI formation by the thermal treatment of AA and AC intermediates and the unsymmetrical benzophenone ring.

In addition, the heating curves of PAI-BAET-BTDA (Figure S8) and PAI-BAET-OPDA (Figure S9) showed endothermic peaks at 184 and 178 °C, respectively. Combined with the TG curves (Figure 6b,c), this thermal occurrence could be because of a physical transition without mass loss. These endothermic peaks could be assigned to the transition of terephthalamide crystalline segments.

The novel PAIs are only soluble in phosphoric acid. The interaction between PI and phosphoric acid was investigated by FTIR analysis. This PI was prepared from OPDA and 4,4′-diaminodiphenyl ether. The FTIR results show that phosphoric acid interacts with PIs mainly by hydrogen bonds rather than by the protonation of PI at RT.

The inherent viscosity (IV) in 85% H₃PO₄ solution (0.5 g/ dL at 30 °C) of PAIs was determined by using an Ostwald viscometer of Cannon-Fenske, size no. 450. The IV values of PI-BAET-PMDA, PI-BAET-BTDA, and PI-BAET-OPDA are 0.466, 0.204, and 1.15, respectively. PI-BAET-OPDA showed the highest IV, whereas PI-BAET-PMDA and PI-BAET-BTDA have lower IV values or lower molecular weights (MWs). This higher MW of PI-BAET-OPDA could be the result of the direct formation of PAI and AA in solution instead of the formation of AC intermediates.

3. CONCLUSIONS

In conclusion, novel PAIs were prepared from the reactions of BAET with PMDA, BTDA, and OPDA. When PMDA was used, even in dried solvents, the intermediate was isolated and identified as an AC salt. BTDA reacted with BAET to form a mixture of AC and AA. In the case of OPDA, the AA intermediate was mainly formed. These intermediates were converted into PAIs by thermal treatment. This nonsolvent dehydration process shows environmental advantages; however, the obtained PIs possess noncrystalline structures. The N-aliphatic imide PI-BAET-PMDA shows a high thermal stability compared to the fully aromatic PI. The presence of carbonyl (BTDA) and ether (OPDA) groups in the dihydrides enhances their stability to moisture compared to the presence of a benzene ring (PMDA); however, these groups exhibit a detrimental effect on the thermal stability of the corresponding PAIs. The DSC and XRD characterizations of the obtained PIs showed that PI-BAET-BTDA has lower crystallinity than PI-BAET-PMDA and PI-BAET-OPDA. Among the three PIs, PI-BAET-OPDA exhibits the highest inherent viscosity. BAET is an inexpensive, low toxic, and nonvolatile diamine; therefore, its application in PAI synthesis is valuable. These PAIs can be used as membrane-modifying agents for industrial separation applications.
4. EXPERIMENTAL SECTION

4.1. Materials. BAET was prepared from waste PET bottles. PMDA, BTDA or carbonyldipthalic anhydride, and ODPA were obtained from Sigma-Aldrich (Munich, Germany). PMDA was recrystallized from acetic anhydride. NMP, DMAC, DMSO, glacial acetic acid, and other solvents were purchased from Sigma-Aldrich (Munich, Germany).

4.2. Reaction of BAET with Dianhydrides. BAET (0.5000 g) was added to a 50 mL round-bottom-flask containing 5 mL of solvent, and then, an equimolar quantity of dianhydride was added slowly to the reaction mixture. The heterogeneous reaction mixture gradually turned into a transparent solution and then became more viscous and opaque.

The intermediate was isolated by treating the transparent viscous solution with 60 mL of cool methanol. The formed solid was filtered and further rinsed with 20 mL of methanol for six times and then rinsed with acetone and finally dried under a vacuum at 60 °C for 24 h. The isolated intermediate or reaction mixture was converted into a PI by a stepwise thermal treatment in an oven.

4.3. Characterization of the Product. An FTIR-TENSOR II Bruker spectrometer (Bruker Optik GmbH, Ettlingen, Germany) was used in transmission mode to record the spectra from KBr pellets. The 1H NMR and 13C NMR spectra were recorded with a Bruker ARX-500 NMR spectrometer operating at 500 MHz (1H) and 125 MHz (13C).

DSC was performed with a METTLER STARE SW 11.00 instrument (Mettler-Toledo Ltd., Thailand). The samples were heated from RT to 400 or 450 °C at a heating rate of 10 °C/min in a nitrogen atmosphere. TG analysis was carried out with a Q500 Universal V4.5A (TA Instrument, NewCastle, DE, USA) by heating from RT to 800 °C at a heating rate of 10 °C/min in a nitrogen atmosphere.

Wide-angle X-ray diffractograms were obtained by a D8-ADVANCE Bruker X-ray diffractometer using Ni-filtered Cu Kα radiation (40 kV, 40 mA). The scanning rate was 0.01°/0.2 s. The samples were measured as fine powders on a silicon sample holder.

ASSOCIATED CONTENT

Supporting Information
The Supporting Information is available free of charge at https://pubs.acs.org/10.1021/acsomega.0c00799.

1H NMR and 13C NMR spectra; isomers of AA and DSC curves (PDF)

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Notes

The authors declare no competing financial interest.

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