INVESTIGATING NONLINEAR DISTORTION IN THE PHOTOPOLYMER MATERIALS

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ABSTRACT

Propagation and diffraction of a light beam through nonlinear materials are effectively compensated by the effect of self-trapping. The laser beam propagating through photo-sensitive polymer PVA/AA can generate a waveguide of higher refractive index in direction of the light propagation. In order to investigate this phenomenon occurring in light-sensitive photopolymer media, the behaviour of a single light beam focused on the front surface of photopolymer bulk is investigated. As part of this work the self-bending of parallel beams separated in spaces during self-writing waveguides are studied. It is shown that there is strong correlation between the intensity of the input beams and their separation distance and the resulting deformation of waveguide trajectory during channels formation. This self-channeling can be modelled numerically using a three-dimension model to describe what takes place inside the volume of a photopolymer media. Corresponding numerical simulations show good agreement with experimental observations, which confirm the validity of the numerical model that was used to simulate these experiments.

Keywords: Self-Written Waveguides; Acrylamide/polyvinyl alcohol; Photopolymer materials; Optical waveguides;

1. INTRODUCTION

Self-Writing of optical waveguides has been demonstrated in photopolymerizable materials \textsuperscript{1}. The self-writing phenomenon arises due to the balance of light wave diffraction via dispersion and confinement due to the increase in index of refraction along the path of beam because of photopolymerization \textsuperscript{2}. Using photopolymerizable materials, self-writing has been applied to create connections between optical fibers, to fabricate micro-tips at the end of optical fibers, biologically inspired microstructures and to manufacture strain sensors \textsuperscript{3-7}. During formation the beam of light writes its own waveguide, and then it is guided by the resulting channel generated. Beam self-trapping occurs along the propagation axis \textsuperscript{8}. However, non-linear material response can lead to the structure produced having non-ideal (irregular) shape characteristics. Such effects constrain photopolymer materials applications. To maximize the potential of these materials, deeper insights into the photo-physical and photo-chemical evolutions taking place during photo-polymerization and waveguide formation through the volume, are of increasing importance \textsuperscript{9-12}. Photopolymer materials are promising optical recording mediums, and are being actively studied for many practical applications such as hybrid optoelectronics, photo embossing, including the manufacture of refractive and diffractive optical elements \textsuperscript{15-20}. Through this work the optical properties required for a material to host self-written waveguides can be identified. The formation of such self-written waveguide structures is investigated and explored both numerically and experimentally. The input beams which expose the photopolymer material are carried by single mode optical fiber optics. During this exposing the beam emerging from top of the sample is imaged using CCD-camera. In this way the evolution of the index change is observed by monitoring the beam shape during waveguide creation. Then by studying the self-bending of light beam, during the formation of self-written waveguides in the dry photopolymer bulk, using parallel laser beams.

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2. MODULATION METHOD

The propagation of light in photopolymerizable materials can be described by a paraxial wave equation approximation as discussed, for example in Eq. 6.1. Using such models, SWWs formation can be examined using numerical simulations. Initially, we presumed that the physical properties such as the refractive index, and the absorptivity of the bulk sample is homogenous (i.e., uniform). A linearly polarized monochromatic incident beam (in x plane) with an angular frequency \( \omega \) is considered. In the bulk geometry, it can be given as:

\[
E(x, y, z, t) = E_0(x, y, z) \exp(i(n_0k_0z - \omega t)) \hat{x},
\]

where \( E_0(x, y, z) \) is the amplitude of the electric field, \( z \) is the light propagation direction, \( x \) and \( y \) are the transverse coordinates, and \( t \) is the time. The refractive index and the wave number inside the homogenous sample are denoted by \( n_0 \) and \( k_0 = 2\pi/\lambda \), respectively. In the material light propagation is described by the wave equation:

\[
\nabla^2 E + \left( k_0^2 n^2 - i\omega \mu \sigma \right) E = 0,
\]

where \( \nabla^2 = \partial^2/\partial x^2 + \partial^2/\partial y^2 + \partial^2/\partial z^2 \) is the Laplace operator. The instantaneous refractive index is given by \( n = n_0 + \Delta n \), where \( \Delta n \) is the change in the refractive index produced by the exposure. \( \mu \) is the absorptivity of the medium which is assumed to be equal to that of free space. The conductivity is denoted by \( \sigma \), and in this analysis, it appears in the form of an absorption parameter \( \alpha = \mu \sigma n_0 \). Substituting the expression in Eq. (1) into Eq. (2) we get:

\[
\frac{\partial^2 E}{\partial z^2} + 2ik_0 n_0 \frac{\partial E}{\partial z} + \nabla^2 E + 2k_0^2 n_0 \Delta n E + k_0^2 \Delta n^2 E + i\omega n_0 \alpha E = 0
\]

where \( \frac{\partial^2 E}{\partial z^2} \) is the 2-D Laplace operator. The refractive index change \( \Delta n \) induced during the self-writing process is small compared to the value of \( n_0 \). In addition, the higher order differential of the electric field can be neglected, i.e., \( 2k_0^2 n_0 \Delta n E \gg k_0^2 \Delta n^2 E \), and \( (\partial^2 E/\partial z^2) \approx 0 \). The resulting paraxial wave equation is:

\[
\frac{\partial E}{\partial z} = \frac{i}{2k_0 n_0} \nabla^2 E + i\omega n_0 \Delta n E - \frac{1}{2} \alpha E.
\]

It is notable that the induced index change and the attenuation parameter in these above equations are functions of both time and location, i.e., \( \Delta n(x, y, z, t) \) and \( \alpha(x, y, z, t) \). We can now utilize our model, which includes saturation effects, to describe the formation of the refractive index including both photosensitivity and photopolymerization effects. To describe the index change induced in the material space, \( \Delta n(x, y, z, t) \), we apply a commonly used model to describe the effects of photopolymerization. In this model, the refractive index evolution during the self-writing process is approximated using a simple phenomenological model:

\[
\frac{\partial \Delta n(x, y, z, t)}{\partial t} = AI(x, y, z, t) \exp\left(1 - \frac{\Delta n(x, y, z, t)}{\Delta n_s}\right),
\]

where \( t \) is the exposure time, and \( \Delta n_s \) is the fixed saturation value of the refractive index change, and \( I(x, y, z, t) \) is the local light intensity. The coefficient, \( A \), is a real coefficient that depends on the material properties, the number of photons \( p \), and the wavelength of the exposure light. In the photo-polymerization process, the number of photons involved in the process is typically assumed to be \( p = 1 \), (i.e., one-photon photosensitivity during photopolymerization process). Eq. (5) is an approximate model. It does not include an accurate description of the polymerization processes. For example in free radical systems, an accurate model of the photopolymer materials should involve calculations of the component concentrations using the related kinetic equations. The refractive index distribution would then be calculated using the Lorentz-Lorenz formula. This phenomenological model is used because the calculations will be simple and it is found that the resulting numerical predictions agree reasonably well with the experimentally results. We note that Eq. (4) is a nonlinear partial differential equation and generally no exact analytic solution will exist except for some specific cases. Therefore, numerical methods are necessary to solve it. Most such methods can be classified into two categories, i.e., finite-difference and pseudo-spectral methods. Generally, pseudo-spectral methods are faster by up to an order of magnitude while still achieving satisfactory accuracy.
3. RESULTS AND DISCUSSION

Our discussion of self-writing optical channels is divided into two parts: in the first part we experimentally measured the time varying amount of light absorbed by a dry acrylamide/polyvinyl alcohol (AA/PVA) based photopolymer bulk sensitized using an initial dye concentration and using a single beam of light. In the second part the model developed is then used to fit the experimentally obtained results. Estimations of some of the physical parameter values appearing in the model are found by carrying out a standard numerical fitting procedure. A laser Gaussian beam is incident on the photopolymer material and propagates along z-axis. The incident laser beam on the medium and the changes induced by localized photopolymerization reactions during the self-focusing and self-trapping processes in a bulk AA/PVA photopolymer material, are recorded. The creating of self-written waveguides is explored using the setup shown in Fig. 1. In this setup, the green light beam (λ = 532 nm) divided into two beams using 50:50 beam splitter (BS), then carried by fiber optic cables (FOC) in to which light is coupled using the micro-objectives. The output beams from the FOCs are focused onto the front face of the photopolymer bulk using optical fiber alignment stage. The generation of the waveguiding structures was detected by a CCD-Camera placed as shown parallel to the sample.

![Fig. 1. Schematic diagram of the setup used to monitor self-written waveguide using the single mode fibers optic beam.](image)

The photopolymer used, AA/PVA, was prepared as previously described 8, 33, 42. In this case, Eosin-Yellowish (EY) (C_{20}H_{6}Br_{2}Na_{2}O_{5}) is used. It is photosensitive to green light. Heating the resulting solution (holding the temperature less than 100 °C) in order to allow the water to evaporate, a conventional magnetic stirrer is used, under red light and in controlled laboratory conditions. It is necessary to wait until 50% of the water content has evaporated during heating, which typically requires over around 6 hours per 100 mL. During this process most of the water content evaporated, then the hot solution should be a highly viscous fluid which was carefully poured into cuvettes (12.5×12.5×4.5 mm). These cuvettes were placed in a vacuum container. The reason for doing so is as follows; to eliminate any air bubbles in the material introduced by the magnetic stirrer and to facilitate cooling of the hot material using the low temperature produced by low air pressure. The material cuvettes were stored in a dark room for a long period (normally several days) to allow uniform solid state formation. Finally the solid bulk AA/PVA photopolymer samples suitable for self-writing experiments were ready for exposure 33.

Self-writing provides a technique for the direct fabrication of waveguide structures within a material. SWWs can form in various materials. The common attribute of such materials is that a refractive index change occurs when they are exposed to light. A standard theoretical model is used to predict both the evolution of the light intensity distribution and the channel formation inside the material during the exposure. Corresponding numerical simulations show good agreement with observations, confirming the validity of the numerical model used to simulate these experiments. The discrepancies between our experimental results and the predictions of the model indicate that there are more complicated photo-physical and photo-chemical processes taking place than predicted by the phenomenological model used 8, 21, 28, 31. Such additional complexity requires the development of a more detailed theoretical model. In the first part of our work, we used one Gaussian writing...
beam to produce a single channel waveguide in a photopolymer bulk. We implemented a simple phenomenological model to describe the material’s response during the propagation. Fig. 2 shows the comparison between the experimental and theoretical results for one Gaussian beam that propagated along $z$ (of wavelength $\lambda = 532 \text{ nm}$ and power $0.5 \text{ mW}$), under room conditions. Additionally, the refractive index change $\Delta n$ distribution is plotted.

![Fig. 2](image)

**Fig. 2.** Numerical simulations and experimental results for light intensity distribution along $z$-axis are shown in (a) and (b), respectively. (c) Shows the contour plotted of refractive index change $\Delta n$ distribution.

In this study the dye used is Eosin-Yellowish (EY), and the thickness of photopolymer bulk (AA/PVA) is 8 mm. The formation of the self-written waveguide in the dry photopolymer material is shown in the Fig. 2, both simulations and experimental measurements are presented. Experimentally, we observed that the Gaussian beam has self-written the optical waveguide channel along the full length of the sample, i.e., $0 \leq z \leq 8 \text{ mm}$. Recalling the simulation results presented in Fig. 2a and the experimental results in Fig. 2b, they both exhibit the same SWW evolution qualitatively, supporting the validity of the model and procedure used. It is worth noting that the position of highest intensity starts to move along the $z$-axis away from the input boundary. This region of relatively higher intensity is referred to as "primary eye". The evolving index change counteracts the diffraction intensity of the input beam. Eventually the cross sectional distribution, at every point in $z$-axis is the same, indicating that self-trapping has occurred. As we can see from Fig. 2c, by this time the refractive index change $\Delta n$ becomes sufficient to overcome the diffraction effects. During each time step the predicted index is used to update the resulting light intensity distribution using Eq. (5), and the optical waveguide shape evolves with time.

A generalized experimental setup like that illustrated in Fig. 1, can be implemented for exposure with fiber optic cables inputs. In this way several waveguides can be fabricated simultaneously in the material. For instance, we can adjust two beams to generate two parallel waveguides in the same direction as the $z$-axis. In general, the beam trajectories are more complex with photopolymerization and index self-modulation, both act simultaneously and dynamically to dominate the formation process. In this case we can start with two fibers optic cables that are relatively separated by large distance (4mm). Fig. 3, shows the
two parallel beams propagating through photopolymer bulk from left to right in the $z$-direction. During this process, the dye molecules are being consumed, which is leading to a gradual reduction during the absorption in the exposed regions. As can be seen from Fig. 3a, the experimental observations for two parallel waveguides are created inside the material. They do not effect each other or effect each other’s trajectories, which is dependent on the separation of the beams. The theoretical results shows good agreement comparing with experimental result, see Fig. 3 b and c. It is worth noting that the beam trajectories are not bending because there is no big change in the refractive index in the area between the two beams, see Fig. 3b. This indicates that not enough photons reach this region, which means there is no reaction to the change in refractive index.

![Image](image)

**Fig. 3.** Evaluate of self-written waveguides for two parallel light beams separated by (4 mm), through studying: (a) the experimental observation of light evolution inside photopolymer material. (b) and (c) the theoretical distributions of refractive index change $\Delta n$ and light beam intensity, respectively.

However, changing of separation distance between two parallel light beams is studied. Fig. 4 shows the self-bending evolution of the self-written waveguides during the exposure of the photopolymer medium using two parallel light beams separated by 1.5 mm. As can be seen from the experimental result the two Gaussian beams are divergent during SWWs process shown in Fig. 4a. This evaluation is due to the higher refractive index changing between trajectories, the refractive index changes by $\Delta n$, between trajectories resulting in changing the direction of beam propagation that also depends on the illumination distances between two beams (separation of two beams). It is estimated that beam diverges with 1.5 mm distance separation, see Fig. 4. To examine this evaluation in Fig. 4c the normalized light intensity value is plotted during the exposure process. We note that there is good agreement between the observed experimental and theoretical results, which supports the validity of the model and procedure used.
Fig. 4. Self-bending or self-diverging of two parallel light beams separated by (1.5mm) during self-written waveguides (a) the experimental observation of light evolution inside photopolymer material; (b) and (c) the theoretical distributions of refractive index change $\Delta n$ and light beam-intensity respectively.

In order to improve our visualization of self-bending evolution, the process is designed by decreasing a separation distance to 0.5 mm, see Fig. 5. As noted, due to the refractive index change $\Delta n$ and reduced distance in this case, two illuminated beams are converging as a single beam, see Fig. 5b. Generally, from this study we can conclude that the distance between parallel light beams inside the photopolymer medium is very important, which can result in two parallel beams diverging into two wide distance channels or merging into a single waveguide.

Fig. 5. Evaluation of self-merging to the two parallel light beams separated by (0.5mm) during self-written waveguides, through studying: (a) the experimental observation of light evolution inside photopolymer material; (b) and (c) the theoretical distributions of refractive index change $\Delta n$ and light beam-intensity respectively.
These results are observed by the complex waveguides created during a bending of Gaussian beams in the AA/PVA photopolymer bulk. The self-writing fabrication method of waveguides shows great potential for future practical applications and integration with existing devices. The self-writing approach offers great potential for the fabrication of couplers and splitters for application in integrated optical devices and components.

4. CONCLUSION

Self-written waveguides can form in photosensitive materials, i.e. materials where light at specific wavelengths induces long lasting refractive index changes. In this paper, self-written channel waveguides have been formed in bulk AA/PVA photopolymer materials. This study includes the experimental and theoretical studies for the self-bending of waveguides during the self-writing process. The results achieved are in good agreement with the numerical simulations and experimental observations, which confirms the validity of the numerical model which is used to simulate these experiments. The investigations of self-diverging and self-merging of two parallel light beams during self-written waveguides are studied, which depends on the separation distance between the light beams. In conclusion, all these results indicate that this type of self-written waveguide shows potential for practical applications such as optical splitters and couplers in integrated optics.

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REFERENCE


