Organic Electro-Optic Waveguide Modulators And Switches

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ABSTRACT

Organic and polymeric materials exhibit many important properties as electro-optic materials. Poled polymer films are particularly well suited for integrated optic devices, due to their large electro-optic coefficients, low dielectric constants, and flexible processability. We review our work toward the fabrication of active polymer integrated optic devices based on a newly developed channel waveguide fabrication procedure, and detail some of the properties of the devices.

1. INTRODUCTION

Organic and polymeric materials have emerged in recent years as promising candidates for advanced device and system applications. This interest has arisen from the promise of extraordinary optical, structural, and mechanical properties of certain organic materials, and from the fundamental success of molecular design performed to create new kinds of materials. From an optical standpoint, organics offer temporal responses ranging over fifteen orders of magnitude, including large nonresonant electronic nonlinearities (fsec-psec), thermal and motional nonlinearities (nsec-msec), configurational and orientational nonlinearities (msec-sec), and photochemical nonlinearities (psec-sec). Additionally, organic and polymeric materials can exhibit high optical damage thresholds, broad transparency ranges, and can be polished or formed to high-optical quality surfaces. Structurally, materials can be made as thin or thick films, bulk crystals, or liquid and solid solutions, and can be formed into layered film structures, with molecular engineering providing different optical properties from layer to layer. Mechanically, the materials can be strong and resistant to radiation, shock, and heat. When coupled with low refractive indices and low D.C. and microwave dielectric constants, the collective properties of these extraordinary materials show great promise towards improving the performance of existing electro-optic and nonlinear optical devices, as well as allowing new kinds of device architectures to be envisioned.

This paper reviews our current progress toward fabricating electro-optic, organic integrated optical devices. Section 2 provides a brief review of the useful properties of glassy, electro-optic polymer materials for integrated optics. Section 3 reviews our methods for fabricating and poling slab waveguide modulators for the characterization of the glassy polymer films and the demonstration of new fabrication techniques. In section 4, we briefly describe a new method, called the selective poling procedure (SPP), for producing active, buried-channel waveguide devices. The SPP is demonstrated by the fabrication of several integrated optical devices, including a Y-branch, Mach-Zehnder interferometer-modulator, a directional coupler switch, and a GHz traveling-wave phase modulator. We conclude with a summary of our work and point to new directions for future research.

2. ORGANIC MATERIALS IN INTEGRATED OPTICS

The synthesis of glassy polymer films containing molecular units with large nonlinear polarizabilities has led to the rapid implementation of organic integrated optics. These films, either spun, cast, or dipped, are amorphous as produced, and can be processed to achieve a macroscopic alignment for the generation of second-order nonlinear optical effects by electric-field poling. Typically, the films are poled by forming an electrode-polymer-electrode sandwich, and applying an electric field normal to the film surface. This produces films with their nonlinear molecular units oriented on the average normal to the film. In this state, the electric field of an optical beam propagating through the film can be maximally modified when the field is parallel to the oriented molecular units, that is, when the propagation vector lies in the film plane. As such, the films are ideally suited for guided wave applications. The successful development of guest-host and side-chain polymer systems incorporating molecular units with large nonlinear polarizabilities in this manner has thus led to the availability of organic thin film materials for integrated optics.

Organic electro-optic materials offer a variety of potential advantages over conventional materials for integrated optical device applications. Table 1 provides a comparison of the potential of organic materials with the current commercial technology, Ti-indiffused LiNbO3 in three major areas of importance: materials parameters, processing technology, and fabrication technology.
Some of these advantages have already been realized in our work on E-O modulators using poled polymer films, described below.

<table>
<thead>
<tr>
<th>CURRENT TECHNOLOGY: Ti:LiNbO$_3$</th>
<th>PROPERTIES OF POLYMERIC ORGANIC E-O MATERIALS</th>
</tr>
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<tbody>
<tr>
<td>$r = 32$ pm/V</td>
<td>$r = 14-53$ pm/V (poled films [1])</td>
</tr>
<tr>
<td>- Larger Modulating Voltage</td>
<td>- Lower Modulating Voltage</td>
</tr>
<tr>
<td>- Little Improvement Expected</td>
<td>- Potentially Much Larger $r$</td>
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<tr>
<th>LIMTED FABRICABILITY</th>
<th>FLEXIBLE FABRICATION</th>
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<tbody>
<tr>
<td>- 1000°C Processing</td>
<td>- Low Temperature Processing</td>
</tr>
<tr>
<td>- Depth Limited to 5 $\mu$m</td>
<td>- Flexible Dimensions</td>
</tr>
<tr>
<td>- Low Index Change $\Delta n$</td>
<td>- Controllable Index Change $\Delta n$</td>
</tr>
<tr>
<td>- Loss $&gt; 0.1$ dB/cm</td>
<td>- Loss $&lt; 0.8$ dB/cm [2]</td>
</tr>
<tr>
<td>- Optical Damage (Photorefractor)</td>
<td>- High Optical Damage Threshold</td>
</tr>
</tbody>
</table>

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<thead>
<tr>
<th>LARGE DIELECTRIC CONSTANT (28)</th>
<th>LOW DIELECTRIC CONSTANT (4)</th>
</tr>
</thead>
<tbody>
<tr>
<td>- Longer Time Constants $= RC$</td>
<td>- Shorter Time Constants $= RC$</td>
</tr>
<tr>
<td>- Large Velocity Mismatch in Traveling Wave Modulator</td>
<td>- Smaller Velocity Mismatch</td>
</tr>
</tbody>
</table>

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<tr>
<th>MASS PRODUCTION DIFFICULT</th>
<th>POTENTIAL FOR MASS PRODUCTION</th>
</tr>
</thead>
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Table 1. Comparison of Integrated Optics Technologies: Current Ti-LiNbO$_3$ and Projected Organics Technologies. [1]-data reported at the Materials Research Society, Boston, MA, Dec., 1987; [2]-data reported in this paper

The major advantages are due to the intrinsic differences in E-O mechanisms in inorganic and organic materials. Organics should provide flat E-O response to well beyond a GHz, and, indeed, measurements of the E-O coefficients and SHG coefficients of certain poled polymer films show little or no dispersion. Additionally, the E-O coefficients of poled polymer films can be made nearly as large as LiNbO$_3$. Pure MNA crystals already exhibit larger E-O coefficients than LiNbO$_3$, but it is our sense that practical, organic integrated optical devices will be made with films, not crystals. It is also true that the dielectric constant of poled polymer films is substantially lower than that of LiNbO$_3$, implying smaller RC time constants and wider frequency bandwidths. In this connection, the bandwidth-length product for a typical poled polymer is of order 120 GHz-cm, compared to 10 GHz-cm for LiNbO$_3$. Finally, the processing technology for integrated optical devices based on poled polymer films is relatively straightforward and fast, requiring only moderate temperatures (100-200 degrees C) for poling, and standard semiconductor fabrication equipment for fabrication of layered waveguide structures.

There are some potential drawbacks to polymers for integrated optics, as well. The microwave loss tangents in glassy poled polymers are not yet known, but are expected to be small. However, the materials may have low thermal conductivity, which could lead to power dissipation problems in practical devices, as well as produce instabilities of the poled states. Further, the stability of poled states at nominal temperatures is not yet well known. These issues represent problems to be solved for practical implementation of organic electro-optic materials.

In light of the potential benefits of poled polymer films for integrated optical devices discussed above, many research groups in the United States, Europe, and Japan have embarked upon dedicated materials synthesis and device fabrication programs to bring this field to fruition in commercial and military products. In the remainder of the paper, we review our progress toward...
developing an understanding of the use of the materials in device architectures, and toward achieving good levels of performance in prototype devices based upon poled polymer films.

3. SLAB WAVEGUIDE DEVICES AND MATERIALS PROPERTIES

The fabrication of simple slab guided wave structures from polymer films permits the systematic study of fabrication techniques for organic devices, as well as providing structures for making direct measurement of Kerr and electro-optic (E-O) coefficients in the waveguide configuration. Figure 1 illustrates a typical slab waveguide device and some of the components used as substrates, electrodes, buffer layers, and the polable, glassy polymers. The polymers used were supplied by the Hoechst Celanese Corporation, and include: 1) guest-host MNA/PMMA films, both poled and unpoled, 2) PC6S, a yellow, pendant side-chain polymer, 3) C-22, a red, pendant side-chain polymer, and 4) HCC-1237, a more active version of C-22. Waveguide structures such as that in the figure were built up by spin-coating the various layers to 2-3 μm thicknesses. The polymers were then poled to produce a non-centrosymmetric structure exhibiting a nonzero electro-optic effect.

![Typical Slab Waveguide Modulator and Components](image)

The poling procedure typically consists of first spin-coating the electrode-coated substrate with the bottom buffer layer and then the active polymer, applying a top electrode, heating the structure above the glass transition temperature of the active polymer, applying a voltage of order 1 MV/cm to the structure, and cooling the sample with the voltage applied. The top electrode is then removed, and a top buffer layer and new electrode can be applied to the poled structure. Guiding of 830 nm light from a semiconductor laser over a 1-3 cm dimension with minimal loss is then achieved by locating the prism couplers directly over the ends of the poled region.

Initial work was performed with PC6S. The optical response was measured by placing the slab modulator in one arm of an external interferometer. The device half-wave voltage was about 48 volts. Frequency response was flat out to 400 kHz. The electro-optic coefficient of the poled PC6S film was measured to be 2.8 pm/V. Similar slab modulators were fabricated with the C-22, although different buffer layers were used with the C-22 devices. The poling fields were comparable to those for PC6S, but the C-22 glass transition temperature was slightly higher. We observed flat E-O modulation out to 85 MHz with the C-22 modulator, and measured r33=16±2 pm/V, about half that of LiNbO3.

Table 2 summarizes the measured physical parameters for PC6S and C-22 slab waveguides. It is most significant that the linear losses in the C-22 guides are below a dB/cm. This allows path lengths of order several cm, and, consequently, half-wave voltages approaching TTL levels. Both materials exhibit even better performance at a wavelength of 1.3 μm. The slab waveguide modulators thus provide a simple and effective method for characterizing the bulk polymer film properties and for developing device fabrication procedures.
Table 2. Physical Parameters Measured for PC6S and C-22 Slab Waveguides

<table>
<thead>
<tr>
<th>Parameter</th>
<th>PC6S</th>
<th>C-22</th>
</tr>
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<tbody>
<tr>
<td>E-O COEFFICIENT (pm/V)</td>
<td>2.8</td>
<td>16.0</td>
</tr>
<tr>
<td>TM REFRACTIVE INDEX (POLED)</td>
<td>1.7</td>
<td>1.58</td>
</tr>
<tr>
<td>TM INDEX DIFFERENCE (POLED-UNPOLED)</td>
<td>0.06</td>
<td>0.005</td>
</tr>
<tr>
<td>WG LENGTH (cm)</td>
<td>1.8</td>
<td>2.5</td>
</tr>
<tr>
<td>MEASURED LOSS (dB/cm)</td>
<td>2.3</td>
<td>0.8</td>
</tr>
<tr>
<td>HALF-WAVE VOLTAGE (volts)</td>
<td>48</td>
<td>7</td>
</tr>
</tbody>
</table>

4. CHANNEL WAVEGUIDE DEVICES

Polymer slab waveguides can be made by spinning thin films onto high quality optical substrates. As spun, the films are isotropic and exhibit no second-order nonlinear susceptibility. In addition to inducing a non-centrosymmetric structure to achieve a macroscopic electro-optic effect, a second major transformation must be engineered in the material to enable the fabrication of integrated optic circuits. Channel waveguides must be formed to confine and guide the light from one active element of the integrated optic circuit to another. The formation of channel waveguides and the poling of the material to produce an active, E-O channel must both be accomplished for device prototypes, and would usually be performed in two distinct steps: fabrication and poling. We have developed a new, powerful method, called the selective poling procedure (SPP), by which active, poled channel waveguides can be fabricated in a single fabrication step.

4.1 Selective Poling Procedure

Figure 2 illustrates the SPP for fabricating active polymer channel waveguides. An electrode pattern defining the channel waveguides is first deposited onto a substrate using standard photolithographic techniques. A planar buffer layer is then applied to optically isolate the active waveguide layer from the metal electrodes. The buffer material must be chosen to have an index lower than the guiding layer and to be compatible with the required processing. Thus, different buffer layers must often be used with different nonlinear polymers. A planar electrode is evaporated directly onto the nonlinear polymer for poling. The nonlinear layer is then poled by applying an electric field above the polymer glass transition and cooling the sample to room temperature under the influence of the field. The degree of alignment induced and the resultant electro-optic coefficient can be calculated based on a statistical average of the molecular susceptibilities. In this case only those regions of the material defined by the electrode pattern on the substrate are poled.

Since most organic nonlinear optical molecules also possess an anisotropic microscopic linear polarizability, the poled region becomes birefringent. The poled regions are uniaxial, with $n_e$ oriented along the direction of the poling field. Consequently, TM and TE waves propagating in vertical and transverse device structures respectively will experience a greater refractive index in the poled regions than in the unpoled regions, and so can be confined in the lateral dimension. Thus, by applying the poling fields using electrodes patterned to define the waveguide network, including both active and passive sections, no further patterning of the active organic layer is required to form the channel E-O waveguide structures. The devices are then completed by etching off the planar poling electrode, applying an upper buffer layer and depositing the patterned switching electrodes, as shown in Figure 2. If electrode removal is not possible, the device may be finished first, and then be selectively poled through the buffer layer.
The SPP permits both vertical and transverse poling of strips. The vertical method, illustrated in Figure 2, locates the electrodes directly over the guides, implying that parallel channels can be located closer together without having an electrode in the middle of the channels. Vertically poled channels should guide TM radiation, while transverse poled channels should guide TE radiation.

Using the SPP, channel waveguides were constructed with both the PC6S and the C-22 materials, and light guiding was observed in both cases. The index change was measured by determining the prism coupling angles in poled and unpoled guides and by measuring the phase change from a double pass reflection through poled and unpoled regions. PC6S has a large index change of $\Delta n = 0.06$ for TM waves, while the C-22 material exhibited $\Delta n = 0.005$ for TM waves. These index differences can be fine tuned by adjusting the poling field or by alterations in the nonlinear optical material. Figure 3 illustrates the confinement of 830 nm light in a poled PC6S channel, as compared with the unconfined light in a PC6S slab. Figure 4 illustrates a simple experimental method to examine the extent of the induced birefringence outside of the electrode region. No such birefringence was observed, indicating that the fringing fields do not cause the guiding region to spread beyond the strip electrodes.
In addition to the obvious simplification of the device fabrication process, the SPP has several other advantages over other methods of producing channel waveguides in poled polymer films. Fringing of the poling fields in the buffer and E-O layers acts to smooth out the edges of the guiding regions. If the edge roughness of the electrodes is small compared to the buffer layer thickness (of order 2 μm), the roughness of the waveguide boundaries should be independent of the resolution of the photolithographic process used, and should result in lower scattering losses in the waveguides. This has been confirmed by us in PC6S channel guides. Figure 5 displays the loss measurements in both a slab and channel guide. The losses are equal, within experimental error. Scattering losses from surface roughness are a major problem in channel guides defined by etching the nonlinear optical material or by channel filling in the substrate. Another advantage of the SPP is that the waveguides defined by the poling process have significantly different index changes for the TE and TM waves, making possible polarization selective elements.
Potential drawbacks to the SPP include partial lateral confinement of TE waves near the guide boundaries due to nonuniformly poled polymer and the fact that the guide cladding is intrinsically the unpoled polymer and cannot be arbitrarily chosen for a particular application.

In order to model the performance that can be expected for integrated optic devices, we developed a wave propagation design tool. Figure 6 shows the results of a model of the input of a Y-branch interferometer. The normalized guide width is $5 \lambda / n_{\text{eff}}$ and an induced index difference of $\Delta n = 0.005$ is assumed, corresponding to the C-22 polymer. Clear confinement in the lateral dimension is predicted for a C-22 Y-branch fabricated by the SPP (Fig. 6a), while no confinement is predicted for an unpoled guide (Fig. 6b).

![Figure 5. Loss Measurements in PC6S Slab Guides (circles) and 150 μm Channel Guides (squares)](image1)

![Figure 6. Predicted Lateral Confinement in a C-22 Y-Branch](image2)
4.2. Device structures fabricated using the SPP

The SPP has been used to fabricate three types of integrated optical device structures: a Y-branch interferometer, a directional coupler, and a traveling wave phase modulator. These device experiments were aimed at developing the necessary processing techniques for poled polymer devices, which differ significantly from those of Ti:LiNbO$_3$, and to determine the effects of secondary, as well as primary, materials parameters on device performance. Device optimization was not carried out for the first prototypes, but is now a routine part of our work.

4.2.1 Y-Branch Interferometer. The prototype Y-branch interferometer was fabricated by first defining the waveguide pattern in an aluminium electrode on a glass substrate. The guides were 7 µm wide. The dimensions of these particular electrodes were chosen to facilitate tests of the poling waveguide formation process and were not scaled to optimize the completed devices made from different materials. A 3 µm lower buffer layer was deposited on the substrate using UV curing epoxy, and then a 2 µm layer of the PC6S was spun onto the substrate. A gold poling electrode was deposited directly onto the the PC6S and the material was poled at 90°C for 5 minutes with an electric field of 100 V/µm. The poling electrode was etched off with dilute Aqua Regia and a glass slide containing the upper electrode over one arm of the interferometer was glued on with a 3 µm thick layer of optical epoxy. The epoxy also served as the upper buffer layer. To facilitate construction the upper electrode was much larger than necessary resulting in increased device capacitance and a reduced maximum modulation rate. Figure 7 illustrates the structure of the completed device and its dimensions. Prism coupling was used to inject 780 nm light into the device and guiding was observed in each arm. Modulation to a few kHz was detected in the output beam, indicating successful confinement and poling of the Y-branch.

![ITO Switching Electrode](image)

Figure 7. Structure of the PC6S Y-Branch Interferometer Made By SPP

4.2.2 Directional Coupler Switch. A second prototype device, a directional coupler, was fabricated using the C-22 material. The electrode defining the waveguide sections consisted of two 7 µm wide sections joining together to form a 13 µm wide, 1 cm long common section at a crossing angle of 3°. Two more symmetric 7 µm sections then diverged at the same angle, resulting in a total device length of 3.8 cm. The structure consisted of the lower aluminium electrode on a glass substrate covered with a 3 µm UV curing epoxy buffer layer. A 2 µm layer of the C-22 material was then spun on and covered with a 3 µm layer of polysiloxane. A planar gold electrode was then deposited for poling and the device was poled at 105°C for 15 minutes with an electric field of 90 V/µm in the C-22 layer. The upper gold electrode was then patterned to form the switching electrode over the central section of the coupler. Figure 8 illustrates the structure of the completed device and its dimensions. Prism coupling was used to inject 830 nm light into one input arm. With an applied voltage of 125 V the output was concentrated in the arm on the same side as the input channel, producing a bar state. With an applied voltage of 65 V a significant fraction of the light was switched to the opposite output arm producing a partial crossed state, as illustrated in figure 9. Complete switching was not observed due to the large crossing angle and the non-optimum waveguide dimensions.
4.2.3 Traveling Wave Modulator. A third device prototype, a traveling-wave phase modulator based on C-22, was fabricated using methods similar to those described above. This device, illustrated in figure 10, was designed to modulate light efficiently at 270 MHz. The device impedance was 9.5 $\Omega$, and a quarter-wave transformer was used to drive the modulator with a 0.5 W electrical input. The total active length was 3 cm. Figure 11 illustrates the response of the device when placed in one arm of an external interferometer. The modulation achieved at 270 MHz (Fig. 11a) was over 60%, almost exactly what we calculate it should have been by using the value of $r_{33}$ measured at low frequencies, indicating little dispersion in the E-O coefficient from DC to 270 MHz. Figure 11b illustrates the response at 1.0 GHz, which was small due to the impedance mismatch. However, this represents the first reported measurement of an E-O effect in a poled-polymer channel waveguide above a GHz. The combination of the unique features of the SPP and the properties of the C-22 and newer polymers shows some of the promise of organic materials for integrated optics.
5. CONCLUSIONS

We have demonstrated the application of a new fabrication technique, the selective poling procedure (SPP), for producing active, buried-channel waveguide devices in poled polymer films. The SPP combines both the fabrication and alignment steps required to make an active polymer device. The method has been demonstrated in several device formats showing promise for
real applications. We have observed a GHz response in a poled polymer channel waveguide, and have observed little or no
dispersion in the E-O coefficient in our materials. The combination of flexible fabrication methods and good material properties
implies that the field of organic integrated optics, now in its infancy, is well on its way toward achieving much of the promise
and expectations of organic electro-optic materials.

Future research on electro-optic polymer devices must address several research topics in poled polymer waveguides. From a
materials standpoint, larger electro-optic coefficients and lower linear absorption are required, and higher glass transition
temperatures are desirable. From a device standpoint, microwave losses must be measured and evaluated, the stability of the
poled states must be determined within the operating range of the devices, and numerous other device fabrication issues, such as
buffer layer selection, end-surface preparation, and fiber pigtailling must be solved. Finally, devices must eventually be integrated
with semiconductor sources and detectors. Such advances are expected to occur within the next several years, and may well
produce a new class of high-speed, cost-effective devices for integrated optics.

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7. REFERENCES

1. R. Lytel, G.F. Lipscomb, and J.I. Thackara, “Recent Developments in Organic Electro-optic Devices”, in Nonlinear
(1988).
2. R. Lytel and G.F. Lipscomb, "Nonlinear and Electro-optic Organic Devices", in Nonlinear Optical and Electro-active
4. R. Lytel, G.F. Lipscomb, P. Elizondo, B. Sullivan, and J. Thackara, "Optical Nonlinearities in Organic Materials:
Yoon, "Development of Polymeric Nonlinear Optical Materials", in Nonlinear Optics and Electro-active Polymers, P.N. Prasad
6. D.J. Williams, "Nonlinear Optical Properties of Guest-Host Polymer Structures", in Nonlinear Optical Properties of
10. G.J. Bjorklund et. al, "Organic and Polymeric Materials", in Research on Nonlinear Optical Materials: An Assessment,
(American Chemical Society), 1983.
12. K.D. Singer and A.F. Garito, "Measurements of Molecular Second-order Optical Susceptibilities Using DC Induced
14. C.C. Teng and A.F. Garito, "Dispersion of the Nonlinear Second-order Optical Susceptibility of an Organic System: p-
15. K.D. Singer, M.G. Kuzyk, and J.E. Sohn, "Second-order Nonlinear Optical Processes in Orientationally Ordered
16. J. Thackara, M. Stiller, E. Okazaki, G.F. Lipscomb, and R. Lytel, "Optoelectronic Waveguide Devices in Thin-Film