

# Synthesis of Cardo Based Poly(arylene ether)s for Flexible Plastic Substrates and Their Properties

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New poly(arylene ether)s (PAEs) with both transparency and heat-resistance were prepared by a polycondensation of FBPODS, an ordered-sequence aromatic dihalide, and cardo typed aromatic diols containing fluorene and/or adamantane moiety and also non-cardo typed 1,5-naphthalene diol. The resulting polymers had their glass transition temperatures ranged from 202 to 247 °C. Based on TGA data, they exhibited excellent thermal stabilities, showing 5% weight loss at 434-487 °C. They had low thermal expansion coefficients of 58-59 ppm at temperature range of 50-200 °C as well as good mechanical properties with moduli of 1757-2143 MPa. The optical transmittance for the PAE films was over 70% at 550 nm, except for the PAE that contains naphthalene moiety (30% at 550 nm). They also showed water uptake of about 0.68% regardless of their chemical compositions. Therefore, the newly developed PAEs show strong potential as plastic substrates for flexible devices for display, solar cell and e-paper.

**Key Words :** Flexible display, Cardo based poly(arylene ether)s, Transparency, High heat resistance, Low CTE

## Introduction

Transparent and heat resistant polymer materials have gained much attention for their possible applications in various high-tech industries such as space, electronics, and semiconductor. The flexibility of the materials combined with transparency and high heat resistance makes such polymers one of the strongest candidates for a substrate for next generation displays, solar cells, and e-paper.<sup>1,2</sup>

Due to their technological importance, there have been intensive efforts to develop transparent and heat-resistant polymer films such as crystalline polyethylene terephthalate and polyethylene naphthalate or amorphous fluorene based polyarylate (Arylite<sup>R</sup>, Ferrania), polynorbornene typed cycloolefin copolymer (Appear<sup>R</sup>-3000, Promerus), polyimide (Neopulim<sup>R</sup> L-343, Mitsubishi Gas Chem), and poly(ether sulfone) (PES<sup>R</sup>, Sumitomo Bakelite). However, these polymers do not satisfy all the requirements as a substrate for high performance devices in an active matrix (AM) mode such as low CTE and water uptake, excellent transparency and easy processibility as well as high heat resistance.<sup>3-5</sup>

Amorphous poly(arylene ether)s (PAEs) can be another strong candidate with their high optical transparency, easy chemical modification, and excellent thermal and mechanical properties as well as good processibility.<sup>6-8</sup> In this paper, we explore new PAEs with both transparency and improved thermal properties.

PAEs are prepared by a nucleophilic substitution polycondensation between novel aromatic diol and dihalide monomers. In particular, cardo typed aromatic diols, which contain a bulky group such as fluorene,<sup>6,8</sup> or adamantane<sup>9-11</sup> moiety in the center of the monomer structure, have been

widely employed to produce corresponding PAEs which show good solubility and high optical transparency without any decrease of thermal stability. It also has been reported that the incorporation of trifluoromethyl, sulfonyl, ketone, and/or ether moieties between benzene rings activates aromatic halide monomers and consequently accelerates a nucleophilic displacement reaction by generating aromatic diphenolate ions from the aromatic diols and results in a high molecular weight polymer during the polycondensation.<sup>12-14</sup>

As a series of our research for flexible plastic substrates,<sup>15</sup> this article reports new cardo-based PAEs produced from a nucleophilic substitution reaction of 4-fluoro-4'-(((fluorobenzoyl)phenyl)-oxy)diphenyl sulfone) (FBPODS), an ordered sequence monomer, with cardo typed aromatic diols such as 9,9-bis(4-hydroxyphenyl)fluorene (BHPF) and 2,2-bis(4-hydroxyphenyl)adamantane (BHPA) or conventional naphthalene diol. Their thermal and mechanical properties, transparency, solubility, and water uptake will also be described.

## Experimental Section

**Materials.** BHPF supplied by Aldrich was recrystallized twice from methylene chloride (MC) and then used as a cardo typed aromatic diol monomer. 4,4'-Difluorodiphenyl sulfone (FPS) was recrystallized from ethanol. 4-Fluoro-4'-hydroxybenzophenone (FHB) were purified by recrystallization from toluene. Sulfolane, anhydrous potassium carbonate, 1,5-dihydroxynaphthalene (1,5-NP) and other chemicals were used as received from Aldrich without further purification. 2-Adamantanone, 1-hexanethiol were used as received from TCI without further purification.

**Instruments.**  $^1\text{H-NMR}$  spectra were recorded on a Varian Gemini-400 spectrometer. Differential scanning calorimetry (DSC) was measured on a TA DSC Q-100 thermal analyzer at a heating rate of  $20\text{ }^\circ\text{C}/\text{min}$ . Thermogravimetric analysis (TGA) measurement was carried out with a TA thermal analyzer at a heating rate of  $20\text{ }^\circ\text{C}/\text{min}$ . X-ray diffraction pattern was obtained on a Bruker D8 Discover using  $\text{Cu K}\alpha$  radiation. Linear coefficient of thermal expansion (CTE) of 40–60  $\mu\text{m}$  thick polymer films was measured with a TA TMA (Thermo-Mechanical Analyzer) Q-400 at a heating rate of  $5\text{ }^\circ\text{C}/\text{min}$  after annealing the samples for 1 h at the temperature  $5\text{ }^\circ\text{C}$  higher than the  $T_g$  of the film samples. Transmittance of the films was recorded on a UV-vis spectrometer (Varian Cary-100). Mechanical properties (ASTM D882) of the films were studied on an Instron Model 4201 tensile tester at a strain rate of  $10\text{ mm}/\text{min}$ . Polymer molecular weights were determined in THF solutions on a Waters GPC against polystyrene beads. Water absorption (ASTM D570) was calculated from the difference between the dry weight and the wet weight of  $1 \times 1\text{ cm}$  polymer film templates after immersion into water for 24 h in a thermostat at  $23\text{ }^\circ\text{C}$ .

**Synthesis of Ordered-Sequence Monomer FBPODS.** FBPODS was synthesized as described in our previous paper and its chemical structure matched well with the reported data: yield, 85%; mp,  $162\text{ }^\circ\text{C}$ .<sup>16</sup>

**Synthesis of 2,2-Bis(4-hydroxyphenyl)adamantane (2,2-BHPA).** 2,2-BHPA (mp  $318\text{ }^\circ\text{C}$ ) was prepared in 33% yield using  $\text{H}_2\text{SO}_4$  by modifying the procedure reported in the papers.<sup>10,17</sup> A 50 mL two-neck flask equipped with a nitrogen inlet adapter, a reflux condenser and a rubber serum was charged with 2-adamantanone (4.0 g, 26.6 mmol), phenol (18.74 mL, 213 mmol), 1-hexanethiol (1.49 mL, 26.6 mmol) and  $\text{H}_2\text{SO}_4$  (1.49 mL, 26.6 mmol). The mixture was slowly heated to  $80\text{ }^\circ\text{C}$ , stirred for 24 h and then cooled down to  $50\text{ }^\circ\text{C}$ . The solution was poured into 20 mL of water and extracted with chloroform three times. The combined organic layers were washed with 40 mL of water three times and dried over  $\text{MgSO}_4$ . The solvent was removed under reduced pressure and the resulting solid was recrystallized from ethanol and dried in vacuo to obtain pure 2,2-BHPA.

$^1\text{H-NMR}$  ( $\text{DMSO-}d_6$ , ppm): 9.02 (s, 2OH), 7.13 (m, 4H), 6.56 (d, 4H), 3.16 (s, 2H), 1.89 (d, 4H), 1.72 (s, 2H).

**Polymer Synthesis.** A 25 mL three-neck flask equipped with a condenser, and a nitrogen inlet adapter was charged with 0.7 g (2.19 mmol) of 2,2-BHPA, 1.00 g (2.22 mmol, 1 mole % excess vs. 2,2-BHPA) of FBPODS, and 15.83 mL of sulfolane. After stirring for 30 min at  $80\text{ }^\circ\text{C}$ , 0.32 g (2.33 mmol, 5 mole % excess vs. FBPODS) of anhydrous potassium carbonate, a weak base, was added to the reaction mixture. The mixture was stirred for 1 h at  $80\text{ }^\circ\text{C}$  which was followed by stirring under nitrogen atmosphere at  $160\text{ }^\circ\text{C}$  for 5 h and then at  $220\text{ }^\circ\text{C}$  for 10 h. After the polymerization was completed, the viscous solution was cooled down to  $30\text{ }^\circ\text{C}$  over 1 h and dissolved in 10 mL of chloroform to remove residual potassium carbonate and potassium fluoride formed during the polymerization. The filtrate solution was poured

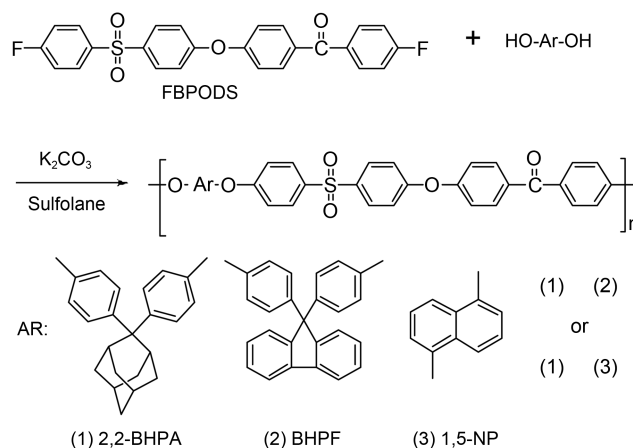
into 400 mL of methanol to precipitate white polymer precipitates. The crude polymer was washed with acetone and hot water 3 times, redissolved in chloroform, reprecipitated in methanol, and dried in vacuo at  $80\text{ }^\circ\text{C}$  for 24 h to obtain a fiber-shaped polymer (1.24 g). Other polymers were also prepared under the same conditions.

## Results and Discussions

**Synthesis of New Cardo Based PAEs.** In general, CTE, heat resistance, and mechanical strength and modulus of a polymer are negatively correlated to its transparency and solubility. In this paper, we present the new polymers obtained from cardo typed aromatic diols and aromatic difluoride FBPODS, which exhibit excellent thermal and mechanical properties as well as good optical transparency.<sup>16</sup>

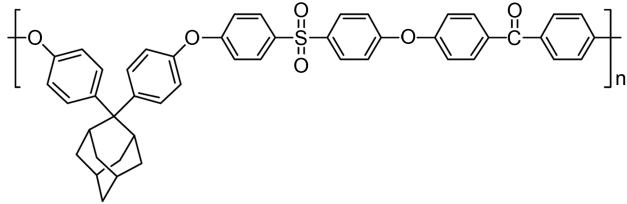
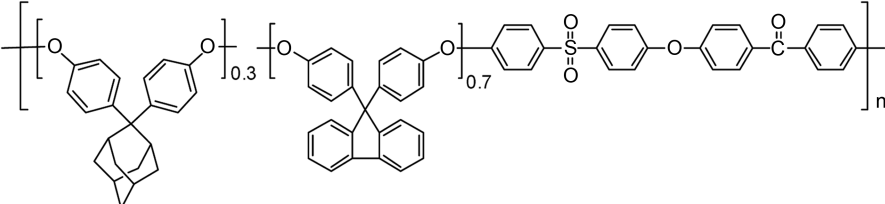
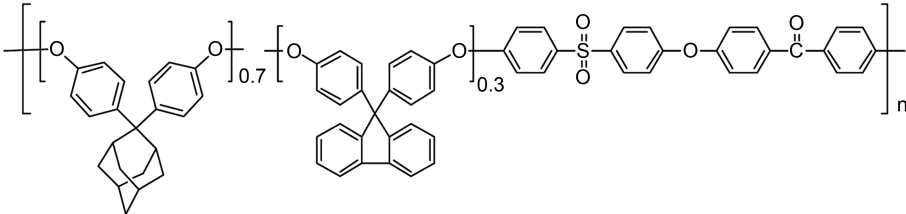
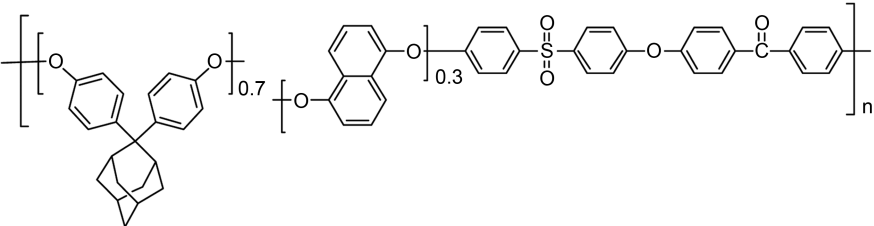
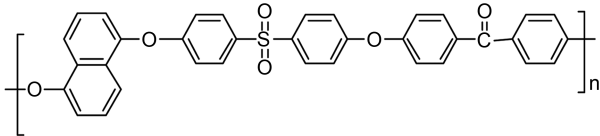
The cardo monomers used in this study were BHPF and 2,2-BHPA that contains fluorene, or adamantane moiety (so-called cardo group) in the center of monomer and two phenol groups next to the cardo group. FBPODS is composed of consecutive benzene rings intercalated with sulfone, ether, and ketone groups. These monomers were polymerized by nucleophilic substitution polycondensation to prepare new cardo based PAEs as shown in Scheme 1.

In order to produce the PAE-1, 2,2-BHPA as a cardo typed aromatic diol, was first polymerized with FBPODS in sulfolane solvent in the presence of potassium carbonate as a weak base. The weak base was used to minimize hydrolytic side reactions responsible for the branched polymers formation.<sup>18</sup> We obtained the best result when potassium carbonate was in 5.0 mol % excess relative to BHPF. To decrease the amount of branched polymers produced even further, the polymerization was carried out at  $160\text{ }^\circ\text{C}$  for 5 h and then at  $220\text{ }^\circ\text{C}$  for 10 h. In addition, 1.0 mol % excess of FBPODS against BHPF was used since FBPODS suppresses the formation of the branched polymers arising from the bisphenoxide ion of BHPF. Eventually, at such conditions, low polydispersity (low CTE) of polymers can be obtained. Therefore, 2,2-BHPA, 1,5-NP, or the mixture of the cardo monomers was also polymerized with FBPODS using the same condition described as above to obtain PAE-2, -3, -4,



**Scheme 1**

**Table 1.** Structure and results of new cardo based PAEs

PAE	Repeating unit	Yield (%)	M <sub>n</sub>	M <sub>w</sub>	M <sub>w</sub> /M <sub>n</sub>
PAE-1		75	21900	33300	1.52
PAE-2		86	40300	55300	1.37
PAE-3		85	27000	40800	1.51
PAE-4		81	21600	32900	1.52
PAE-5		80	-	-	-

or -5. Among the new PAEs, PAE-5 obtained from 1,5-NP and FBPODS was not only insoluble in most solvents such as chloroform, THF, toluene, ethyl acetate and so on but also very deep in color so that any properties were not measured except for its thermal property by using TGA and DSC.

As summarized in Table 1, the weight average molecular weights of the corresponding PAEs were in the range of 32900-55300. The GPC chromatogram of the new PAEs showed single, relatively, narrow peaks (polydispersity index of 1.37 to 1.52), indicating that no branching reactions occurred during the polymerization.<sup>6,8</sup> This is a noticeable improvement over other kinds of cardo typed PAEs which have wide molecular weight distributions.<sup>12,19</sup>

The structure of the PAEs was confirmed by FT-IR and <sup>1</sup>H-NMR spectra [Supplementary Materials (SM), Fig. 1] and they coincided reasonably well with the structure of repeating units shown in Scheme 1. This proves that the backbones of the newly synthesized PAEs are comprised of ordered arrays of cardo typed PAE, amorphous poly(ether

sulfone) (PES), and crystalline poly(ether ketone) (PEK). However, the real chemical microstructure of the polymers is likely to be a random distribution of head-to-tail and head-to-head structures due to the unsymmetrical structure of FBPODS.

All of the new PAEs were found to be amorphous by their wide angle X-ray diffraction patterns (SM Fig. 2). Also, DSC thermograms (SM Fig. 3) show no evidence of crystal melting, which are indicative that the new PAEs are of amorphous morphology. Amorphous characteristics of the polymers can be explained by the structural irregularity of cardo typed aromatic diols that employ bulky chains and the tetrahedral aryl sulfonyl linkages which will inhibit the ordered packing of the polymer main chain.

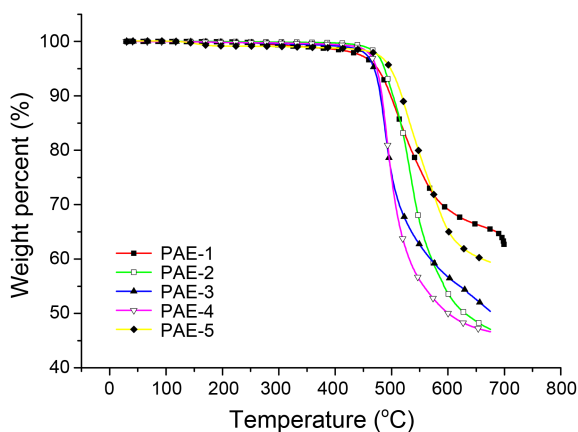
**Thermal Properties.** Table 2 describes the thermal properties of the new PAEs. The T<sub>g</sub> obtained by DSC for the cardo typed PAE-1, -2, -3, and -4 ranged from 232 to 247 °C. PAE-1 containing 2,2-BHPA revealed the highest T<sub>g</sub> of 247 °C. However, PAE-5 prepared from non-cardo typed mono-

**Table 2.** Thermal properties of new cardo based PAEs

PAE	T <sub>g</sub> (°C)	Temp. of 5% wt loss (°C)	Temp. of 10% wt loss (°C)	Temp. of 20% wt loss (°C)	Char. wt % at 600 °C
PAE-1	247	434	502	526	69
PAE-2	244	487	480	493	53
PAE-3	243	469	482	494	56
PAE-4	232	472	518	547	50
PAE-5	202	497	498	535	65

mer 1,5-NP and FBPODS showed the lowest T<sub>g</sub> (202 °C). Additionally, the T<sub>g</sub> of PAE-2 (244 °C), -3 (243 °C), or -4 (232 °C), which is composed of the mixture of 2,2-BHPA and BHPF or of 2,2-BHPA and 1,5-NP, was almost consistent with a theoretical value (246, 246, or 234 °C, respectively) estimated by the Fox equation [ $1/T_g = (w_1/T_{g1}) + (w_2/T_{g2})$ ]: Where w<sub>1</sub> and w<sub>2</sub> are the mass fractions of monomer units existing in copolymers.<sup>20</sup> This verifies that PAE-2, -3, and -4 have a random distribution of the aromatic diol monomers inside the polymer chains. The T<sub>g</sub> of the PAE-1, -2, -3, and -4 were lower than those of conventional Appair<sup>R</sup> (330 °C), Arylite<sup>R</sup> (325 °C), and Neopulim<sup>R</sup> (303 °C) but higher than that of PES (223 °C).<sup>1,3</sup>

According to TGA thermograms shown in Figure 1, the new PAEs showed high thermal stability as summarized Table II. The temperature for the 5% and 10% weight losses was in the range of 434–497 °C and 480–518 °C, respectively, depending on their structure. All the PAEs displayed high char yields of 50–69% at 600 °C under a nitrogen atmosphere. Among them, especially, PAE-5 exhibited the most excellent thermal stability even though its T<sub>g</sub> was lower than other PAEs. These results show that the chemical structure of a polymer exerts different influence on its phase transition temperature and thermal stability, respectively. That is, a phase transition temperature such as T<sub>g</sub> depends on the interaction and free volume between polymer chains, but thermal stability such as decomposition temperature mainly correlates with their morphology and bond strength. Therefore, non cardo typed monomer 1,5-NP, which has a smaller molecular size compared to the other cardo typed monomer

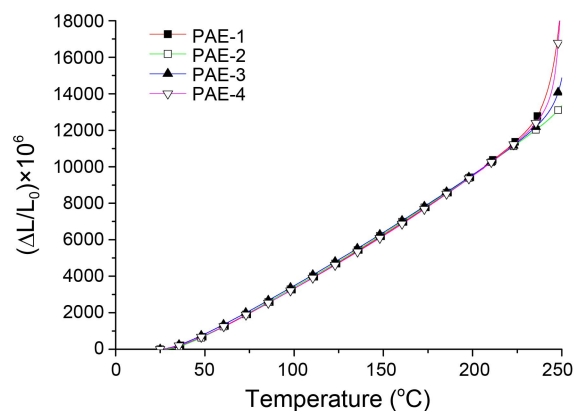
**Figure 1.** TGA thermograms of the new PAEs: (■), PAE-1; (□), PAE-2; (▲), PAE-3; (▼), PAE-4; (◆) PAE-5.

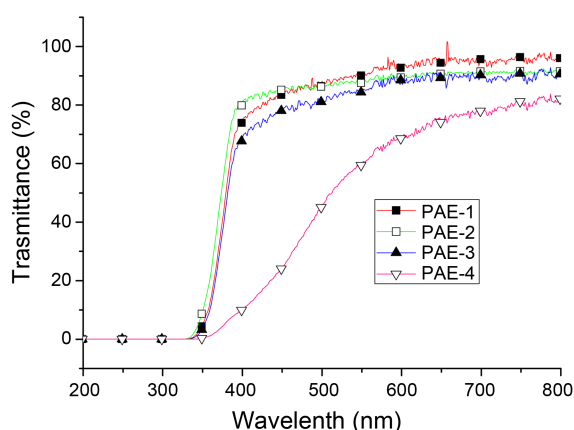
BHPF and 2,2-BHPA, reduces the free volume and forms microcrystalline phase between the polymer chains to improve its thermal stability.

**Dimensional Stabilities.** In general, CTE ( $\alpha$ ), indicating the dimensional stability of a polymer, is defined by the following equation:  $(\Delta L/L_0) = (\alpha_m \cdot \Delta T)$  where L<sub>0</sub> is the specimen length at room temperature;  $\Delta L$  is a change in length between room temperature and experimental temperature;  $\Delta T$  is the temperature interval between room temperature and experimental temperature; and  $\alpha_m$  is a the mean CTE.<sup>21</sup> In this research, CTE along the length (Y) direction of the film (8.0 mm × 4.0 mm) was measured by using a TMA (SM Fig. 4).

Figure 2 shows the change of the length (L<sub>0</sub> = 8.0 mm) of the PAE films as a function of temperature, which is measured from room temperature to their T<sub>g</sub>: The new PAEs had CTE values between 58 and 59 ppm at a specific temperature range, which varies with the T<sub>g</sub> of the PAEs used.

PAE-1 containing adamantane moiety (2,2-BHPA) showed very low CTE (58.2 ppm) at a relatively wide temperature range (50–200 °C), which is much lower than that of the poly(BHPF/FBPODS) (74 ppm) from BHPF and FBPODS, which was synthesized in our previous research.<sup>15</sup> Furthermore, it is very interesting that PAE-3 exhibited somewhat lower CTE (58.0 ppm) than PAE-1, even though 30 mol % of bulky fluorene group was introduced into the PAE-1. This is likely due to the fact that the compact adamantane groups of 70 mol % occupy the vacant space (so-called free volume) resulting from the bulky fluorene groups located perpendicular to the polymer main chain, and these results

**Figure 2.** The change of length of the film along Y direction versus temperature for the new PAEs: (■), PAE-1; (□), PAE-2; (▲), PAE-3; (▼), PAE-4.



**Figure 3.** UV-VIS Spectra of the new PAEs: (■), PAE-1; (□), PAE-2; (▲), PAE-3; (▽), PAE-4.

are in agreement with our earlier research.<sup>15</sup> However, as the content of BHPF introduced in the polymer chain raised from 30% (PAE-3) to 70% (PAE-2), CTE value is inferior from 58.0 to 58.7 ppm. Additionally, PAE-4, which contains 30% of naphthalene groups, exhibited the lowest CTE (57.9 ppm). This result meets to our expectation that naphthalene group, which forms microcrystalline structure in the polymer chain, would lower its CTE value. Such microcrystalline phase made it difficult to fabricate the PAE-4 film and the resulting film had low optical transmittance. These CTE values are comparable to those of the conventional advanced polymers such as Arylite<sup>R</sup> (ca. 63 ppm at 200 °C), PES, (ca. 67 ppm at 200 °C), or Neopulim<sup>R</sup> (ca. 58 ppm at 150 °C).<sup>1,3</sup>

On the basis of these CTE values, dimensional stability of the PAEs is likely to be closely related to the amount of free volume involved in the polymer chains and also to the polydispersity of the polymer rather than their  $T_g$  which is affected directly by the bending and rotational mobility of the polymer chain.

**Mechanical Properties.** Tensile properties of the PAEs are shown in Table 3. Moduli of the PAEs had values between 1757 and 2143 MPa and their elongation at break was in the range of 3–21%. Among them, PAE-2 showed the highest modulus, which is comparable to those of Arylite<sup>R</sup> (2900 MPa), Neopulim<sup>R</sup> (ca. 2230 MPa), and PES<sup>R</sup> (2200 MPa).<sup>1</sup> In strength-strain curves (SM Fig. 5) for the PAEs, PAE-2 and -3 showed decreased tensile stress (34.7 and 51.2 MPa, respectively) at the yield point and thereafter to elongate. This demonstrates that PAE-2 and -3 has tough-ductile mechanical properties. The modulus of PAE-1 containing

**Table 3.** Tensile properties of new cardo based PAEs

PAE	Modulus (MPa)	Tensile stress at break (MPa)	Elongation at break (%)
PAE-1	1964	50	3.3
PAE-2	2143	38.2	21.2
PAE-3	1957	47.2	9.5
PAE-4	1757	42.9	3.4

cage typed adamantane group (2,2-BHPA) was 1964 MPa, which is close to that of Appear<sup>R</sup> (1900 MPa), which contains similar cage typed norbornane moiety. The modulus of PAE-2 and -3, a random copolymer of PAE-1 and poly-(BHPF/FDPODS) with the modulus of 2350 MPa,<sup>15</sup> are fairly proportional to the composition ratio of each polymer. However, PAE-4 with naphthalene group had the lowest modulus value. These moduli results showed a good agreement with the DSC and TGA behaviors, which are closely related to the  $T_g$  and thermal stability of the PAEs.

**Solubility and Film Transparency.** The cardo based aromatic diols were soluble in most solvents besides ethyl acetate and THF, but FBPODS was freely soluble in most solvents except for methanol, water, and *n*-hexane. The PAEs were soluble not only in high polar solvents such as sulfolane, DMF, DMSO, and NMP but also in common solvents like THF and chloroform.

The film should retain the transparency of > 80% at 450–800 nm in order to be applied as flexible plastic substrates for display, solar cell, and e-paper.<sup>2</sup> As shown in Figure 2, all the new films, which were cast from chloroform solution (3.5 wt %), showed a high optical transmittance of > 70% in the wavelength range 450–800 nm. In particular, PAE-1, -2, and -3 films displayed very high transparency of 91, 88, 85% at 550 nm, respectively. These transmittance values were similar to those of the advanced materials such as Arylite<sup>R</sup> (91%), Appear<sup>R</sup> (91%), and Neopulim<sup>R</sup> (90%) and higher than that of PES<sup>R</sup> (89%).<sup>1</sup> PAE-4 film showed a very low optical transmittance of 60% at 550 nm due to its microcrystalline phase derived from the naphthalene units in the polymer chain.

**Water Uptake.** Normally, it has been reported that the flexible plastic substrates should keep very low moisture permeability (below 0.1 g per 1.0 m<sup>2</sup> film a day) to be incorporated in an AM-TFT device for display or e-book device. The novel PAEs had low water absorption of about 0.68% regardless of their chemical composition. The water uptake value is somewhat higher than that of Arylite<sup>R</sup> (0.4%), but much more lower than those of Neopulim<sup>R</sup> (2.1%) and PES<sup>R</sup> (1.4%).<sup>3</sup>

## Conclusion

New amorphous poly(arylene ether)s were synthesized by a nucleophilic substitution reaction between sequentially ordered aromatic difluoride FBPODS and cardo based aromatic diols. Some of the resulting polymers exhibited high optical transparency of over 85% at 550 nm as well as excellent thermal and dimensional stabilities, which are comparable to those of advanced materials such as Arylite<sup>R</sup>, Appear<sup>R</sup>, Neopulim<sup>R</sup>, and PES<sup>R</sup>. Therefore, the newly developed PAEs show strong potential as plastic substrates for flexible devices for display, solar cell, and e-paper.

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### References

1. MacDonald, W. A. *J. Mater. Chem.* **2004**, *14*, 4.
  2. Saran, N.; Parikh, K.; Suh, D. S.; Muñoz, E.; Kolla, H.; Manohar, S. K. *J. Am. Chem. Soc.* **2004**, *126*, 4462.
  3. Ito, H.; Oka, W.; Goto, H.; Umeda, H. *Jpn. J. Appl. Phys.* **2006**, *45*, 4325.
  4. Liu, P.; Wu, Y.; Li, Y.; Ong, B. S.; Zhu, S. *J. Am. Chem. Soc.* **2006**, *128*, 4554.
  5. Angiolini, S.; Avidano, M.; Bracco, R.; Barlocco, C.; Young, N. D.; Trainor, M.; Zhao, X. M. *SID 03 DIGEST* **2003**, *47*, 1.
  6. Huang, W. Y.; Liaw, B. R.; Chang, M. Y.; Han, Y. K.; Huang, P. T. *Macromolecules* **2007**, *40*, 8649.
  7. Hayashi, H.; Takizawa, A.; Ikeda, K.; Takarada, W.; Kikutani, T.; Koyama, Y.; Takata, T. *Polym. J.* **2009**, *41*, 609.
  8. Kawasaki, S.; Yamada, M.; Kobori, K.; Jin, F.; Kondo, Y.; Hayashi, H.; Suzuki, Y.; Takata, T. *Macromolecules* **2007**, *40*, 5284.
  9. Liaw, B. R.; Huang, W. Y.; Huang, P. T.; Chang, M. Y.; Han, Y. K. *Polymer* **2007**, *48*, 7087.
  10. Pixton, M. R.; Paul, D. R. *Polymer* **1995**, *36*, 3165.
  11. Mathias, L. J.; Lewis, C. M.; Wiegel, K. N. *Macromolecules* **1997**, *30*, 5970.
  12. Chen, G.; Zhang, S.; Chen, T.; Wu, Y. *J. Appl. Polym. Sci.* **2007**, *106*, 2808.
  13. Seesukphronrarak, S.; Kawasaki, S.; Kobori, K.; Takata, T. *J. Polym. Sci. Part A: Polym. Chem.* **2007**, *45*, 3073.
  14. Seesukphronrarak, S.; Takata, T. *Polym. J.* **2007**, *39*, 731.
  15. Kong, G. P.; Hong, B. R.; Kim, M. K.; Han, Y. K.; Chun, H. A. *Macromolecular Research* **2011**, *19*(6), 608.
  16. Han, Y. K.; Chi, S. D.; Kim, Y. H.; Park, B. K.; Jin, J. I. *Macromolecules* **1995**, *28*, 916.
  17. Kawasaki, S.; Yamada, A.; Kobori, K.; Sakamoto, H.; Kondo, Y.; Jin, F.; Takata, O. *J. Appl. Polym. Sci.* **2009**, *111*, 461.
  18. Devaux, J.; Daoust, D.; Legras, R.; Dereppe, J. M.; Nield, E. *Polymer* **1989**, *30*, 161.
  19. Chern, Y. T.; Shiue, H. C. *Macromolecules* **1997**, *30*, 4646.
  20. Fox, T. G. *Bull. Am. Phys. Soc.* **1956**, *1*, 123.
  21. Deng, D. Q.; Xu, L. *Cryogenics* **2003**, *43*, 465.
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