

Effect of Chemical Structure and Preparation Process on the Aggregation Structure and Properties of Polyimide Film

Ying Wang, Yang Yang, Zhenxing Jia, Jiaqiang Qin, Yi Gu

College of Polymer Science and Engineering, Sichuan University, Chengdu 610065, People's Republic of China

Correspondence to: J. Qin (E-mail: jqin@scu.edu.cn)

ABSTRACT: In this article, polyimide (PI) films were fabricated via the three-step method including the reactions of condensation polymerization, chemical imidization, and thermal imidization. In comparison with the conventional two-step method to produce PI films, there was an additional step in the present method, i.e., chemical imidization. The aim of chemical imidization was to get PI intermediates with different pre-imidization degree (pre-ID). And PI component in PI intermediates acted as *in-situ* rigid-rod segments and induced orientation in the films of PI intermediates. Then the orientations of molecular chains were preserved in the following thermal imidization, and caused the difference in aggregation structure and property of the final PI films. The test results indicated that the orderly degree of molecular chains and mechanical properties of PI films increased with pre-ID increasing. Furthermore, this tendency was much more obvious for more rigid backbone structure. © 2012 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* 000: 000–000, 2012

KEYWORDS: polyimides; films; crystallization; mechanical properties; thermal properties

Received 3 March 2012; accepted 17 May 2012; published online

DOI: 10.1002/app.38068

INTRODUCTION

Aromatic polyimides (PIs) have been widely used in technology fields due to their excellent thermal stability, good dimensional stability, outstanding mechanical properties, and high resistance to chemicals and radiations.^{1–4} PI molecular chains consist of abundant imide and aromatic rings. Besides of common Van der Waals force, there are other interactions among the molecular chains, such as charge transfer interaction, preferred layer packing and mixed layer packing.⁵ The insoluble and infusible characteristics of PIs resulted from the above factors make them difficult to process. So far, most of PI films are prepared from the conventional two-step method, and the derived products are almost amorphous without special treatment.⁶ Thus, the mechanical properties were not high enough to meet the demand of high-performance PI films in high-tech applications.

Orderly structures such as orientation and crystallization can dramatically enhance the mechanical and thermal properties, and decrease linear coefficients of thermal expansion (CTE) of polymer materials. Thermal cure of a slightly drawn film of poly(amic acid) (PAA) showed marked spontaneous orientation toward the stretching direction.⁷ Parallel to the draw direction the modulus of PMDA-ODA PI films increased, while the modulus decreased perpendicular to the draw direction.⁸ Kochi et al.⁹ prepared a high-modulus and high-strength PI material by a

technique of thermal imidization after cold drawing of PAA films. The studies^{10–12} on the spontaneous molecular orientation of PI showed CTE decreased almost linearly with an increase in the degree of in-plane orientation.

In order to increase the degree of crystallinity, researchers designed the backbone structure of PI in two ways: to increase the mobility of molecular chains and the amount of rigid molecular chains. Since molecular chains could move freely in solution, PI particles obtained from imidization in solution were highly crystalline.¹³ It has been found that introduction ether linkage or carbonyl group into diamine could improve the mobility of molecular chains and increase the orderly degree.^{14–16} However, the flexible groups of PI main chains give rise to decreasing of mechanical properties. To enhance the degree of crystallinity and mechanical properties of PI simultaneously, some rigid-rod segments which could easily induce the formation of crystalline structure were introduced to the backbone, and found the crystallinity, in-plane orientation, hardness and modulus of the PI films increased with increasing contents of rigid-rod segments.^{17–19}

PAA converts to PI by thermal or chemical imidization, and the intermediate in imidization is a copolymer composed of amic acid and imide, called copolyamic acid-imide (PAA-PI).²⁰ According to the previous papers,^{20–22} the mechanical properties of PI materials can be enhanced with the precursor (PAA)

Additional Supporting Information may be found in the online version of this article.

© 2012 Wiley Periodicals, Inc.

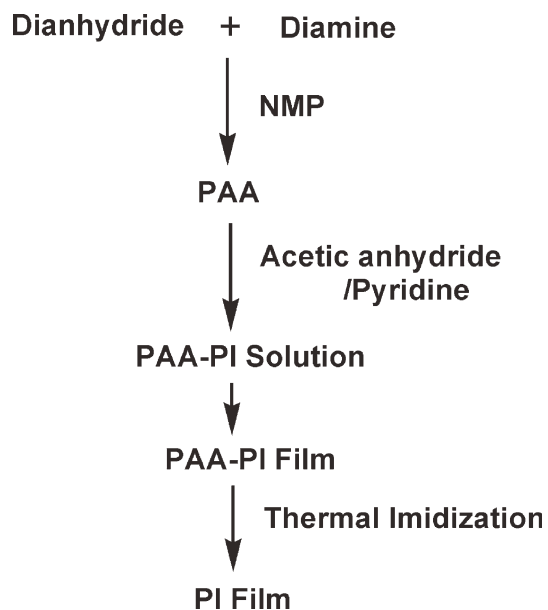


Figure 1. Schematic process for producing polyimide films via the PAA-PI precursors with different imidization degrees.

partly imidized. Qin et al.²³ found amic acid-imide copolymer with different contents of carboxyl group could be controlled. Zhai et al.^{20,24} studied the phase separation in PAA-PI solution and the morphologic ordering of the final PI films derived from PAA-PI solutions with different imidization degree. However, these works have not systematically investigated the aggregation structure, mechanical property and their relationship. Imide group is rigid and can act as *in-situ* rigid-rod segments. We conjecture that PI films based on PAA-PI precursors may have different aggregation structure and properties.

In this article, we selected three PI systems possessing different rigidity of backbone. The condensation polymerization of diamine and dianhydride to give poly(amic acid) (PAA), then dehydrating reagent was added to chemically convert PAA into partly imidized PAA. The partly imidized PAA solution was coated onto glass plates, and fully imidized to produce PI films by thermal imidization. The aggregation structure and properties were carefully measured. Moreover, the three different PI systems were studied and compared in detail to reveal the effect of chemical structure and preparation process on the aggregation structure and property of the final PI films.

EXPERIMENTAL SECTION

Materials

3,3',4,4'-oxydiphthalic anhydride (ODPA), 3,3',4,4'-biphenyltetracarboxylic dianhydride (BPDA) and 4,4'-oxydianiline (ODA) were supplied by Synthetic Resin Research Institute, Shanghai, China. 3,3',4,4'-benzophenonetetracarboxylic dianhydride (BTDA) was purchased from Acros Organics of New Jersey, USA. ODPA, BTDA, and BPDA were recrystallized from the mixture of acetic anhydride and acetic acid (3/1, v/v), then dried at 180°C for 4 h prior to use. ODA was recrystallized from ethanol and dried under N₂ atmosphere before use. *N*-methyl-2-pyrrolidinone (NMP, from Qunli Chemical, Shanghai,

China) was distilled under reduced pressure over phosphorous pentoxide. Other reagents, such as acetic anhydride, acetic acid, pyridine and ethanol, purchased from Chengdu Kelong Chemical, were used without further purification.

Preparation of PAA-PI Precursors and Polyimide Films

As a typical experimental procedure for the synthesis of PI film (see Figure 1), an equimolar amount of dianhydride was stepwise added into a solution of ODA and anhydrous NMP in a three-necked flask equipped with a mechanical stirrer at room temperature. The solid content was restricted at 10%. After the mixture was continuously stirred for 12 h, viscous PAA solution obtained was divided equally into six parts, and different mole amount of dehydration reagents (100, 80, 60, 40, 20, and 0 mol %; mole ratio of acetic anhydride/pyridine = 5/4), which is relative to original amino groups was added. After a predetermined time (as described later), viscous and homogeneous PAA-PI solutions with different pre-IDs were cast onto glass plates, which were then thermally baked in a vacuum oven at 80°C for 2 h, 140°C for 2 h, 220°C for 2 h, and 300°C for 2 h, successively. The free-standing polyimide films were stripped from the glass substrates with the help of hot water followed by drying in an oven at 100°C. The synthesized PI films with different pre-ID of 100, 80, 60, 40, 20, and 0 mol %, were abbreviated as PI-100, PI-80, PI-60, PI-40, PI-20, and PI-0, successively.

Three different PI systems were studied and compared together. Their chemical structures were shown in Figure 2, and assigned to be A, B, and C, respectively.

Characterizations

Fourier transform infrared (FTIR) spectra were recorded on a Nicolet 560 Fourier transform spectrophotometer with the wave number ranging from 400 to 4000 cm⁻¹. Imidization degree (ID) of dried PAA-PI sample was determined according to a reported method, in which the band of 1380 cm⁻¹ was selected for quantifying ID, and the aromatic band at 1500 cm⁻¹ (C—C stretching of the *p*-substituted benzene backbone) was selected

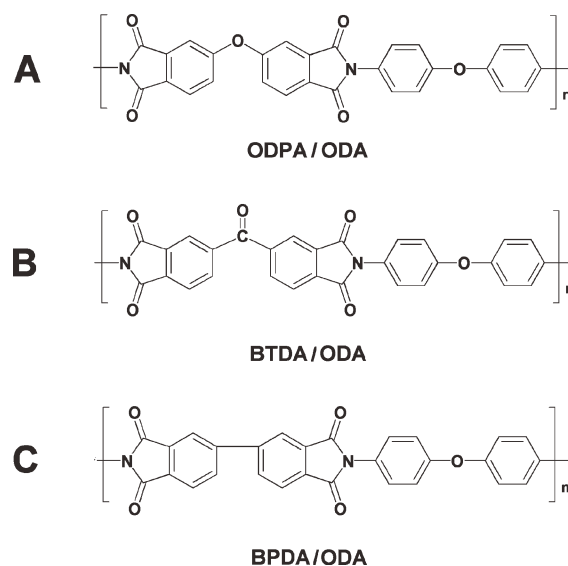


Figure 2. Structures of polyimide.

Table I. Theoretical and Experimental Imidization Degree of PAA-PIs

Imidization degree (%)						
Theoretical	0	20	40	60	80	100
Experimental	A	0	28	46	55	73
	B	0	14	34	57	80
	C	0	14	37	62	84

as the internal standard. ID was calculated using the following equation:

$$ID(\%) = \frac{(S_{1380}/S_{1500})_{T=50^{\circ}\text{C}}}{(S_{1380}/S_{1500})_{T=300^{\circ}\text{C}}} \times 100 \quad (1)$$

Where S is the area of absorption band, $T = 50^{\circ}\text{C}$ is the cured temperature of the samples, $T = 300^{\circ}\text{C}$ is taken as the temperature of completely imidized PI.

Mechanical tensile properties of PI films were performed at room temperature on an Instron-4320 Tensile Apparatus. Samples of PI film were cut into 50 mm × 10 mm strips along the film casting direction. Average values of more than five measurements were taken for each sample. The experimental uncertainties in tensile strength and modulus were ± 1 MPa and ± 0.05 GPa, respectively.

The wide-angle X-ray diffraction (WAXD) was performed at room temperature on an X-ray diffractometer (X²-Pert-MPD, Philip) with a graphite monochromator (operating at 40 kV and 40 mA) with nickel-filtered with Cu/K-α1 radiation ($\lambda = 0.15418$ nm). Data were collected for a continuous scan at a rate of 2°/min over a range of $2\theta = 5^{\circ}\sim 50^{\circ}$.

Differential scanning calorimetry (DSC) was recorded on a Netzsch STA 449C thermal analysis system in nitrogen at a heating rate of 10°C/min. Dynamic mechanical analysis (DMA) were recorded on a Netzsch DMA 242C in nitrogen at a heating rate of 5°C/min, a frequency of 2 Hz.

RESULTS AND DISCUSSION

Synthesis of PAA-PI Precursors and Determination of Imidization Degree

PI films is generally prepared by a two-step method, that is, PAA derived from the condensation polymerization of aromatic

diamine and dianhydride followed by thermal imidization to produce PI. In this article, PI films were obtained from the partly imidized precursors, i.e., PAA-PI, as shown in Figure 1.

Insoluble gel may form as chemical imidization progressing owing to the decreasing solubility of PAA-PI. Because of the different rigidity of molecular chains, the solubility of PAA-PI with the fixed pre-ID is different in these PI systems, and it increases in the order of $C < B < A$. To obtain freestanding PI films, the PAA-PI solution should be cast on substrate then baked before gel appears. But the time of chemical imidization (t_c) should be as long as enough to give PAA-PI solution with the desired pre-ID. Hence t_c was different among these PI systems, which was set as 8, 2, and 1 h for A, B, and C system, respectively.

Determination of Imidization Degree

FTIR is used widely for determination of reaction extent.^{25–27} We also used it to determine the ID of dried PAA-PI precursors. All the thin film samples were baked at 50°C for 24 h to remove most of the solvent. With the pre-ID increases from 0 to 100%, the results indicate that the intensity of characteristic bands of PAA (1655 cm^{-1}) decreases gradually, and those of PI (1780, 1724, and 1380 cm^{-1}) increase. The FTIR spectrum of PAA-PI precursors were given in Supporting Information (Supporting Information S1-A, S1-B, and S1-C). It is known that isoimide rings form accompany with imide ones in chemical imidization. However, the absorbance band of isoimide (1800 cm^{-1}) has not been detected in FTIR spectrum of PAA-PI films. Hence the main product obtained from chemical imidization is PAA-PI, and isoimide can be ignored.

According to a reported method,²⁸ the band at 1380 cm^{-1} (stretching vibration of C–N) was selected for quantifying pre-ID, and the aromatic band at 1500 cm^{-1} (C–C stretching of the *p*-substituted benzene backbone) was selected as the internal standard. All the experimental pre-IDs are calculated according to the eq. (1) and summarized in Table I, in which it can be seen that all the experimental data are well coincident with the theoretical ones. These results state clearly that the pre-IDs can be successfully controlled by adjusting the amount of dehydrating reagents.

Mechanical Properties of the Final PI Films

The mechanical properties of the final PI films were measured, and the results of A, B, and C systems were summarized in Figure 3. It can be seen that tensile strength, tensile moduli and

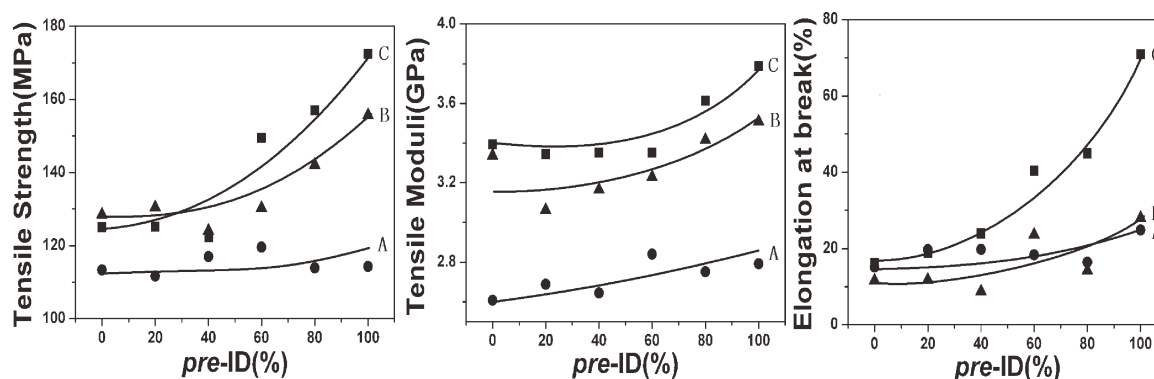


Figure 3. Mechanical properties of polyimide films with different pre-IDs: (A) ●, ODPA-ODA, (B) ▲, BTDA-ODA, (C) ■, BPDA-ODA.

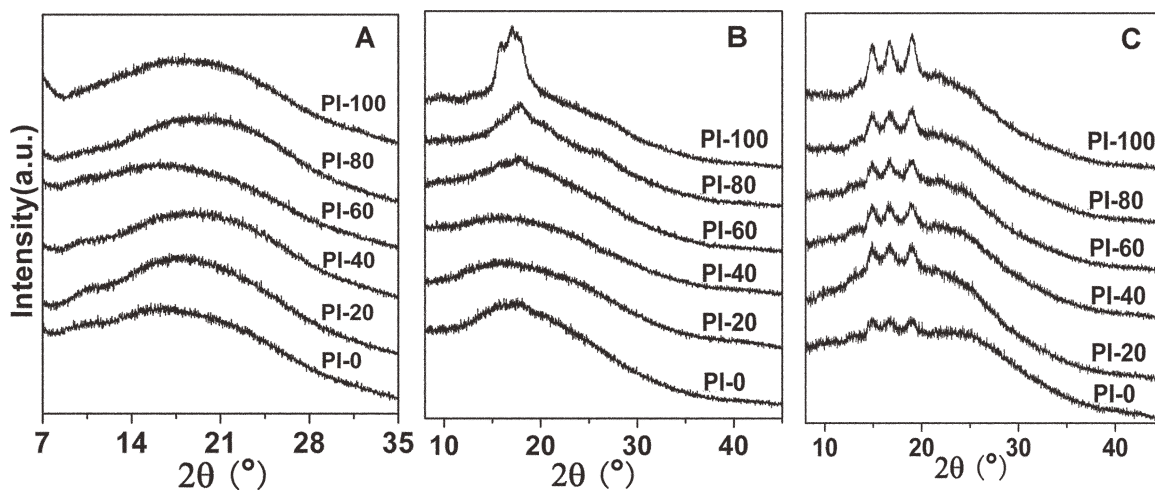


Figure 4. WAXD pattern of polyimide films with different pre-IDs: (A) ODPA-ODA, (B) BTDA-ODA, (C) BPDA-ODA.

elongation at break increase with increasing pre-IDs for all PI systems, and they increase faster in the order of $A < B < C$. Compared with PI-0, the tensile strength of PI-100 respectively increases by 1.0%, 23.2%, and 37.9% for A, B, and C system, and those data of the tensile moduli are 7.1%, 7.6%, and 11.7%, and they are 63.7%, 158.6%, and 335.8% for elongation at break. Because of the transformation of isoimide to imide under heat treatment,²² coupled with the FTIR results, the possibility of different chemical structure of the final PI films in the fixed system can be excluded. As mentioned above, rigid imide segments formed in the step of chemical imidization. These rigid segments induce the regular packing of molecular chains. So the enhancement of mechanical properties could be attributed to the orderly structure formed in PI films. By comparing A, B, and C systems, it can be seen that PI with a higher rigidity is more affected by this preparation process.

Aggregation Structure of the Final PI Films

The aggregation structures of the final PI films were measured by WAXD. The results suggest that PI films based on ODPA-ODA [Figure 4(A)] display amorphous peaks, and the relative intensity of these peaks reveals a slight increase with the increasing pre-ID. Compared with ODPA-ODA PI films, the intensity of BTDA-ODA ones [Figure 4(B)] shows a much more significant increase as the pre-ID increase from 0% to 100%. And the sample of PI-100 displays a rather strong peak which indicates the formation of crystalline structure. Those PIs of BTDA-ODA [Figure 4(C)] show three diffraction peaks at the range of $15^\circ \sim 20^\circ$ (14.82° , 16.66° , 18.94° , respectively), which indicate the crystalline structures exist in films, and the increase of intensity of diffraction peaks is the most obvious one among the three systems. According to a reported literature method,²⁹ we calculated the degrees of crystallinity of PI films. For PI films based on BPDA-ODA, as the pre-IDs increase from 0% to 100%, the degrees of crystallinity of PI films are 7.0%, 7.0%, 10.0%, 11.0%, 12.9%, and 18.0%, respectively. And for PI-100 of PI (BTDA-ODA), the degree of crystallinity is 7.3%. The other samples are beyond the calculation range because they have

amorphous peaks in the WAXD patterns. These results suggest that the aggregation structure of PI films can be readily controlled by adjusting the amount of dehydrating reagents in the preparation procedure.

In a fixed PI system, the content of rigid segments increases with PAA converting to PI. Rigid structure restricts the mobility of molecular chains. And the reduction of the mobility of molecular chains is favorable for their regular packing.^{30,31} In other words, the orientation extent of molecular chains increases with increasing pre-ID. It is also reasonable to assume that the T_g of the film raises significantly with increasing pre-ID. And this prevented the relaxation of the molecules and retained the induced orientation.³² While molecular reorganization occurs during the heating required for imidization. ODPA has the most flexible structure due to the ether linkage, BTDA with carbonyl group is more rigid than ODPA, and BPDA is the most rigid one because of the biphenyl structure, as shown in Figure 2. The hindrance potentials for internal rotation of aromatic ether and ketone are only of the order of 1-3 kcal/mol while the potential for biphenyl is ca. 10 kcal/mol.⁹ Flexible functional groups may contribute to the relaxation of orderly packing in thermal imidization.^{33,34} In other words, for the samples with a fixed pre-ID, the orientation extent of BPDA-ODA PIs is the largest among the three PI systems. Thus, the preparation process has the greatest effect on the most rigid PI system,³⁵ corresponding to the results described above.

It is reasonable that the orderly packing of molecular chains contributes to the enhancement of tensile strength and moduli. In addition, the plastic of deformation of aromatic polyimide can be explained in terms of local order consisting of bundles of parallel-packed main chains.⁹ Thus, PI film with a higher orderly degree of molecular chains should have a higher value of elongation at break. Moreover, all of PI films reveal relatively weak diffraction peaks as well as the big amorphous halo, thus the diameter of crystals forms in PI films is very small. Hence, PI films derived from this pre-imidization process form many micro-crystals [especially for PI (C) films and PI-100 of PI

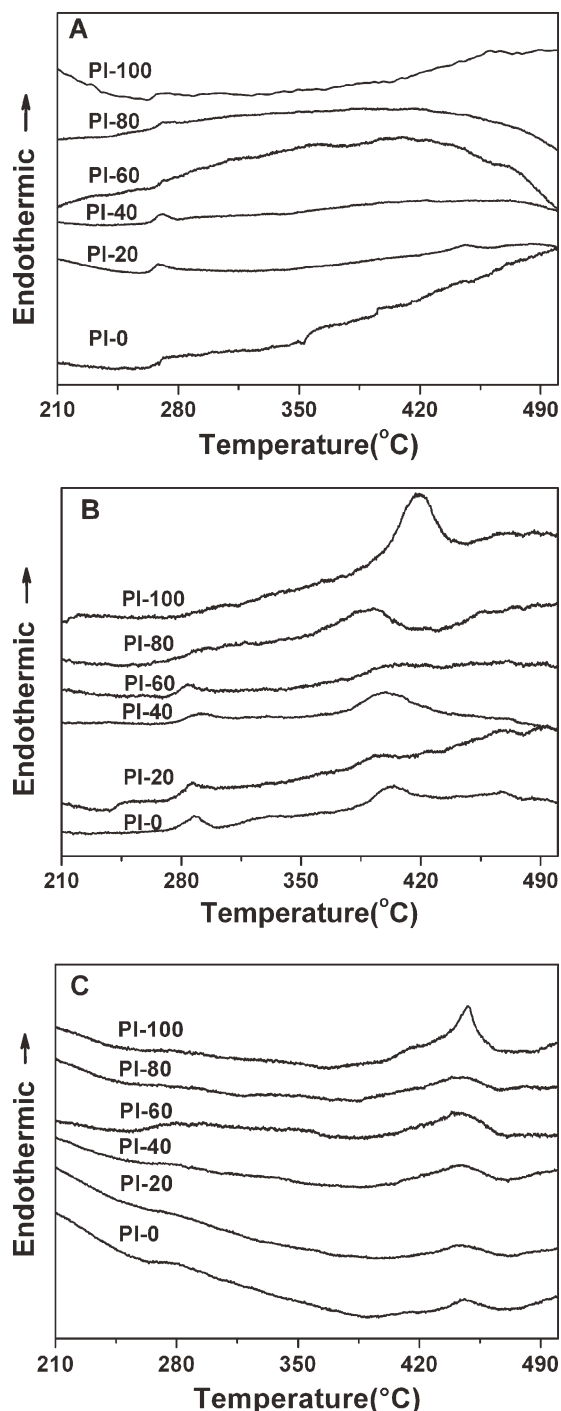


Figure 5. DSC results of PI films with different pre-IDs: (A) ODPA-ODA, (B) BTDA-ODA, (C) BPDA-ODA.

(B)], which could reinforce as well as toughen these PI films. The results of aggregation structure are in well accordance with the data of mechanical properties shown in the last section.

Thermal Properties of the Final PI Films

DSC measurement was conducted with the final PI films to confirm the formation of crystalline structure proposed above. As can be seen in Figure 5, PI films based on B and C systems

show obvious melt endothermic peaks at ca. 400 and 450°C, respectively, and the relative height of these peaks increase with increasing pre-ID. While there is no such peak presenting in those PIs of A system. Melting enthalpy is proportional to the degree of crystallinity of material. The melting enthalpies of PI films from B and C systems are calculated from the DSC curves by the software of NETZSCH-TA4 (there are no melting peaks in the DSC curves of PI (A) films, so they could not be analyzed in quantitative). With the pre-ID increases from 0 to 100%, the melting enthalpies of PI (B) films are 2.47, 5.25, 6.54, 7.42, 9.05, 13.91 J/g; the melting enthalpies of PI (C) films

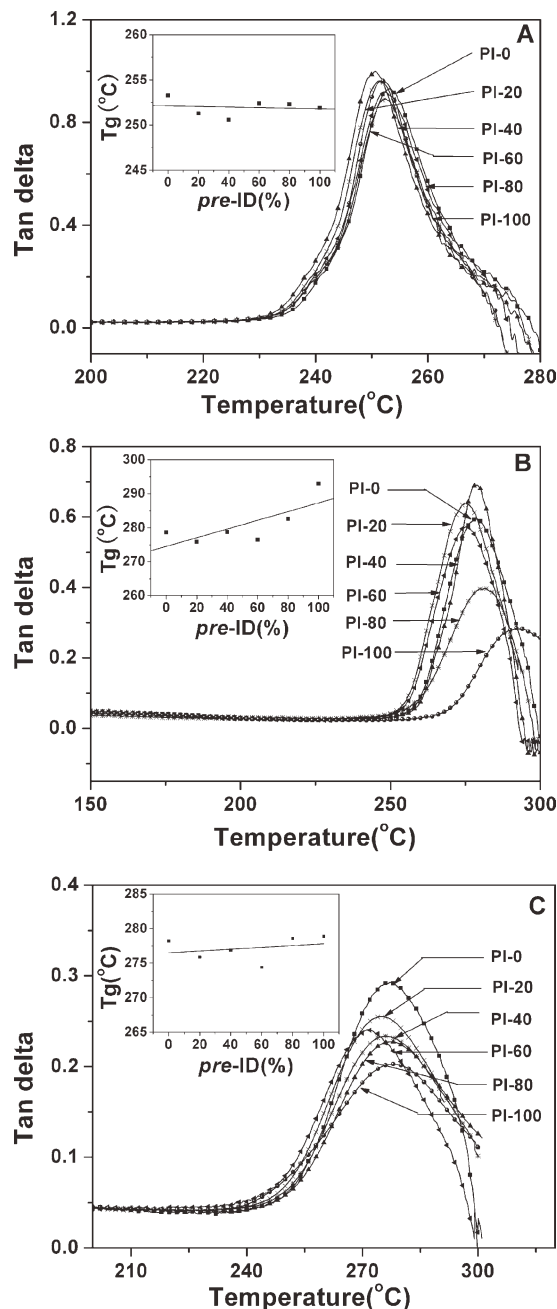


Figure 6. DMA results of PI films with different pre-IDs: (A) ODPA-ODA, (B) BTDA-ODA, (C) BPDA-ODA.

are 3.70, 5.49, 10.73, 10.46, 15.67, 20.55 J/g. It indicates the degree of crystallinity of PI (B) and (C) films increase with increasing pre-ID. The results successfully confirm the crystalline structure in BTDA-ODA and BPDA-ODA PI films, and the amorphous morphology in ODPDA-ODA PI films, corresponding well with the WAXD data.

Furthermore, the glass transitions show at ca. 266, 288, and 270°C for A, B and C PI films, respectively. And it becomes less obvious with increasing pre-ID. It is reasonable to assume that this trend of glass transitions could be attributed to two aspects. On one hand, the free volume decrease due to the regular packing of molecular chains. As can be seen in WAXD and DSC results, the orderly degree of PI films enhances with increasing pre-ID. That is, the molecular chains are more compact with higher pre-ID. On the other hand, the orderly packing of molecular chains, especially crystalline structure, restricts the mobility of molecular chains. Therefore, the glass transitions become less obvious with increasing pre-ID.

Figure 6 shows the relationship of $\tan \delta$ and temperature. The maxima of $\tan \delta$ curves which correspond with T_g , are shifted to the direction of higher temperature with increasing pre-IDs. It indicates that the mobility of the PI films is much more restricted with increasing pre-IDs. The orderly packing of PI chains leads to a decrease in the free volume between the molecular chains. Thus, PI films with higher pre-IDs have higher T_g , corresponding well with DSC result. Moreover, the magnitude of $\tan \delta$, which is the ratio of loss modulus and storage modulus, indicates the relative amount of mechanical energy dissipated during cyclic stress in DMA testing. A material is perfectly elastic if $\tan \delta$ is zero; perfectly viscous if $\tan \delta$ is infinite; and equally elastic and viscous if $\tan \delta$ is one.^{36,37} The value of $\tan \delta$ decreases as the pre-ID increases from 0 to 100% in a fixed PI system. It means PI molecular chains become less elastically with higher pre-IDs. The free movement of the molecular chains is restricted by the orderly structure, resulting in an increase in rigidity and mechanical properties. In addition, the value of $\tan \delta$ shows at ca. 0.9~1.0, 0.2~0.7, and 0.2~0.3 for A, B, and C system, respectively, corresponding well to their chemical structures. Generally, polyimide films prepared without the process of stretching have very low degree of crystallinity. In this article, these films derived from pre-imidization process also have relatively low degree of crystallinity, which could be seen from the above quantitative analysis of WAXD results. Hence, the difference of T_g s from DSC and DMA is minor. However, the trends of increasing T_g s and decreasing $\tan \delta$ with increasing pre-IDs are still obvious.

CONCLUSIONS

A method of chemical imidization followed by thermal imidization was adopted to produce PI films. The imidization degree of PAA-PI precursors could be successfully controlled in the step of chemical imidization. Orderly molecular chains were induced by the rigid imide segments and preserved in thermal imidization, leading to the various aggregation structure and properties of the final PIs. The molecular chains arranged more

regularly with higher pre-ID, resulting in an increase in mechanical properties, an increase in T_g , and a decrease in elasticity. And the influence of this preparation process is greater for much more stiff PI system, that is, the variation extent enhanced with the increasing order as follows: ODPDA-ODA < BTDA-ODA < BPDA-ODA. This work provided the theoretic indicators for controllable preparing of high-performance polyimide films.

ACKNOWLEDGMENTS

This work was financially supported by the National Natural Science Foundation of China (Grant No. 50803041) and the Cooperation Project in Industry, Education and Research of Guangdong Province and Ministry of Education of China (2011A090200014).

REFERENCES

- Mittal, K. L. *Polyimides: Synthesis, Characterization, and Applications*; New York: Plenum, **1984**.
- Ghosh, M. K.; Mittal, K. L. *Polyimides: Fundamentals and Applications*; New York: Dekker, **1996**.
- Mittal, K. L. *Polyimides and Other High Temperature Polymers*; Utrecht: VSP, **2001**.
- Ding, M. X. *Polyimides: Chemistry, Relationship between Structure and Properties and Materials*; Chinese Science Press: Beijing, **2006**.
- Baklagina, Y. G.; Milevskaya, I. S. *Polyamic Acids and Polyimides, Synthesis, Transformations and Structures*; CRC Press: Boca Raton, **1993**.
- Denisov, V. M.; Svetlichnyi, V. M.; Gindin, V. A.; Zubkov, V. A.; Kol'tsov, A.; Koton, M. M.; Kudryavtsev, V. V. *Polym. Sci. U.S.S.R.* **1979**, *21*, 1644.
- Hasegawa, M.; Okuda, K.; Horimoto, M.; Shindo, Y.; Yokota, R.; Kochi, M. *Macromolecules* **1997**, *30*, 5745.
- Hardaker, S. S.; Samuels, R. J. *J. Polym. Sci. Part B: Polym. Phys.* **1997**, *35*, 777.
- Kochi, M.; Yokota, R.; Iizuka, T. Y.; Mita, I. *J. Polym. Sci. Part B: Polym. Phys.* **1990**, *28*, 2463.
- Ishii, J.; Shimizu, N.; Ishihara, N.; Ikeda, Y.; Sensui, N.; Matano, T.; Hasegawa, M. *Eur. Polym. J.* **2010**, *46*, 69.
- Ebisawa, S.; Ishii, J.; Sato, M.; Vladimirov, L.; Hasegawa, M. *Eur. Polym. J.* **2010**, *46*, 283.
- Ishii, J.; Takata, A.; Oami, Y.; Yokota, R.; Vladimirov, L.; Hasegawa, M. *Eur. Polym. J.* **2010**, *46*, 681.
- Nagata, Y.; Ohnishi, Y.; Kajiyama, T. *Polym. J.* **1996**, *28*, 980.
- Brandom, D. K.; Wilkes, G. L. *Polymer* **1995**, *36*, 4083.
- Brillhart, M. V.; Cebe, P. J. *Polym. Sci. Part B: Polym. Phys.* **1995**, *33*, 927.
- Hsiao, B. S.; Kreuz, J. A.; Cheng, S. Z. D. *Macromolecules* **1996**, *29*, 135.
- Lee, C.; Iyer, N. P.; Han, H. J. *Polym. Sci. Part B: Polym. Phys.* **2004**, *42*, 2202.

18. Kim, Y.; Ree, M.; Chang, T.; Ha, C. S.; Nunes, T. L.; Lin, J. S. *J. Polym. Sci. Part B: Polym. Phys.* **1995**, *33*, 2075.
19. Khatua, S. C.; Maiti, S. *Eur. Polym. J.* **2002**, *38*, 537.
20. Zhai, Y.; Yang, Q.; Zhu, R. Q.; Gu, Y. J. *Mater. Sci.* **2008**, *43*, 338.
21. Su, J. F.; Chen, L.; Tang, T. T.; Ren, C. B.; Wang, J. J.; Qin, C. X.; Dai, L. X. *High Perform. Polym.* **2011**, *23*, 273.
22. Park, S. K.; Farris, R. J. *Polymer* **2001**, *42*, 10087.
23. Qin, J. Q.; Zhao, H.; Liu, X. Y.; Zhang, X. Y.; Gu, Y. *Polymer* **2007**, *48*, 3379.
24. Zhai, Y.; Huang, Y.; Zhu, R. Q.; Gu, Y. *Chem. Eng. Comm.* **2010**, *197*, 289.
25. Pryde, C. A. *J. Polym. Sci. Part A: Polym. Chem.* **1989**, *27*, 711.
26. Fukukawa, K. I.; Ueda, M. *Macromolecules* **2006**, *39*, 2100.
27. Musto, P.; Ragosta, G.; Scarinzi, G.; Mascia, L. *Polymer* **2004**, *45*, 1697.
28. Marek, M.; Schmidt, P.; Schneider, B.; Bednář, B.; Králíček, J. *Makromol. Chem.* **1990**, *191*, 2631.
29. Godocikova, E.; Balaz, P.; Boldizarova, E. *Hydrometallurgy* **2002**, *65*, 83.
30. Jang, W.; Seo, J.; Lee, C.; Paek, S. H.; Han, H. J. *Appl. Polym. Sci.* **2009**, *113*, 976.
31. Jou, J. H.; Huang, P. T. *Macromolecules* **1991**, *24*, 3796.
32. Russell, T. P.; Gugger, H.; Swalen, J. D. *J. Polym. Sci. Polym. Phys.* **1983**, *21*, 1745.
33. Sensui, N.; Ishii, J.; Takata, A.; Oami, Y.; Hasegawa, M.; Yokota, R. *High Perform. Polym.* **2009**, *21*, 709.
34. Hasegawa, M.; Ishii, J.; Shindo, Y. *Macromolecules* **1999**, *32*, 6111.
35. Chung, H.; Joe, Y.; Han, H. J. *Appl. Polym. Sci.* **1999**, *74*, 3287.
36. Chou, W. J.; Wang, C. C.; Chen, C. Y. *Compos. Sci. Technol.* **2008**, *68*, 2208.
37. Muhammad, K.; Shaukat, S.; Zahoor, A. J. *Macromol. Sci. Part A* **2007**, *44*, 55.