

# Synthesis and Optical Properties of Polyimide-Silica Hybrid Thin Films

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Received March 18, 2002. Revised Manuscript Received July 29, 2002

In this study, two series of the polyimide–silica hybrid optical thin films, PMDA–ODA/SiO<sub>2</sub> and 6FDA–ODA/SiO<sub>2</sub>, were synthesized using an in situ sol–gel reaction combined with spin coating and multistep curing. The hybrid thin films were prepared by the aminoalkoxysilane-capped poly(amic acid)s and tetramethoxysilane (TMOS) as the precursors. Highly transparent hybrid thin films were obtained at a silica content as high as 54.9 wt %. The FE-SEM study suggests the nanosize domain of the silica moieties in the hybrid thin films. The AFM study demonstrates that the hybrid thin films have excellent planarity. The prepared hybrid thin films show good thermal stability. The refractive indices of the prepared hybrid thin films decrease linearly with increasing silica content while the Abbe number of the hybrid thin films shows the opposite trend. The birefringence of the prepared polyimide–silica hybrid thin films is reduced by incorporating the silica moiety. Optical planar waveguides were prepared from the prepared hybrid thin films using a thermal oxide as the cladding layer. The optical losses of these planar waveguides at 1310 nm are in the range of 0.5–1.9 dB/cm, which are due to the higher harmonics of the aromatic C–H vibration, the O–H vibration, and the extrinsic loss. The optical losses of the prepared polyimide–silica hybrid films are lower than their parent polyimides and thus have potential applications in optical devices.

## Introduction

Organic–inorganic hybrid materials have been widely studied recently because novel material properties can be produced through systematic design of the organic and inorganic segments.<sup>1</sup> Moreover, the hybrid materials offer the possibility of combining the advantages and overcoming the disadvantages of the two different materials. Controlling of morphology and functional segment size of the hybrid materials produces high performance films, coating, and membranes.<sup>2</sup>

Applications of hybrid materials in optoelectronic devices have been studied recently,<sup>3–11</sup> including wave-

guide materials,<sup>4,5</sup> photochromic materials,<sup>6</sup> conductive materials,<sup>7</sup> low-dielectric constant materials,<sup>8,9</sup> and high refractive index thin films.<sup>10,11</sup> Our laboratory has successfully prepared trialkoxysilane-capped poly(methyl methacrylate)(PMMA)–titania<sup>10,11</sup> and PMMA–silica<sup>12</sup> hybrid optical thin films. The prepared hybrid films showed high optical transparency, tunable refractive indices, and an absorption maximum. However, the organic moiety of PMMA possibly restricts their applications in high-temperature optoelectronic devices. Furthermore, the complete condensation of the inorganic segment in the hybrid materials requires a high curing temperature. Therefore, a high-temperature organic polymer in preparing hybrid materials for optoelectronic devices is essential.

Polyimides have been recognized as high-temperature polymers for optoelectronics due to their excellent thermal stability, chemical resistance, mechanical properties, and dielectric properties. Different polyimide–silica hybrids have been prepared,<sup>2,13–25</sup> which show

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outstanding thermal stability and mechanical strength. The transparency of the polyimide–silica hybrids prepared without a coupling agent was limited to the silica content <10 wt % in earlier studies.<sup>13,14</sup> Above this value, silica particles larger than 1  $\mu\text{m}$  were observed and the hybrid materials became opaque. To enhance the interaction between the organic moiety and the inorganic network, a coupling agent<sup>9,17–25</sup> was added to connect the polyimide and silica. Besides, low molecular weight poly(amic acid) was more suitable for preparing the hybrids since a high covalent bonding density between organic and inorganic moieties was obtained.<sup>20</sup> This improves the interaction between polyimides and silica, which decreases the size of silica particles and increases the silica content in the hybrid materials without phase separation. The low molecular weight poly(amic acid)s have also been utilized for preparing thin films with improved planarity.<sup>26</sup>

Although the structures and thermal and mechanical properties of polyimide–silica hybrid materials have been widely studied, the thin film properties of polyimide–silica hybrid materials have not been fully explored, especially the optical properties. In addition, application of polyimides in optical waveguides has developed recently, especially of fluorinated polyimides and their copolymers.<sup>27</sup> However, poor adhesion on some fluorinated polyimides restricts their applications. Therefore, hybrid materials based on fluorinated polyimides and silica may solve the above problem.

In this study, PMDA–ODA/SiO<sub>2</sub> and 6FDA–ODA/SiO<sub>2</sub> hybrid thin films were prepared using an in situ sol–gel reaction followed by spin coating and multistep curing, as shown in Figure 1. The dianhydrides and diamines for preparing polyimides were pyromellitic dianhydride (PMDA) and 4,4'-hexafluoroisopropylidene-diphthalic anhydride (6FDA), and oxydianiline (ODA), respectively. The coupling agent and tetraalkoxysilane were aminopropyltrimethoxysilane (APrTMOS) and tetramethoxysilane (TMOS), respectively. The stoichiometries used for preparing the hybrid thin films are listed in Table 1. The molecular structure, morphology, thermal properties, and optical properties of the prepared hybrid films were studied. Optical planar waveguides were fabricated from the prepared hybrid films on a silicon wafer with a thermal oxide and their properties measured at 1319 nm. The effect of the silica content on the optical properties of the prepared films and waveguides were discussed.

## Experimental Section

**Materials.** Pyromellitic dianhydride (PMDA, 99%, Acros), 4,4'-hexafluoroisopropylidenediphthalic anhydride (6FDA, 99%, Lancaster), and oxydianiline (ODA, 99%, TCI) were dried

under vacuum for 1 h at 50 °C before 3-aminopropyltrimethoxysilane (APrTMOS, 98+%, TCI), tetramethoxysilane (TMOS, 98+%, TCI), and *N,N*-dimethylacetamide (DMAc, 99.98%, Tedia) were used as received.

**Preparation.** *Preparing the PMDA–ODA/SiO<sub>2</sub> and 6FDA–ODA/SiO<sub>2</sub> Hybrid Thin Films.* The synthetic procedures for preparing hybrid thin films are described below. PMDA or 6FDA was added into a flask and dissolved in DMAc with stirring. After the dianhydride was dissolved in DMAc, ODA was added and reacted at room temperature under nitrogen. After 4 h, a 20 wt % yellow solution of a low molecular weight poly(amic acid) was obtained. The theoretical molecular weights of the prepared poly(amic acid)s are 1055 and 1733 for PMDA–ODA and 6FDA–ODA, respectively. Then, APrTMOS was added and reacted for another 2 h. The final molar ratio of dianhydride, diamine, and coupling agent was 3:2:2. Various molar ratios of TMOS and calculated quantities of deionized water were added as shown in Table 1. The molar ratio of H<sub>2</sub>O to the alkoxy function group of APrTMOS and TMOS was 1:1. After 16–28 h of reaction time (varied with the amount of inorganic precursor employed), the resulting homogeneous solution was then spin-coated on a 4-in. silicon wafer at 3000 rpm for 60 s. The coated film was then cured on a hot plate at 60 °C for 20 min, 100 °C for 20 min, 150 °C for 20 min, 300 °C in a furnace for 60 min under nitrogen, and finally at 400 °C in a vacuum furnace for 60 min. For prism coupler measurements on the polyimide–silica films, a 1.5- $\mu\text{m}$  thermal oxide layer was grown on the wafer surface before coating the reaction solution.

*Preparation of the PMDA–ODA and 6FDA–ODA Polyimide Thin Films.* The polyimides, PMDA–ODA and 6FDA–ODA, were prepared according to the literature<sup>28</sup> in comparison with the hybrid polyimide–silica thin films. For the preparation of PMDA–ODA and 6FDA–ODA, the ODA was added into a flask and dissolved in DMAc with stirring. After 20 min, PMDA or 6FDA was added and reacted at room temperature with stirring under nitrogen. The molar ratio of dianhydride to diamine was 1:1, and the total concentration of the reaction solution was 15 wt %. After 20 h, the reaction solution was spin-coated on a 4-in. silicon wafer at 3000 rpm for 60 s. The coated film was then cured on a hot plate at 60 °C for 20 min, 100 °C for 20 min, 150 °C for 20 min, and finally at 360 °C in a furnace for 120 min under nitrogen. For prism coupler measurements on the polyimide films, the procedure was similar to those of polyimide–silica materials except an adhesion promoter, 3-aminopropyltriethoxysilane (0.1 vol % in DI water), was coated on the thermal oxide first before coating poly(amic acid) solution. It is used to enhance the adhesion between the polyimide films and the substrate<sup>29</sup> because of worse adhesion of pure polyimide with a thermal oxide than polyimide–silica materials.

**Characterization.** Mid and near-infrared spectra of the prepared polyimide–silica hybrid thin films and polyimide thin films were recorded with a BIO-RAD Model QS-300 and Jasco Model V-570 spectrophotometer, respectively. The theoretical harmonics of C–H and O–H bond stretching vibrations can be calculated from the fundamental vibration and the anharmonicity constant:<sup>30</sup>

$$\nu_n = \frac{\nu_1 n - \nu_1 \chi n(n+1)}{1 - 2\chi} \quad (1)$$

where  $\nu_1$  is the fundamental vibration position (wavenumber),  $\nu_n$  is the  $n$ th overtone position, and  $\chi$  is the anharmonicity constant ( $\chi = 1.9 \times 10^{-2}$  for the C–H bond).

The microstructure of the hybrid materials was examined by a field emission scanning electron microscopy (FE-SEM, Hitachi, Model S-800). An atomic force microscope (AFM, Digital Instrument Inc., Model DI-5000) was used to probe the

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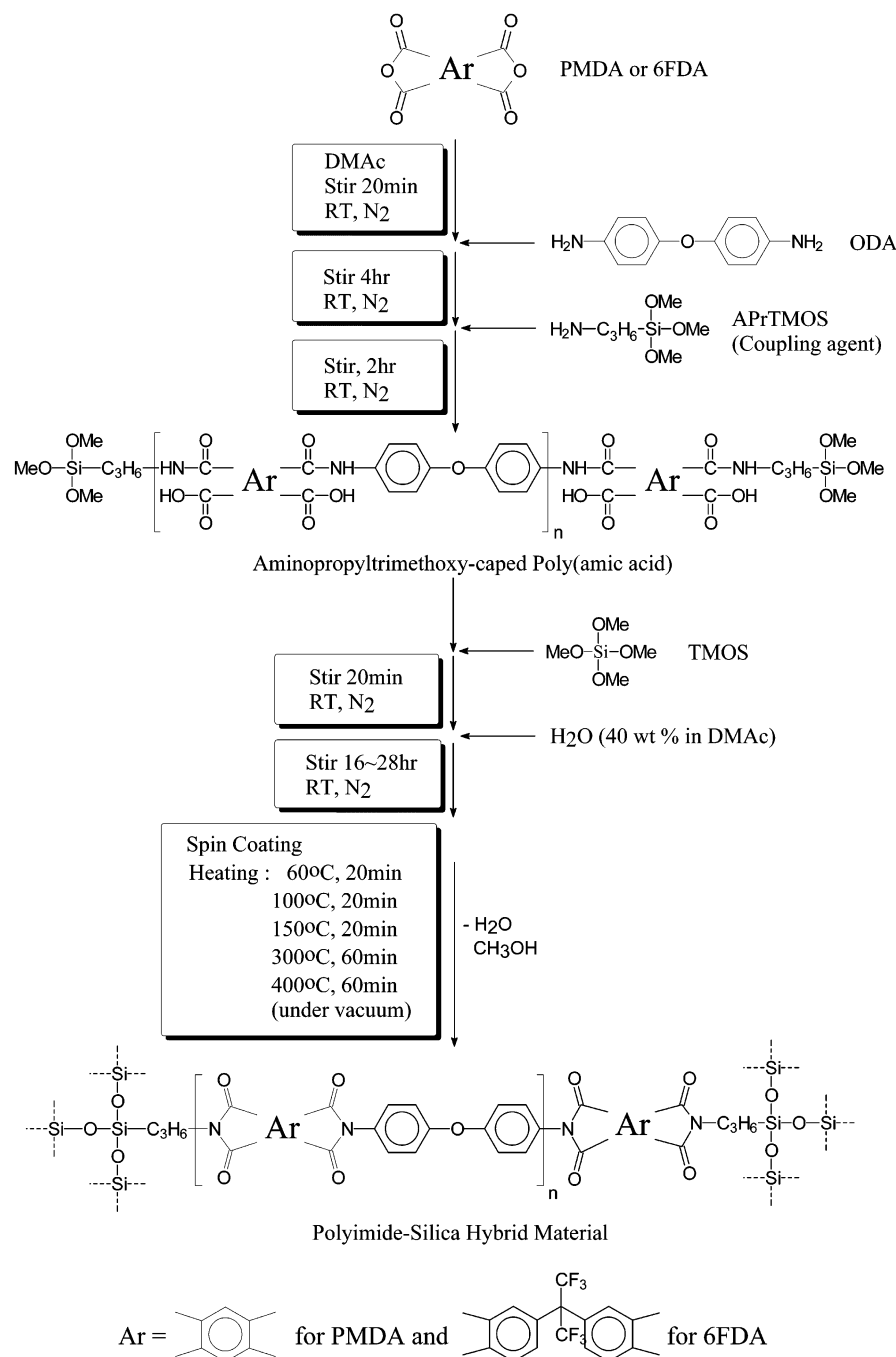
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**Figure 1.** Reaction scheme for preparing polyimide-silica hybrid thin films, **SP**, and **SF**.

surface morphology of the coated films. Thermogravimetric analysis (TGA) was performed under nitrogen flow using a Dupont Model 951 thermogravimetric analyzer at a heating rate of 10 °C/min. A n&k analyzer (n&k Technology, Inc., Model 1200) was used to measure the film thickness, refractive index, and the extinction coefficient of the prepared film in the wavelength range of 190–900 nm. The extinction coefficient has been normalized with film thickness. The Abbe numbers ( $\nu_D$ ) of the prepared polyimide and hybrid thin films were calculated according to the following equation:  $\nu_D = (n_D - 1)/(n_F - n_C)$ , where  $n_C$ ,  $n_D$ , and  $n_F$  are the refractive index at the spectral line of C (656 nm), D (589 nm), and F (486 nm), respectively.

The transverse electric (TE) and transverse magnetic (TM) refractive indices of the prepared films were measured by a prism coupler (Metricon Model 2010) at the wavelengths of 633 and 1319 nm. The birefringence ( $\Delta n$ ) defined as the difference between the in-the-plane and out-of-plane refractive indices (the difference of  $n_{TE}$  and  $n_{TM}$ ) were also calculated.

The optical losses of the planar waveguides at 1310 nm were measured by a cutback method.<sup>31</sup> The measurement system was designed by the Center for Measurement Standards, Industrial Technology Research Institute (Hsinchu, Taiwan).

## Results and Discussion

Figure 2 illustrates the FTIR spectra of the prepared polyimide and polyimide-silica hybrid thin films **SP0**, **SP1**, **SP6**, **SF0**, **SF1**, and **SF4**. The characteristic absorption bands of the imide group are observed at 727, 1378, 1728, and 1776  $\text{cm}^{-1}$  for all samples as shown in Figure 2. The major difference between the pure poly-

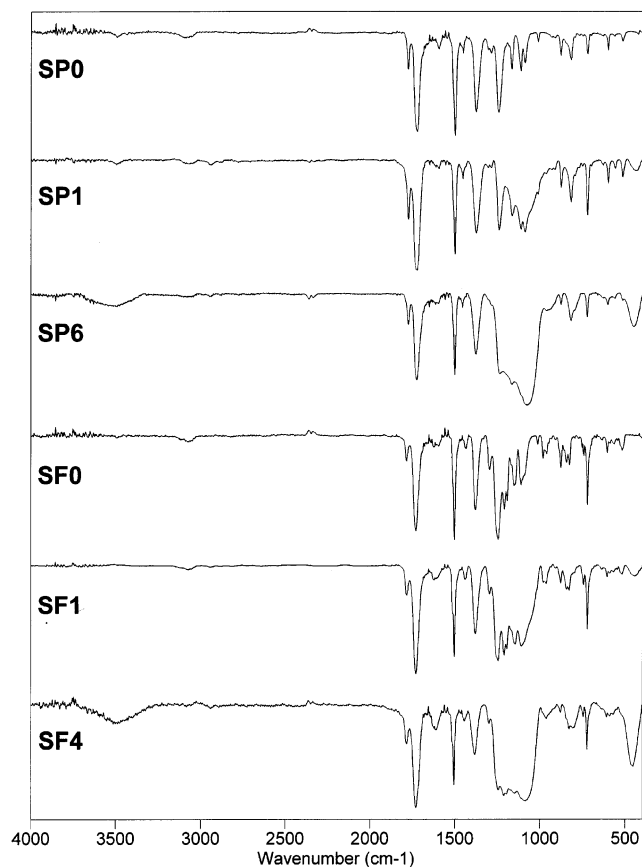
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**Table 1. Monomer Mixtures, Theoretically Compositions, and Solid Contents (wt %) for Preparing the Polyimide and Hybrid Thin Films<sup>a</sup>**

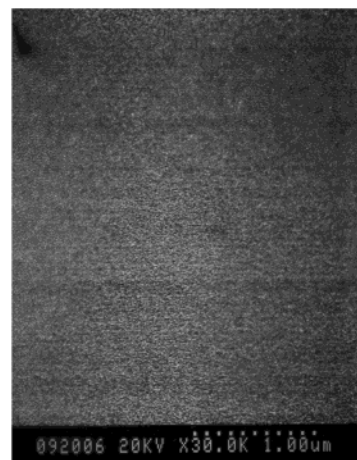
sample	solution (exp.)				solid content	film (calc.)	
	PMDA (SP) 6FDA (SF)	ODA	APrTMOS	TMOS		PI	SiO <sub>2</sub>
SP0	52.1	47.9	0	0	15.0	100	0
SP1	38.1	23.3	20.9	17.7	19.6	82.6	17.4
SP2	32.4	19.8	17.7	30.1	19.5	75.5	24.5
SP3	24.9	15.2	13.6	46.3	19.2	64.5	35.5
SP4	20.2	12.4	11.1	56.4	19.1	56.3	43.7
SP5	17.0	10.4	9.3	63.3	19.0	50.0	50.0
SP6	14.7	9.0	8.0	68.3	18.9	45.1	54.9
SF0	68.9	31.1	0	0	15.0	100	0
SF1	49.3	14.8	13.3	22.5	20.8	83.5	16.5
SF2	34.0	10.2	9.2	46.6	21.7	67.8	32.2
SF3	25.9	7.8	7.0	59.3	22.4	57.1	42.9
SF4	20.0	6.0	5.4	68.6	23.0	47.7	52.3

<sup>a</sup> PMDA(or 6FDA):ODA:APrTMOS = 3:2:2 (mole ratio).

**Figure 2.** FTIR spectra of prepared thin films **SP0**, **SP1**, **SP6**, **SF0**, **SF1**, and **SF4**.

imide and polyimide–silica films is the absorption band around 1000–1100 cm<sup>-1</sup>. This band gradually increases intensity with increasing silica content, consistent with the formation of the three-dimensional Si–O–Si network in the hybrid films.<sup>14,19</sup> The broad absorptions around 3200–3700 cm<sup>-1</sup> are assigned to the Si–OH residue, formed in the hydrolysis of alkoxy groups of APrTMOS or TMOS. This band is barely detectable in the spectrum of **SP1** or **SF1** with a lower silica content but increases its intensity in that of **SP6** or **SF4** with a high silica content. The Si–OH residue plays a significant role in the optical loss and will be discussed later.

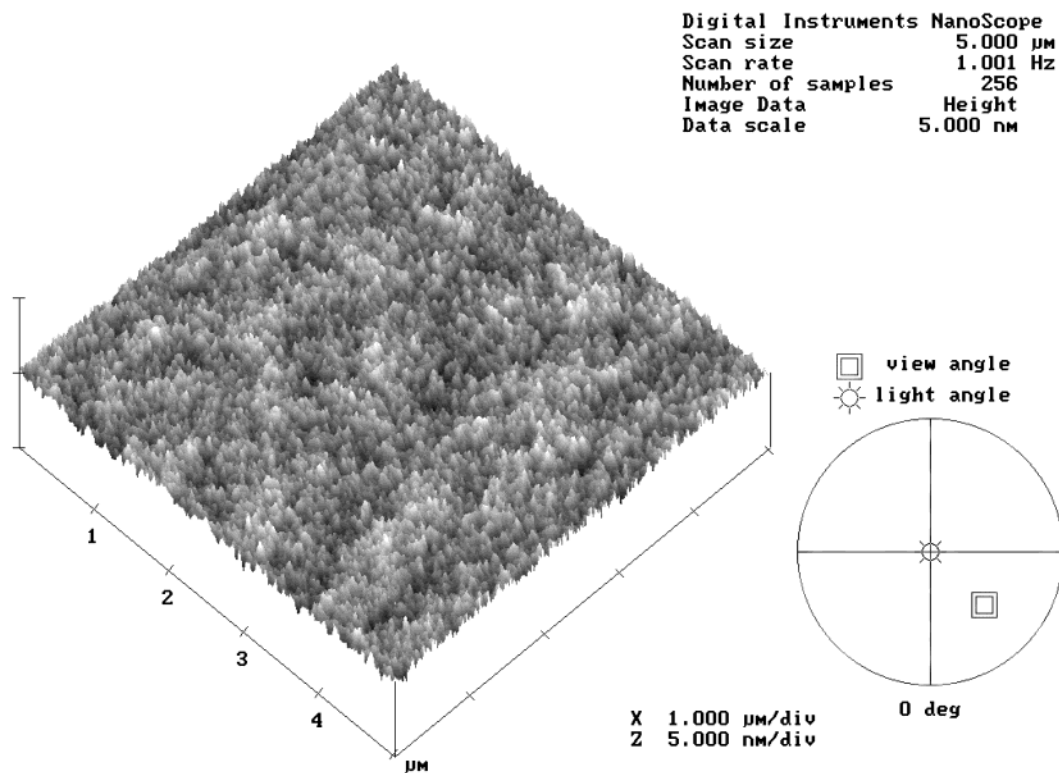
Figure 3 shows the FE-SEM micrograph of the prepared polyimide–silica hybrid thin film, **SF4**. There is no significant inorganic domain observed in the

**Figure 3.** FE-SEM diagram of hybrid thin film, **SF4**.

prepared hybrid films. The silica domains in the hybrid materials are estimated to be smaller than 20 nm from the figure. Figure 4 shows the AFM diagrams of the prepared hybrid thin film, **SF4**. The average roughness ( $R_a$ ) and mean square roughness ( $R_q$ ) of the prepared polyimide and hybrid films are listed in Table 2. The mean square roughness ranges from 0.26 to 0.34 for the **SP** system and from 0.28 to 0.30 nm for the **SF** hybrid thin films. The mean square roughness is less than that of the model polyimide thin films (0.63 for PMDA–ODA and 0.41 for 6FDA–ODA), which confirms the excellent planarity of the prepared hybrid thin films. Low roughness reduces the surface scattering loss at the waveguide surface<sup>31</sup> and good optical waveguides are expected from the prepared polyimide–silica hybrid thin films.

Figure 5 illustrates the TGA curves of the prepared polyimide and hybrid films **SF0**–**SF4** at a heating rate of 10 °C/min under nitrogen flow. The thermal decomposition temperatures ( $T_d$ ) of prepared films are shown in Table 2. They are all above 525 °C for both the **SP** and the **SF** hybrid thin films. From Figure 5, the increasing residues at 900 °C from **SF0** to **SF4** also suggest the successful incorporation of the silica moiety in the hybrid materials. The slightly lower decomposition temperatures relative to the pure polyimides are probably due to the low molecular weight of polyimide segments in the hybrid materials. Another possibility is the Si–OH residue groups in hybrid thin films since it increases with the silica content because of the incomplete condensation reaction.

The thickness ( $h$ ) of the prepared polyimide and hybrid thin films is in the range of 1.32–2.95 μm, as shown in Table 3. Figure 6a,b shows the refractive index ( $n$ ) dispersions of the prepared polyimide and hybrid films **SP0**–**SP6** and **SF0**–**SF4** in the wavelength range of 200–900 nm, respectively. The refractive indices of the prepared hybrid thin films at 633 nm are listed in Table 2. The refractive indices decrease from 1.74 to 1.53 and 1.59 to 1.50 for the **SP** and **SF** series, respectively. Increasing the incorporation of silica results in a decrease in the refractive index since the refractive index of silica is lower than that of polyimide. The lower refractive indices of the **SF** materials relative to those of the **SP** are due to the low electronic polarizability of the hexafluoroisopropyl group. From Table 2, the Abbe number increases from 14.6 of **SP0** to 22.0 of **SP6** and from 20.4 of **SF0** to 27.2 of **SF4** for prepared thin films,



**Figure 4.** AFM diagram of the hybrid thin film, SF4.

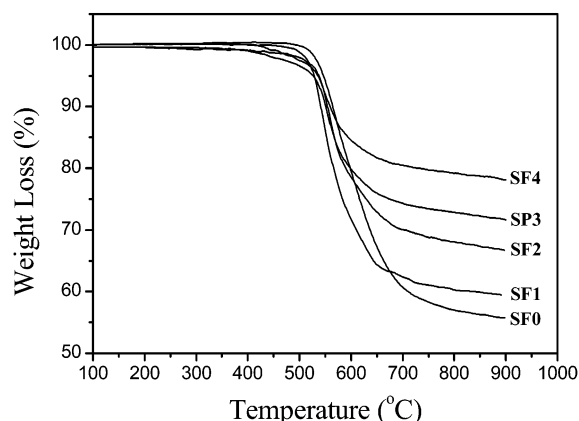
**Table 2. Properties of the Prepared Polyimide and Hybrid Thin Films**

sample	$R_a$ (nm)	$R_q$ (nm)	$T_d^a$ (°C)	$h^b$ (μm)	$n^b$	$\nu_D$	$\alpha^c$ (dB/cm)
SP0	0.49	0.63	591	1.32	1.74	14.6	2.9
SP1	0.26	0.33	551	2.29	1.66	16.5	1.1
SP2	0.27	0.34	551	2.91	1.62	17.9	1.9
SP3	0.22	0.28	548	1.49	1.61	19.7	1.3
SP4	0.20	0.26	531	2.95	1.58	20.5	0.9
SP5	0.21	0.28	548	2.29	1.56	21.1	1.3
SP6	0.21	0.28	542	1.92	1.53	22.0	0.9
SF0	0.33	0.41	548	1.78	1.59	20.4	1.7
SF1	0.22	0.28	529	1.98	1.57	20.2	0.5
SF2	0.23	0.30	538	2.58	1.54	22.7	0.8
SF3	0.24	0.30	526	2.78	1.52	24.5	1.0
SF4	0.24	0.30	537	2.73	1.50	27.2	1.2

<sup>a</sup> 5 wt % decomposition temperatures. <sup>b</sup> Measured at the wavelength of 633 nm using an n&k analyzer. <sup>c</sup> Measured at the wavelength of 1310 nm.

respectively. This suggests that the Abbe numbers of the hybrid thin films increase with increasing silica content. This indicates that the optical dispersion of the prepared thin films results from the polyimide moieties.

The transverse electric refractive indices ( $n_{TE}$ ) and transverse magnetic refractive indices ( $n_{TM}$ ) at 632.8 and 1319 nm of the prepared thin films were measured by a prism coupler, as shown in Table 3. The variations of the TE and TM mode refractive indices with the silica content for the hybrid thin films are shown in Figure 7a,b. The relationship between the measured refractive indices with the silica content and polyimide structure is the same as those measured by the n&k analyzer as described above. The birefringence corresponds to the optical anisotropy of the thin films.<sup>29</sup> The birefringence of the SP hybrid thin films is larger than the SF hybrid thin films because PMDA-ODA has a more planar rigid backbone than 6FDA-ODA that enhances the ordering along the substrate, increasing the anisotropy and



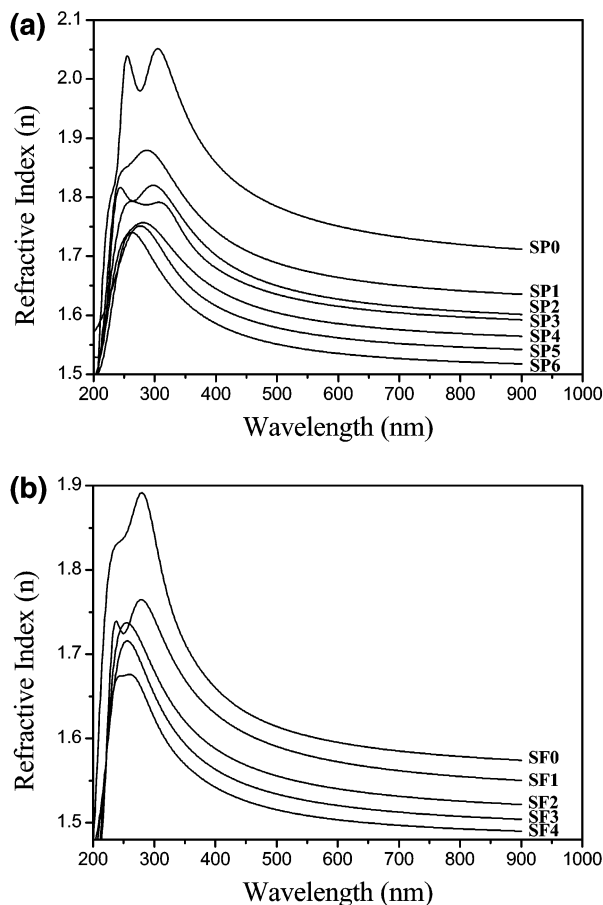
**Figure 5.** TGA curves of SF0-SF4 at a 10 °C/min heating rate under nitrogen flow.

**Table 3. TE and TM Mode Refractive Indices and Birefringence of the Prepared Thin Films**

sample	632.8 nm			1319 nm		
	$n_{TE}$	$n_{TM}$	$\Delta n^a$	$n_{TE}$	$n_{TM}$	$\Delta n^a$
SP0	1.718	1.637	0.081	1.679	1.607	0.072
SP1	1.641	1.593	0.048	1.611	1.568	0.042
SP2	1.626	1.585	0.041	1.597	1.560	0.037
SP3	1.599	1.566	0.033	1.573	1.543	0.029
SP4	1.584	1.553	0.031	1.560	1.532	0.028
SP5	1.564	1.541	0.023	1.540	1.521	0.019
SP6	1.552	1.533	0.019	1.530	1.513	0.017
SF0	1.592	1.584	0.008	1.567	1.560	0.007
SF1	1.559	1.551	0.009	1.538	1.530	0.008
SF2	1.538	1.531	0.007	1.518	1.512	0.006
SF3	1.526	1.518	0.008	1.507	1.500	0.007
SF4	1.511	1.503	0.008	1.493	1.486	0.007

<sup>a</sup> The birefringence,  $\Delta n = n_{TE} - n_{TM}$ .

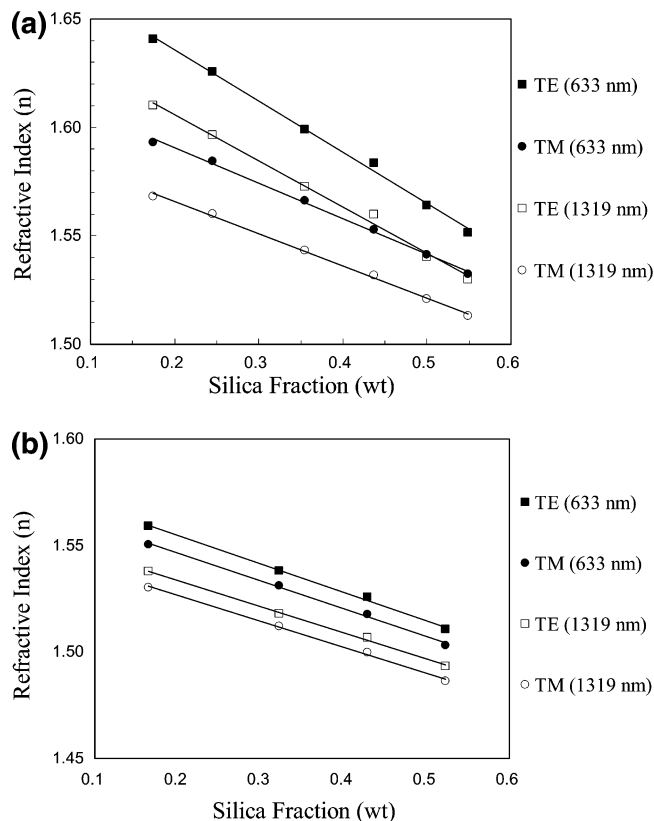
birefringence.<sup>29</sup> The birefringence of the SP hybrid thin films decreases with increasing silica content since silica is a low birefringence material. For the case of the SF



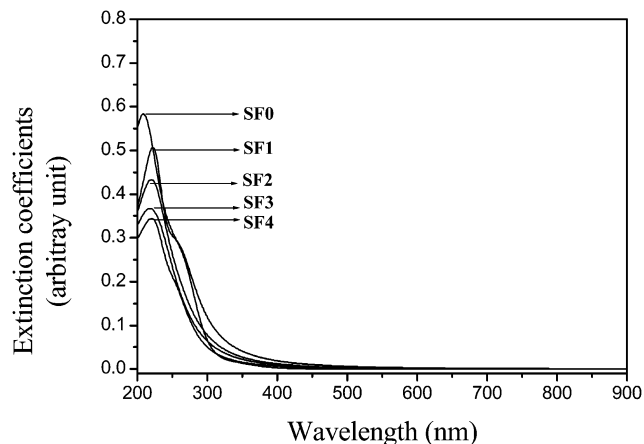
**Figure 6.** (a) Refractive indices ( $n$ ) of prepared thin films **SP0–SP6** in the wavelength range of 190–900 nm. (b) Refractive indices ( $n$ ) of prepared thin films **SF0–SF4** in the wavelength range of 190–900 nm.

hybrid thin films, the decreasing of birefringence with silica content is not as marked as the **SP** series since the birefringence of 6FDA–ODA is small.

Figure 8 shows the dispersion of the extinction coefficient of the studied films **SF0–SF4** in the wavelength range of 200–900 nm. The extinction coefficients are  $<0.02$  after the wavelength in the wavelength range of 400–900 nm for the studied hybrid thin films, which indicates excellent optical transparency in the visible region. The absorption bands in the UV region from Figure 8 are due to the chromophoric units and the charge transfer of polyimide structures.<sup>28</sup> Figure 9 showed the near-IR (NIR) spectra of prepared thin films **SP0**, **SP1**, **SP6**, **SF0**, **SF1**, and **SF4** in the wavelength range of 860–1800 nm. The theoretical and measured absorption peak sites are listed in Table 4. The calculated harmonic absorption peaks are very close to the measured absorption peaks in the prepared hybrid materials. There are three main peaks around 1660, 1412, and 1128 nm in PMDA–ODA and 6FDA–ODA polyimide thin films, which are assigned to peaks of **a**, **b**, and **c**, respectively. The peaks **a**, **b**, and **c** are assigned to the second harmonic of the aromatic C–H bond stretching vibration ( $2\nu_{C-H}$ ), a combination of the second harmonic of the aromatic C–H bond stretching vibration and the deformation vibration ( $2\nu_{C-H} + \delta_{C-H}$ ), and the third harmonic of the aromatic C–H bond stretching vibration ( $3\nu_{C-H}$ ), respectively.<sup>27</sup> In Figure 9, the peak, **d**, at 1458 nm is assigned to the second



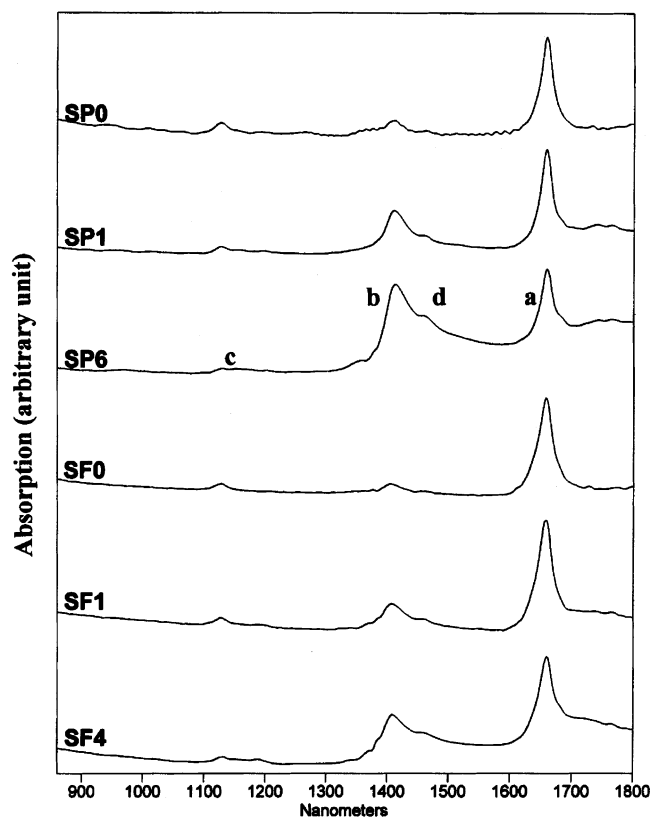
**Figure 7.** (a) TE and TM mode refractive indices with the silica content for the hybrid thin films **SP1–SP6** at 633 and 1319 nm. (b) TE and TM mode refractive indices with the silica content for the hybrid thin films **SF1–SF4** at 633 and 1319 nm.



**Figure 8.** Extinction coefficients ( $k$ ) of prepared thin films **SF0–SF4** in the wavelength range of 190–900 nm.

harmonic of the O–H bond stretching vibration ( $2\nu_{O-H}$ ), which increases with increasing silica content. The increasing absorption is due to the Si–OH residue groups. As discussed in Figure 2, the Si–OH residue increases with silica content due to the incomplete condensation. Although the O–H harmonic peak is not at the communication wavelengths, it will increase the absorption at the wavelengths of 1319 and 1550 nm and thus increase the absorption loss at these wavelengths.

The optical losses of the model polyimides and their hybrid thin films at 1310 nm were measured by a cutback method and are listed in Table 2. The measured optical losses of prepared polyimide planar waveguides



**Figure 9.** Near-IR spectra of prepared thin films **SP0**, **SP1**, **SP6**, **SF0**, **SF1**, and **SF4** in the wavelength range of 860–1800 nm.

in this study are 2.9 and 1.7 dB/cm for PMDA–ODA and 6FDA–ODA planar waveguides, respectively. The optical losses of the planar waveguides prepared from polyimide–silica hybrid thin films are in the range of 0.5–1.9 dB/cm, which are smaller than those of their corresponding polyimide planar waveguides. Introduction of the silica segment results in a decrease of the optical loss since the intrinsic optical loss is associated with the C–H overtone of the polyimide moiety. The optical losses of the prepared hybrid thin films do not have a clear correlation with the silica content in the **SP** hybrid thin films. It is because the loss of optical waveguide may result from both intrinsic absorption and extrinsic scattering losses. Besides the C–H over-

**Table 4.** Theoretical and Measured Absorption Peak Sites of the Prepared Thin Films

	C–H ( $3\nu$ )		C–H ( $2\nu + \delta$ )		O–H ( $2\nu$ )		C–H ( $2\nu$ )	
	exp. (nm)	calc. <sup>a</sup> (nm)	exp. (nm)	calc. (nm)	exp. (nm)	calc. <sup>a</sup> (nm)	exp. (nm)	calc. <sup>a</sup> (nm)
<b>SP0</b>	1128	1129	1412	1400	1464	1458	1660	1660
<b>SP1</b>	1128	1129	1412	1400	1462	1458	1660	1660
<b>SP6</b>	1130	1129	1414	1400	1458	1460	1660	1660
<b>SF0</b>	1128	1129	1406	1400	1460	1458	1658	1658
<b>SF1</b>	1128	1129	1408	1400	1456	1458	1660	1658
<b>SF4</b>	1130	1129	1408	1400	1454	1458	1660	1658

<sup>a</sup> Calculated by eq 1.

tone loss, the Si–OH residue, the core (polyimide–silica)-cladding (thermal oxide) boundary imperfection, and film quality may result in additional optical losses. Since the prepared polyimide–silica hybrid films have a lower optical loss than their parent polyimides, they might be able to replace the polyimides used in optical waveguide applications.

### Conclusions

In this study, polyimide–silica hybrid optical thin films were successfully prepared using an in situ sol–gel process combined with spin coating and multistep baking. The prepared hybrid thin films have a homogeneous structure and nanoscale size domain of the silica moieties. Excellent surface planarity and optical transparency are obtained at a high silica content. The refractive indices of the prepared polyimide–silica hybrid thin films decrease linearly with increasing silica content while the Abbe numbers show the opposite trend. The birefringence of the prepared polyimide–silica hybrid thin films is reduced by incorporating the silica moieties. The optical losses of the polyimide–silica hybrid films are lower than their parent polyimides. The excellent film uniformity, optical transparency, and tunable optical properties provide the potential applications of the polyimide–silica hybrid thin films in optical devices.

**Acknowledgment.** We thank the National Science Council of Taiwan for the financial support of this work (NSC89-2216-E002-030 and NDL 90-C036).

CM0202310