3D direct-write waveguides in diffusion photopolymers

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Abstract: Three-dimensional direct-write lithography can be used to pattern waveguides into diffusion photopolymer using low-power CW lasers as an alternative to femtosecond writing in glass. We show that polymer chemistry permits significant control of waveguide shape.

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1. Introduction and motivation

Direct-write lithography with femtosecond pulses can fabricate 3D integrated optics in inorganic materials, enabling high density, multi-dimensional optical circuitry [1,2]. We demonstrate that inexpensive, low-power, continuous wave (CW) lasers can write similar 3D structures in thick diffusion-mediated photopolymers [3,4]. This system has a number of advantages including potentially lower waveguide scattering loss due to the CW exposure and the gradient-index material response. Unlike femtosecond writing, which has a threshold-like response, the analog index control in polymer materials expands the range of possible devices to include, for example, gradient-index optics and sinusoidal Bragg gratings. Polymers can also be easily machined or cast to incorporate passive or active subcomponents, leading to fully hybrid optoelectronic circuits [5,6]. In this paper we demonstrate how the flexibility of polymer chemistry results in an additional benefit which is the ability to tailor the size and shape of the index profile in ways unavailable to high-power writing in glass.

2. Polymer initiation kinetics

Nonlinear absorption at the focus of a femtosecond pulse can locally heat normally transparent inorganic materials, inducing a structural change that raises the index of refraction. The analogous process in photopolymers uses a CW laser undergoing single- or multi-photon absorption which initiates polymerization, locally depleting the monomer concentration which in turn drives diffusion to increase the refractive index. Any of these steps can potentially be modified to control the shape of the index structure. Here we concentrate on the second step, the initiation of polymerization just after the absorption of an incident photon. For simplicity we will utilize a single-photon dye, although the development applies equally well to two- and three-photon absorption.

Equation (1) is a simplified rate equation for this process in which activated initiator concentration, $I^*$, is increased through absorption of optical intensity, $I_o$, by unactivated initiator, $I$, and decreased by unimolecular and bimolecular termination. The unimolecular termination rate constant, $k_{t1}$, is typical of ionic initiators, while the bimolecular termination rate constant, $k_{t2}$, is common to radical initiators. The initiation rate, $k_i$, and termination rate constants are controlled by choice of initiator species as well as additives such as inhibitors.

$$\frac{d[I^*]}{dt} = k_i I_o [I] - k_{t1} [I^*] - k_{t2} [I^*] [I^*]$$

Solving this equation in steady state, we find

$$[I^*] = \begin{cases} \frac{k_i}{k_{t1}} I_o & k_{t1} \gg k_{t2} [I^*] \\ \frac{k_i}{k_{t2}} I_o^{1/2} & k_{t1} \ll k_{t2} [I^*] \end{cases}$$

Equation (2) states the well-known result that the activated initiator concentration in steady state is proportional to the optical intensity when unimolecular termination dominates, and proportional to the square root of intensity when bimolecular termination dominates [7]. Intermediate cases between these two extremes are commonly described by an exponent, $\alpha$, varying from 1 to $1/2$, even though this is not the formal solution to (1) except at the two extremes.
If we assume that all other polymerization process steps are linear, the initiation process dominates the refractive index change locally in the polymer. The final refractive index is then proportional to the optical intensity to a power, $\alpha$. Using 3D direct-write lithography, this material is exposed to a translating CW Gaussian beam. The resulting index distribution can be calculated by integrating the intensity of a Gaussian beam raised to a power, $\alpha$, translating in $x$, through the $y$-$z$ plane.

$$\delta n(y,z) \propto \int I_{\text{GAUSSIAN}}^{\alpha}(x,y,z) dx \propto \left[1 + \left(\frac{z}{z_0}\right)^2\right]^{1/2\alpha} \exp\left[-2\alpha\left(\frac{y}{w_0}\right)^2\right]$$

where $w_0$ and $z_0$ are the waist radius and Rayleigh range of the Gaussian beam. Equation (3) reveals that for $\alpha = 1$, the expected index change is localized to the focus and is elliptical, just as in the case of femtosecond writing in glass. This ellipticity can be suppressed by shaping the writing beam [8]. As $\alpha$ decreases towards $\frac{1}{2}$, the ellipticity increases until at $\alpha = \frac{1}{2}$, where there is no confinement along the $z$ direction, as is shown in Figure 1. Since $\alpha$ can be modified by chemical formulation, this result demonstrates the dramatic control possible over the index distribution.

3. Experimental demonstration.

All of the experiments described here use one millimeter-thick InPhase Technologies Tapestry™ HDS3000 photopolymer packaged between one millimeter glass substrates. A frequency-doubled Nd:YAG laser at 532 nm is focused at a depth of 1.5 mm in the material with a 0.55 NA molded asphere corrected for 1.2 mm of spherical aberration. To limit depth-dependent spherical aberration and permit diffraction-limited access to the complete one millimeter polymer depth, the lens is stopped down to 0.3 NA.

The material is mounted on a high-precision, linear, 3D stage system with two tip/tilt stages, all under computer control [9]. In order to locate and orient the sample, a confocal microscope operating outside the material sensitivity band, at 633 nm, is implemented through the same aspheric objective. Although the polymer is well index-matched to the glass substrates, the confocal microscope can detect the small Fresnel reflection from the glass/polymer boundary. The $z$-coordinate of this reflection is measured at multiple transverse ($x,y$) locations in order to automatically adjust the sample tip/tilt to be normal to the lens axis and to identify the range of depths occupied by polymer.

The material is moved at a velocity of 2 mm/sec and the write shutter is opened to expose the material with a focused beam power of 1 $\mu$W. This power level is significantly lower than that used by femtosecond machining of glass, illustrating the very different physical processes. This greater sensitivity can be exploited, for example, to write 3D polymer index features much more quickly than is possible with a MHz pulsed laser – speeds of 20 meters/second with a CW, 50 mW laser at 0.55 NA have been demonstrated [10]. Higher CW power levels can be used to initiate two- and three-photon dyes for smaller feature size.
Figure 2 shows differential phase contrast microphotographs of the index distribution for a set of parallel uniform lines written 500 µm into the depth of the material and spaced by 35 µm. This polymer is initiated with a radical species, and thus, from Equation (3) we expect an increase of the axial feature size. Since these phase images are differential, a direct fit cannot be performed, but $\alpha$ appears to be roughly 0.7 in this material at these intensities. Volumetric data-storage experiments with cationic materials do not show this axial stretching of the index, consistent with the expected $\alpha$ value of one [10].

![SECTION VIEW](image1)

![TOP VIEW](image2)

Figure 2. Differential interference contrast phase microphotographs of the 3D index distribution. The $yz$ view on the left is a 20 µm-thick microtome slice, while the $xy$ view on the right is taken through the glass substrates.

4. Conclusions

We have shown theoretically and experimentally that very weak CW focused beams can write localized gradient-index features in the volume of a photopolymer. This integrated optics platform should exhibit most of the features of femtosecond nonlinear writing into glass but with the additional advantages of continuous writing, gradient-index response and analog index control. In this paper we have demonstrated that modifications of the organic chemistry enable significant control over the index profile.

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