

Water Sorption Behaviors of the BPDA-Based Polyimide Films Depending upon the Structural Isomers of Diamine

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Received 11 February 2000; accepted 23 June 2000

ABSTRACT: For the biphenyltetracarboxylic dianhydride (BPDA)-based polyimide thin films, the water sorption behaviors were gravimetrically investigated by using a thin film diffusion analyzer. The water sorption behaviors of the polyimide thin films are quite different and strongly dependent upon the sort of polyimide. The diffusion coefficients of the polyimide thin films vary in the range of 1.6×10^{-10} to $12.4 \times 10^{-10} \text{ cm}^2/\text{s}$ and the water uptakes vary from 1.52 to 5.25 wt %. Both the diffusion coefficient and water uptake of the polyimide thin films are in the increasing order: BPDA-*p*PDA < BPDA-*p,p'*ODA < BPDA-*p,m'*ODA < BPDA-*m*PDA ~ BPDA-*p,p'*DDS < BPDA-*m,m'*DDS. Specifically, the polyimide films with *para*-oriented linkages in backbone structure showed relatively lower diffusion coefficient and water uptake than the corresponding polyimide films with *meta*-oriented linkages because of the well-developed crystalline structure and good intermolecular chain ordering. In addition, the polyimide thin films having higher chain order showed relatively lower diffusion coefficient and water uptake. The crystallinity and intermolecular chain ordering in the morphological structure are critical parameters in controlling the water sorption behaviors of the polyimide thin films. © 2001 John Wiley & Sons, Inc. *J Appl Polym Sci* 79: 2121–2127, 2001

Key words: BPDA-based polyimide films; diffusion coefficient; water uptake; morphological structure

INTRODUCTION

High-temperature aromatic polyimides are used in the fabrication of microelectronic devices such as interdielectric, alpha-particle-protecting, and passivation layers, owing to relatively high thermal stability, high chemical resistance, and high mechanical toughness due to the aromatic ring and imide ring units on the chain backbone.^{1–4}

However, despite the relatively high chemical resistance characteristics, polyimides still absorb water.^{2–10} Specifically, the water sorption of the polyimide films causes reliability problems, such as displacement, package cracking, delamination, and mechanical failures in thin films. Above all, it increases dielectric constants, conductivities, and dielectric loss of the polyimide films. Also, the water sorption behaviors are closely related to stress relaxation of the polyimide thin films.¹¹ Therefore, the phenomenon of water sorption is an interesting subject in the design and selection of advanced structural materials for the fabrication of advanced microelectronic devices with higher performance and better reliability. One of

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Contract grant sponsor: Korea Research Foundation (KRF); contract grant number: 1998-001-E01258.

Journal of Applied Polymer Science, Vol. 79, 2121–2127 (2001)
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possible criterion of material for the applications of electronics might be based on how quickly, and to what extent, water permeates into it.

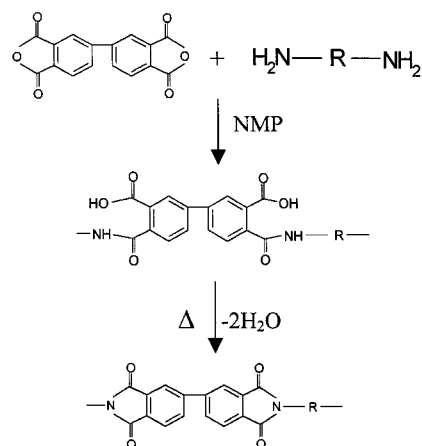
Since a representative poly(*p,p'*-oxydiphenylene pyromellitimide) (PMDA-ODA), which has been widely used in the microelectronic industry, was commercialized in the 1960s, many studies have been conducted on the development of new organic materials and optimization of process.^{2,3} Another high-performance polyimide, poly(*p*-phenylene biphenyltetracarboximide) (BPDA-PDA) is recently known to have superior chemical and physical properties,^{2,11} in comparison with the PMDA-based polyimides including PMDA-ODA. Also, for the development of many BPDA-based polyimides, many studies have been conducted and some of them have been commercialized.

The purpose of this study was to gain a better understanding of the relationship between the water sorption behaviors and the chemical/morphological structures of the polyimide thin films. To investigate this, the polyimide thin films prepared from BPDA as a dianhydride and two type diamines (*para*-oriented and *meta*-oriented): *p* and *m*-phenylene diamine (PDA), *p,p'* and *p,m'*-oxydianiline (ODA), and *p,p'* and *m,m'*-diaminodiphenyl sulfone (DDS), were gravimetrically investigated by using a thin film diffusion analyzer.^{11–19} The water sorption results were analyzed using a Fickian diffusion model in thin films and interpreted in terms of the chemical and morphological structures.

EXPERIMENTAL

Materials and Film Preparation

BPDA was supplied from Chriskev Co. and other monomers and solvent were purchased from the Aldrich Chemical Co. (Milwaukee, WI): PDA, ODA, DDS, and *N*-methyl-2-pyrrolidinone (NMP). Poly (*p*-phenylene biphenyltetracarboxamic acid) (BPDA-*p*PDA PAA) solution was prepared under a nitrogen atmosphere by adding BPDA to PDA in anhydrous NMP.^{1–4} Before polymerization, all the monomers were purified by the sublimation under reduced pressure. Other poly(amic acid)s were prepared in the same manner as BPDA-*p*PDA PAA was synthesized: BPDA-*m*PDA PAA, BPDA-*p,p'*ODA PAA, BPDA-*p,m'*ODA PAA, BPDA-*p,p'*DDS PAA, and BPDA-*m,m'* DDS PAA (see Fig. 1). The resulting solutions had a solid content of 15 wt %.



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
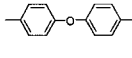
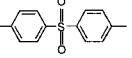

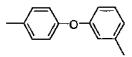
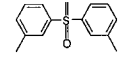
PDA	ODA	DDS
 <i>p</i> PDA	 <i>p,p'</i> ODA	 <i>p,p'</i> DDS
 <i>m</i> PDA	 <i>p,m'</i> ODA	 <i>m,m'</i> DDS

Figure 1 Synthesis of the BPDA-based polyimides.

All of the polyimide precursor solutions were spin coated on silicon (100) substrates. Coated wafers were prebaked at 80°C for 30 min on a hot plate. The prebaked samples were then placed in a curing oven and cured by the following cure schedule^{11–19}: 150°C/30 min, 230°C/30 min, 300°C/30 min, and 400°C/60 min.^{8–11} The ramping rate for each step was 2.5°C/min and the cooling rate was 2.0°C/min for the curing process. The thickness of polyimide thin films was controlled in the range 10–15 μm by using a spin coater. The thickness of the polyimide thin films was measured using a surface profiler (model P-10; Tencor Instruments Co.). Then, the cured films were taken off from the substrates with the aid of deionized water and washed with distilled water several times and dried under a vacuum. The fully cured films were cut into rectangular pieces approximately 10 × 15 mm and fully dried in a vacuum to use for the water sorption.

Water Sorption

For the fully cured polyimide thin films, the water sorption behaviors were measured at 25°C and

100% relative humidity (RH) as a function of time by using the Thin Film Diffusion Analyzer (model D-200; CAHN Instruments, Inc.) with the resolution of 0.1 μg over 20-mg weight loading. These were described in our previous literature.^{11–19} All the sorption isotherms were analyzed with Fick's second law given by Crank and Park^{20,21}:

$$\frac{M(t)}{M(\infty)} = 1 - \frac{8}{\pi^2} \sum_{m=0}^{\infty} \frac{1}{(2m+1)^2} \times \exp\left(\frac{-D(2m+1)^2\pi^2 t}{L^2}\right) \quad (1)$$

where $M(t)$ is the water sorption at a time t , $M(\infty)$ is the water sorption at $t = \infty$, D (cm^2/s) is the diffusion coefficient of water and polymer systems, and L is the film thickness, respectively. The diffusion coefficient and water uptake are obtained by simulating the normalized experimental data to eq. (1). Here, water uptake means the amount of sorbed water per unit mass of dried film in equilibrium state.

Film Characterization

For the morphological structure of the polyimide thin films, wide-angle X-ray diffraction (WAXD) patterns were corrected from a horizontal X-ray diffraction apparatus (D/MAX-200B; Rigaku Co.) with nickel-filtered radiation. One diffractometer was set up for the transmission mode (i.e., reflections from lattice planes normal to the film surface) with a thin asymmetric cut (101) quartz plate monochromator bent to a section of a logarithmic spiral and located in the diffracted beam. The other was used for the reflection mode (i.e., reflections from lattice planes parallel to the film surface) with a curved graphite monochromator in the diffracted beam. The $\text{CuK}\alpha$ radiation source ($\lambda = 1.54\text{\AA}$) was operated at 35 kV and 40 mA and all measurements were performed in $\theta/2\theta$ mode. The 2θ scan data were collected in the range of 5–60° at 0.02° intervals with a scan speed of 0.3–0.5° min^{-1} , depending upon whether a reflection or transmission scan was being made.

Dynamic mechanical thermal properties and glass transition temperatures (T_g s) of the BPDA-based polyimide thin films were measured in a nitrogen ambient over the range of 25–480°C, using a dynamic mechanical thermal analyzer (model Mark III; Polymer Laboratories Co.). The heating rate and frequency were 5.0 K/min and 1 Hz, respectively.

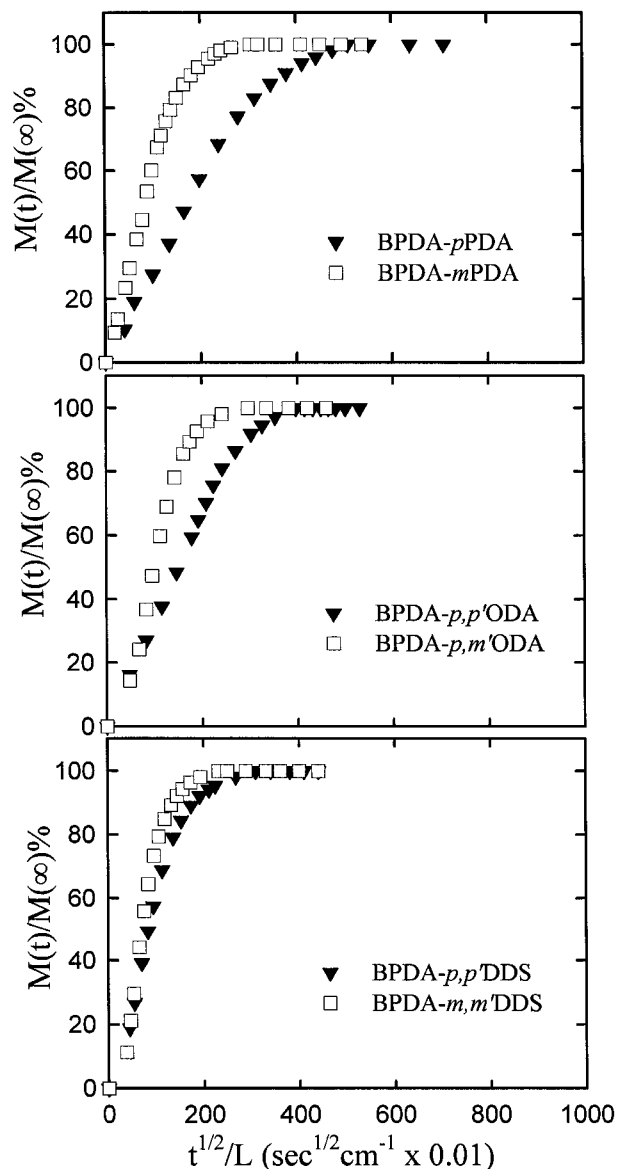


Figure 2 Water sorption isotherms for the BPDA-based polyimide thin films at 25°C and 100% RH.

RESULTS AND DISCUSSION

Water Sorption

The water sorption isotherms of the BPDA-based polyimide thin films were gravimetrically investigated at 25°C and 100% RH, and are depicted in Figure 2. All of the water sorption isotherms were reasonably well fitted by a Fickian diffusion model [i.e., eq. (1)], regardless of morphological heterogeneities^{11–19,22–24} due to the ordered and disordered phases. It indicates that the water sorption behaviors of the polyimide thin film

Table I Diffusion Coefficients and Water Uptakes of the BPDA-Based Polyimide Thin Films

Polyimide Structure	Film Thickness (μm)	Diffusion Coefficient $D \times 10^{-10}$ (cm^2/s)	Water Uptake (wt %)
BPDA- <i>p</i> PDA	14.08	1.6	1.52
BPDA- <i>m</i> PDA	10.81	7.4	4.75
BPDA- <i>p,p'</i> ODA	12.80	3.0	1.62
BPDA- <i>p,m'</i> ODA	10.94	6.7	2.32
BPDA- <i>p,p'</i> DDS	10.19	7.2	5.14
BPDA- <i>m,m'</i> DDS	12.03	12.4	5.25

could be macroscopically interpreted by using a Fickian diffusion model. Thus, the diffusion coefficient and water uptakes were simulated with eq. (1), and are summarized in Table I.

The diffusion coefficient and water uptake were estimated to be 1.6×10^{-10} cm^2/s and 1.52 wt % for BPDA-*p*PDA polyimide thin film and 7.4×10^{-10} cm^2/s and 4.47 wt % for BPDA-*m*PDA polyimide thin film, respectively. It indicates that a larger amount of water was diffused more quickly into the *meta*-oriented BPDA-*m*PDA polyimide thin film than the corresponding *para*-oriented BPDA-*p*PDA polyimide thin film. BPDA-ODA and BPDA-DDS polyimide thin films showed the same trends with the water sorption as BPDA-PDA polyimide thin films. That is, the *para*-oriented polyimide thin film (BPDA-*p,p'*ODA and BPDA-*p,p'*DDS) showed relatively lower diffusion coefficient and lower water uptake than the corresponding *meta*-oriented polyimide thin film (BPDA-*p,m'*ODA and BPDA-*m,m'*DDS). However, the isomeric BPDA-DDS polyimide thin films showed no significant difference in the water uptake, unlike BPDA-PDA and BPDA-ODA polyimide thin films. This suggests that there may be no apparent morphological difference in the isomeric BPDA-DDS polyimide thin films.^{15–19}

From the water sorption results, it was found that the polyimide thin films with *para*-oriented linkage (*p*, *p,p'*, or *p,p'*) showed relatively lower diffusion coefficient and water uptake than the corresponding polyimide thin films with *meta*-oriented linkage (*p*, *p,m'*, or *m,m'*). In addition, both water uptake and diffusion coefficient are quite different from each other depending on the sort of polyimide. The diffusion coefficients of water in thin films vary in the range of 1.6×10^{-10} to 12.4×10^{-10} cm^2/s , and are in the increasing order: BPDA-*p*PDA < BPDA-*p,p'*ODA < BPDA-*p,m'*ODA < BPDA-*p,p'*DDS \sim BPDA-*m*PDA < BPDA-*m,m'*DDS. The water uptakes vary from 1.52 to 5.25 wt %, and are in

the increasing order: BPDA-*p*PDA < BPDA-*p,p'*ODA < BPDA-*p,m'*ODA < BPDA-*m*PDA < BPDA-*p,p'*DDS < BPDA-*m,m'*DDS. These different water sorption behaviors of the polyimide thin films may be a function of several parameters^{8,9,11–19,25–28}: vapor pressure (RH), temperature, and characteristics of the sample itself, including film thickness, chemical structure, and morphology. In the present work, polymer films were prepared from the same sample preparation method. Accordingly, it is simply expected that the water sorption behaviors of the polyimide thin films may be solely dependent on the morphological and chemical structures.^{15–19}

Water sorption behaviors may be due to the differences in the chemical affinity to water molecules and the concentration of polar groups such as carbonyl group ($>\text{C}=\text{O}$), carboxyl group ($-\text{COOH}$), and sulfonyl group ($>\text{SO}_2$).^{29,30} It is well known that the solubility of a given polymer in various solvents is largely determined by its chemical structure, and structural similarity favors solubility itself. Also, the polymer films with higher content of polar groups have higher diffusion coefficient and water uptake. From the chemical affinity to water molecules and the content of polar group, one can simply expect that the water sorption is in the increasing order: BPDA-PDA < BPDA-ODA < BPDA-DDS. However, these results are not consistent with the water sorption behaviors of the polyimide thin films. Additionally, BPDA-*p*PDA has the similar chemical group, the similar value of solubility parameter, and the content of polar groups for the corresponding isomeric BPDA-*m*PDA. Despite the similarity in the chemical structure, however, there exist significant differences in the diffusion coefficient and water uptake of BPDA-*p*PDA and BPDA-*m*PDA polyimide thin films (see Table I). For BPDA-ODA and BPDA-DDS polyimide thin films, the water sorption results were also dependent on the struc-

Table II Glass Transition Temperatures of the BPDA-Based Polyimide Films

Polyimide Structure	Glass Transition Temperature (°C)
BPDA- <i>p</i> PDA	360
BPDA- <i>m</i> PDA	375
BPDA- <i>p,p'</i> ODA	295
BPDA- <i>p,m'</i> ODA	300
BPDA- <i>p,p'</i> DDS	400
BPDA- <i>m,m'</i> DDS	330

tural isomers of diamine. It suggests that the water sorption of the polyimide thin films may be significantly affected by the morphological structure: crystallinity, chain order, and packing order.^{15–19}

Morphological Structure

BPDA-*p*PDA/BPDA-*m*PDA, BPDA-*p,p'*ODA/BPDA-*p,m'*ODA, and BPDA-*p,p'*DDS/BPDA-*m,m'*DDS have different isomeric molecular structures (see Fig. 1). *Meta*-oriented polymers have higher configurational entropy because of larger degree of conformational freedom in main chain than do *para*-oriented polymers.³¹ These differences may lead to the inherent changes in the microstructure or morphology of the polyimide thin films. It was simply expected that the polyimide film with *para*-oriented linkages (*p*PDA, *p,p'*ODA, and *p,p'*DDS) would have more linearity than the corresponding polyimide film with *meta*-oriented linkages (*m*PDA, *p,m'*ODA, and *m,m'*DDS). However, highly linear chains have the potential to pack well as long as the T_g is low enough to permit some reorientation during annealing.^{15,17,18,25} All of the polyimide thin films showed lower or near T_g to final curing temperature and significant decrease in modulus (see Table II and Fig. 3). It indicates that the polyimide thin films with *para*-oriented linkages have relatively more linearity and more well-packed structure than those with *meta*-oriented linkages, which may induce to lower water sorption in the polyimide thin films with *para*-oriented linkages.

For the morphological structure of the BPDA-based polyimide thin films, WAXD measurements were performed. WAXD patterns are depicted in Figure 4. Here, the transmission pattern gives a structural information for the film plane, whereas the reflection pattern does a structural information for the out-of-film plane.^{11–19,32}

BPDA-*p*PDA exhibited multiple diffraction peaks in both the transmission and reflection patterns. However, the characteristic diffraction peaks in the transmission pattern, which correspond to the intermolecular ordering, are very prominent. It indicates that polymeric chains of BPDA-*p*PDA polyimide film are predominantly aligned in the film plane and have higher intermolecular packing order. However, WAXD patterns of BPDA-*m*PDA are significantly different from those of BPDA-*p*PDA. It showed no characteristic peaks in the transmission and reflection patterns, but only a big amorphous halo peak with flat region over the angle range of 10–35° (2 θ). It indicates that, though there exist some chances to form local ordering in polymer chain, BPDA-*m*PDA forms the amorphous structure. The transmission and reflection patterns of BPDA-*p,p'*ODA and BPDA-*p,m'*ODA polyimide thin films showed multiple diffraction peaks seated on a big amorphous halo peak in the diffraction patterns over the angle range of 10 to 30° (2 θ). In comparison, however, the diffraction peaks of the *meta*-oriented BPDA-*p,m'*ODA polyimide thin film are not so apparent. It indicates that *para*-oriented BPDA-*p,p'*ODA polyimide

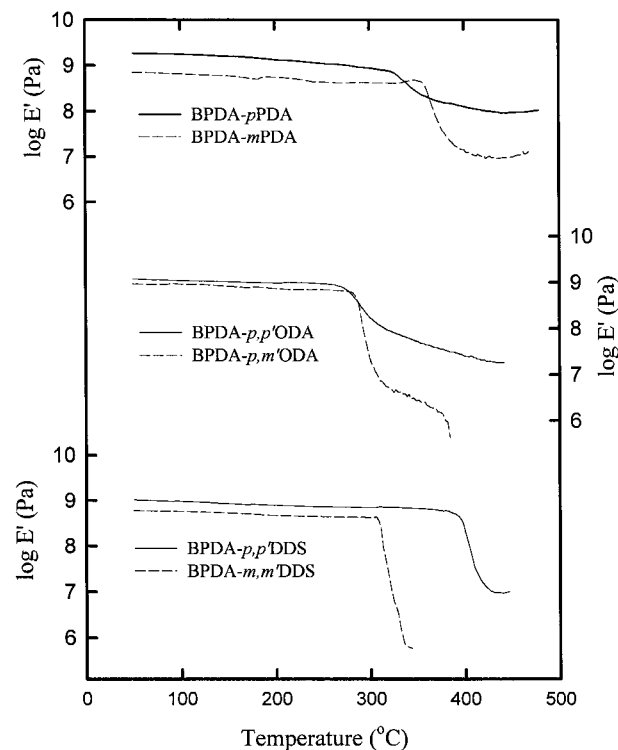


Figure 3 Variations of dynamic storage moduli with temperature for the BPDA-based polyimide films.

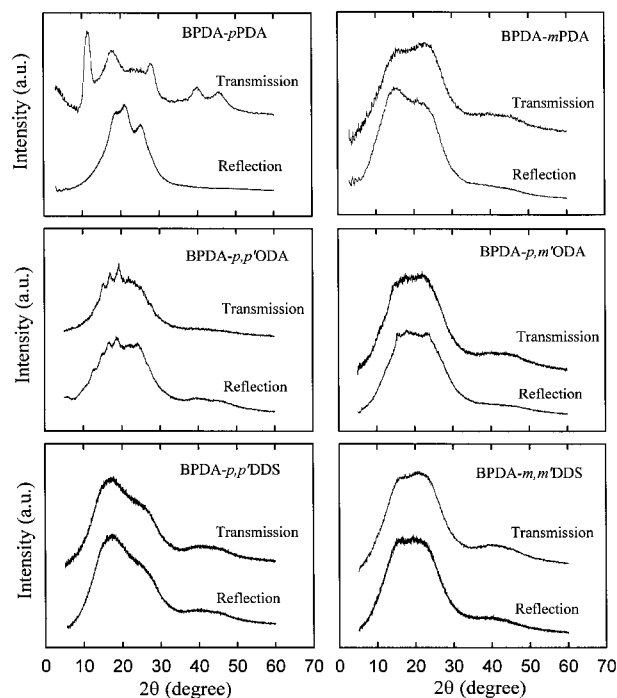


Figure 4 WAXD patterns of the isomeric BPDA-based polyimide thin films.

thin film have a relatively well-developed crystalline structure, whereas *meta*-oriented BPDA-*p,m'*ODA polyimide thin film does not. From WAXD patterns of the isomeric BPDA-PDA and BPDA-ODA polyimide thin films, it can be deduced that *para*-oriented polyimide thin films exhibited relatively more ordered and well-developed crystalline structure than the corresponding *meta*-oriented polyimide thin films. This may cause relatively lower diffusion coefficients and smaller water uptakes in *para*-oriented BPDA-PDA and BPDA-ODA polyimide thin films than the corresponding *meta*-oriented polyimide thin films.^{15–19}

For the isomeric BPDA-DDS polyimide thin films, there exists no significant difference in the transmission and reflection patterns unlike BPDA-PDA and BPDA-ODA isomeric polyimide thin films. The characteristic diffraction peaks of the transmission and reflection patterns indicate that both BPDA-*p,p'*DDS and BPDA-*m,m'*DDS polyimide thin films are nearly amorphous and structureless. These WAXD patterns of BPDA-DDS polyimide thin films showed good agreement with the water sorption behaviors. Comparing other polyimides, BPDA-DDS polyimide thin films having amorphous structure exhibited relatively high diffusion coefficient and water uptake.

Specifically, the amorphous structure in two isomeric BPDA-DDS polyimides may result from the bulky sulfonyl group, which inhibits close packing of polymer chains because of the steric hindrance.^{17,29,30}

From the present study, and that of other researchers,^{33,34} it can be deduced that the overall population of more ordered phase in the polyimide film is in the increasing order: BPDA-*m,m'*DDS \sim BPDA-*p,p'*DDS \sim BPDA-*m*PDA < BPDA-*p,m'*ODA < BPDA-*p,p'*ODA < BPDA-*p*PDA. The water sorption behaviors of the BPDA-based polyimide thin films do follow this order. The BPDA-based polyimide thin films having higher ordering in polymer chains showed relatively lower diffusion coefficient and smaller water uptake.

Therefore, it was concluded that the water sorption behaviors are affected by the morphological structure, in addition to the chemical structure. Specifically, the water sorption behaviors are significantly affected by the morphological structure, which includes packing order and crystallinity. In comparison with the polyimide film with *meta*-oriented linkages, those with *para*-oriented linkages showed relatively high crystalline and well-developed structure, and good intermolecular chain ordering, which may induce to lower diffusion coefficient and water uptake in the *para*-oriented polyimide thin films.

CONCLUSIONS

Water sorption behaviors of the BPDA-based polyimide thin films were gravimetrically investigated by using a thin film diffusion analyzer. The water sorption isotherms were reasonably well fitted by a Fickian diffusion model despite the morphological heterogeneities and the difference of chemical backbone structure. For the polyimide thin films, the water sorption behaviors are quite different and strongly dependent on the sort of polyimide. The diffusion coefficients vary in the range of 1.6×10^{-10} to 12.4×10^{-10} cm²/s and the water uptakes vary from 1.52 to 5.25 wt %. Both diffusion coefficient and water uptake in films are in the increasing order: BPDA-*p*PDA < BPDA-*p,p'*ODA < BPDA-*p,m'*ODA < BPDA-*m*PDA \sim BPDA-*p,p'*DDS < BPDA-*m,m'*DDS. Specifically, the polyimide thin films with *para*-oriented linkages showed relatively lower diffusion coefficient and smaller water uptake than the corresponding films with *meta*-oriented linkages be-

cause of the well-developed crystalline structure and good intermolecular chain ordering. The morphological structure is a more critical parameter than the chemical structure in controlling the water sorption behaviors of the polyimide thin films. The polyimide thin films having relatively higher chain order showed lower diffusion coefficient and water uptake.

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