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## Photoinduced Refractive Index Modulation of Photosensitive Polyimides and Their Application in Passive Waveguide Fabrication

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A novel photosensitive poly(amide-co-imide) was synthesized from the copolymerization of p-phenylenediacryloyl chloride, bis(3,4-dicarboxylphenyl)hexafluoropropane dianhydride and 4,4'-oxydiphenyldiamine followed by subsequent chemical imidization. The resulting polymer showed intrinsic photosensitivity without additives, good processability in solution and high thermal stability up to 350 °C. From the refractive index measurement of the polymer by prism coupling method, it was found that the refractive index could be varied precisely with respect to the exposure time (from 1.6444 to 1.6230 for 2 µm thick film, TE mode, measured at 1.55 µm). Fabrication of passive waveguide was attempted by the photo-patterning process, i.e., photo-modulation of refractive index. Straight waveguide was easily fabricated and single mode near-field pattern was observed by using 1.55 µm near-IR light.

**Keywords** photosensitive polyimide, photo-induced refractive index modulation, polymer waveguide, single mode near-field pattern

### INTRODUCTION

Polymers are one of the most promising candidates for the optical materials to fabricate waveguide due to the several advantages over the inorganic materials such as SiO<sub>2</sub>, LiNbO<sub>3</sub>, and GaAs. They can be spin-coated on almost any substrates and fabricated easily and inexpensively, and the polymer waveguides show faster response time and wider bandwidths. Optical waveguides have been produced from several polymers such as polyacrylates, polystyrene, poly(organo-

silane), benzocyclobutene and so on.<sup>[1]</sup> More recently, fluorinated polyimides have been used mainly due to their high thermal stability as well as other desirable properties.<sup>[2-4]</sup> Reactive ion etching (RIE) is usually employed to fabricate optical waveguides from such polyimides because of its precise patterning characteristics. However, there are several disadvantages in RIE, such as restriction of the processed area for less than 5  $\mu\text{m}$ , the multiple and complicated processes, and longer fabrication time. To solve this problem, numerous attempts have been made. For example, electron-beam irradiation technique was reported to modulate the refractive index by bleaching the polyimide structure, and the authors successfully fabricated a channel-waveguide using electron-beam irradiation.<sup>[5]</sup> Nevertheless, complicated and expensive equipment was required and the resulting waveguide showed considerable transmission loss due to the damages in the polyimide induced by electron-beam bombardment. Recently, we have reported syntheses and characterization of a new photosensitive poly(amide-co-imide) based on phenylenediacryloyl moiety, which showed good photo-sensitivity at elevated temperature and became insoluble with irradiation of 365 nm UV light.<sup>[6]</sup> In this paper, we report the photo-induced refractive index (RI) change of such polyimide, and its application in passive waveguide fabrication.

## EXPERIMENTAL

**Syntheses of poly(amide-co-imide):** The poly(amide-co-imide), 6FOD, was synthesized from 0.4 mole of phenylenediacryloyl chloride (PDACl) and 0.6 mole of bis(3,4-dicarboxylphenyl)hexafluoropropane dianhydride (6FDA), and 1 mole of 4,4'-oxydiphenyldiamine (ODA) as reported in a previous paper.<sup>[6]</sup> In this experiment, 6FOD-46 ( $x = 0.4$  in Figure 1) was used for RI measurements and waveguide fabrications.

**Refractive Index (RI) Measurement:** Polymer thin film (2 ~ 4  $\mu\text{m}$  thick) was produced by spin coating a filtered (0.4  $\mu\text{m}$  Millipore) NMP solution (14.5 wt %) of 6FOD-46 at 700 rpm on a Si wafer and drying in a vacuum oven for 14 h at 60 °C. RI was measured by prism coupling method by using 1.55  $\mu\text{m}$  light source while the film was irradiated with 500W tungsten lamp at 150 °C.

**Fabrication of Straight Waveguide:** Straight waveguide was fabricated by using the photo-induced RI patterning technique. Firstly, 20  $\mu\text{m}$  of Cyclotene<sup>®</sup> lower cladding was produced on a Si wafer as described in the literature.<sup>[7]</sup> Then, 6FOD-46 was spin-coated and dried in vacuum to produce 2 ~ 4  $\mu\text{m}$  film. UV was irradiated by 500 W tungsten lamp at 150 °C for 250 min all over the area, first, and then for 120 min the irradiation was continued over the area except the core (by using 3  $\mu\text{m}$  x 50 mm mask pattern). Finally, 10  $\mu\text{m}$  of Cyclotene<sup>®</sup> upper cladding was produced over the 6FOD layer. The resulting waveguide was examined by observing a near field pattern with a 1.55  $\mu\text{m}$  light source and the IR camera.

## RESULT AND DISCUSSIONS

The synthesis of 6FOD-46 was shown schematically in Figure 1. The polymer structure was identified by  $^1\text{H-NMR}$ , IR, and UV spectra as described in the previous paper.<sup>[6]</sup> The resulting polymer was soluble in aprotic polar solvents and stable up to 350 °C. The polymer underwent crosslinking without sensitizer upon irradiation of 365 nm via [2+2] cycloaddition of double bonds. From the gel fraction measurement, the sensitivity was found to be 990 mJ/cm<sup>2</sup> at room temperature, and 343 mJ/cm<sup>2</sup> at 160 °C.

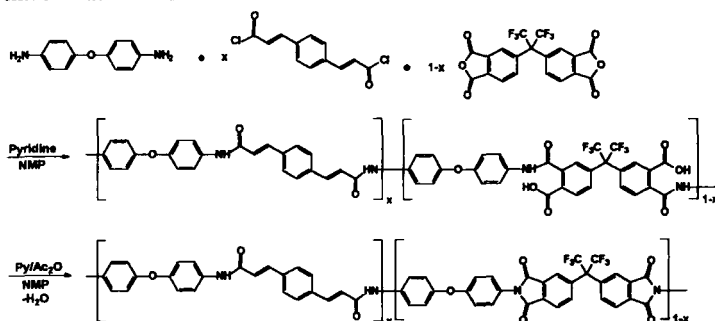


FIGURE 1. Synthetic schemes of poly(amide-co-imide), 6FOD

The RI of the irradiated polymer film was measured by prism coupling method. As shown in Figure 2, the RI of TE mode (in-plane polarized) was decreased with respect to the irradiation time (from 1.6444 to 1.6230 for 2  $\mu\text{m}$  thick film), while that of TM mode (out-of-plane polarized) was almost not changed or slightly increased. This is attributed to the photo-induced molecular conformational change during the [2+2] cycloaddition. Intermolecular distances between the molecules of out-of-plane slightly decrease, while those of in-plane

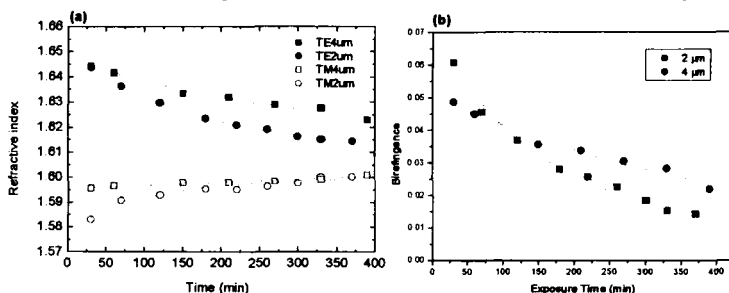


FIGURE 2. (a) Refractive index and (b) optical birefringence change of 6FOD-46 with respect to the irradiation time (500W tungsten lamp at 150 °C)

molecules increase owing to the  $sp^2 \rightarrow sp^3$  conformational change. As a result, the optical birefringence ( $\Delta n$ ) was decreased (from 0.0456 to 0.0144, for  $2\mu\text{m}$  thick film) with the increased irradiation time. It is notable because, in the case of electron-beam photo-modulation for fluorinated polyimides,<sup>[5]</sup> the RI of polyimide was increased with respect to the irradiation time due to the fluorine abstraction induced by electron-beam bombardment. Fabrication of a straight waveguide was attempted by using this RI photo-modulation technique. Straight waveguide was fabricated by irradiating the film with a mask pattern of  $3\mu\text{m} \times 5\text{mm}$  line, which resulted in the RI of unexposed core slightly higher (c.a. 0.005) than that of the exposed cladding area. Single mode near-field pattern was observed by using  $1.55\mu\text{m}$  near-IR light as in Figure 3. The elliptic shape was probably attributed to the RI mismatching between core and cladding and the un-optimized design. The propagation loss was estimated roughly to be larger than 1.0 dB, which is mainly due to the absorption of N-H vibrational overtone near at  $1.55\mu\text{m}$ . Further works are under progress including the optimization of waveguide design and structural modification of the polymer. The details of experiments and results will be published elsewhere.

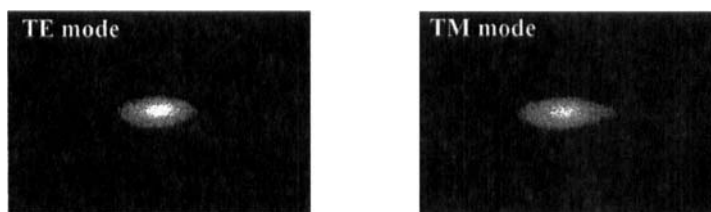


FIGURE 3. Near field mode patterns observed by IR camera at  $1.55\mu\text{m}$  for the polymer waveguide fabricated by the photo-defining technique.

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