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SELF-WRITTEN NET-WAVEGUIDES USING PHOTOPOLYMER MEDIA

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ABSTRACT

Recently the numerical and experimental results of optical self-written waveguides (SWWs) has been demonstrated intensively in the photopolymer media. In order to further understand the mechanism of self-written net-waveguide in photosensitive polymers, light-induced material response is analyzed. Optical net-waveguide trajectories formed using solid bulk of acrylamide/polyvinyl alcohol (AA/PVA) photopolymer material. As part of this work presents a studying of non-linear optics in photopolymer systems to form a net-waveguides. Which deals with the nonlinearity behaviors of transmitted light in photopolymer media, during refractive index changed throughout the optical self-propagating process. The self-interactions of crossing beams inside photopolymer material during SWWs process are studied. It is shown that there is good agreement between the numerical simulation results and experimental observations. These are confirmed 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

Photopolymerization process and nonlinearity optics behaviors are distinguished fields that have been studied for decades. In recent years the photopolymer media have been used attractively with much attention for use in optical interconnects and in integrated devices for optical communications in the access network and the home network areas ¹. Which are intersect with the observation of wave propagation through photopolymer media during self-written waveguide (SWW) process ²⁻⁵.

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 ⁶⁻¹⁰. 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 ¹¹⁻¹⁸. 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 ¹⁹⁻²⁴. 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 ²⁵⁻³³.

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For the first time it is demonstrated that permanent optical net-waveguides can be written in a solid acrylamide/polyvinyl alcohol (AA/PVA) photopolymer material ³⁴. In this article presents a studying of non-linear optics in photopolymer systems to form a net-waveguides. This has led to studies on the nonlinear dynamics of transmitted light in photopolymer media, particularly for optical self-propagating during refractive index changed. A standard theoretical model is used to predict both the evolution of the light intensity distribution and the trajectories formation inside the material during the exposure ^{2, 3, 11}. The experimental results and the numerical simulations are compared, and good agreement is obtained. The diffraction of a light beam as it propagates through a medium can be effectively compensated by self-trapping. A laser beam propagating through a nonlinear medium can generate a waveguiding action, i.e. a higher refractive index, along the direction of the light propagation ¹¹⁻¹⁸.

In this paper, the photopolymerization process is studied in three-dimensional the bulk samples (i.e. millimeters thick). In this way, the optical properties required for a material to host self-written waveguides (SWWs) can be identified. The formation of such SWW structures is investigated both numerically and experimentally. The input beams which expose the photopolymer material are, in all cases, input into the sample throughout pin holes using mask. During the exposure of the photopolymer, the scattered light, emerging from the top side of the sample is imaged using a CCD-camera. In this way the evolution of the index change (SWW) is observed by monitoring the waveguide shape during formation. In this paper the self-interactions of crossing beams to create net-waveguide trajectories is examined. Self-interactions of crossing beams are shown to take place during the formation of net-SWWs, in a dry photopolymer bulk exposed by two simultaneously input crossing laser beams.

2. PHOTOPOLYMERIZATION PROCESS:

A photopolymer material undergoes photopolymerization when exposed to incident light of an appropriate wavelength (depending on the photosensitizer chosen). The mechanism of photo-polymerization that occurs in the PVA/AA material is known as free radical polymerization ³³⁻³⁷. This reaction mechanism results in the recording of permanent refractive index changes. Throughout the course of the photochemical process, four main phases of the reaction exist: (i) Initiation, (ii) Propagation, (iii) Termination and (iv) Inhibition ³⁸.

The initiation: process involves two steps. The first step is the reaction between the photo-initiator and the electron donor, which leads to the production of initiator (primary) radicals, R^{\bullet} . These radicals can react with the monomers to produce chain initiators, $M^{\bullet 35, 39}$.

$$I \xrightarrow{h\nu} 2R^{\bullet}, \tag{1a}$$

$$R^{\bullet} + M \xrightarrow{k_i} M_1^{\bullet} = Chain \, Initiator \,, \tag{1b}$$

where *M* is the monomer and k_i is the chain initiation kinetic constant. Eq (1b), is the second step in the initiation process (chain initiation), in which the primary radicals resulting from the absorption of photons react with the monomer to produce the chain initiating species M_1^{\bullet} . The kinetic rate constant for this step is k_i , the chain initiation kinetic constant $\frac{2, 21, 40-42}{2}$.

The resulting monomer radical, M_1^{\bullet} , from the initiation stage can now react with other monomer molecules, M. The reaction forms a growing chain. This growing chain is known as a macroradical chain and has a free radical at its tip. Eq. (2) indicates how a monomer is added to a growing macroradical chain of repeated n monomeric units.

$$M_n^{\bullet} + M \xrightarrow{k_p} M_{n+1}^{\bullet}$$
. (Growing polymer chain) (2)
Where k_p (cm³ mol⁻¹ s⁻¹) is the propagation rate constant and M_n^{\bullet} is the macroradical of *n* repeated units.

Termination: is the process that brings the propagation stage to an end, i.e. causes the polymer chains to stop growing. There are three different reactions that can lead to termination $\frac{40, 43}{2}$:

$$M_n^{\bullet} + M_m^{\bullet} \frac{\kappa_{tc}}{\kappa_{td}} M_{n+m}, \tag{3 a}$$

$$M_n^{\bullet} + M_m^{\bullet} \xrightarrow{\kappa_{la}} M_n + M_m, \qquad (3 b)$$

$$M_n^{\bullet} + R^{\bullet} \xrightarrow{\kappa_{LP}} M_n R, \qquad (3 c)$$

- Eq. (3 a) shows bimolecular termination by combination. This occurs when two separate growing polymer chains meet and form one long "dead polymer chain" as described by the term M_{n+m} . k_{tc} (cm³ mol⁻¹ s⁻¹) is the combination rate kinetic constant.
- Eq. (3 b) shows bimolecular termination by disproportionation. This occurs when two separate growing polymer chains meet and terminate as two individual chains described by M_n and $M_m \cdot k_{td}$ (cm³ mol⁻¹ s⁻¹) is the disproportionation rate kinetic constant.
- Eq. (3 c) presents primary radical termination. This is when a growing macroradical reacts with a primary radical to form an inactive dead polymer chain.

Inhibition: is the reaction mechanism caused by the reaction of primary radicals and macroradicals with an inhibitor such as oxygen. An inhibitor is a substance that decreases the rate of a chemical reaction by removing radicals. The following reactions can occur:

$$R^{\bullet} + Z \xrightarrow{k_{Z,R}^{\bullet}} (R + Z^{\bullet}, and/or \ R \ Z^{\bullet}), \qquad (4 a)$$

$$M_n^{\bullet} + Z \xrightarrow{\kappa_{Z,M^{\bullet}}} (M_n^{\bullet} + Z^{\bullet}, and/or M_n Z^{\bullet}), \qquad (4 b)$$

³Dye^{*} +
$$Z \xrightarrow{R_{Z,D}} Leuco Dye,$$
 (4 c)

where Z is the inhibitor concentration and $k_{Z,R^{\bullet}}$, $k_{Z,M^{\bullet}}$ and $k_{Z,D}$ are the inhibition rate constants. In free radical polymerization the effects of inhibition suppress the creation of macroradicals by scavenging primary radicals as shown in Eq. (4 a). They also inhibit the macroradicals that have already been created as is shown in Eq. (4 b). Inhibition serves to stop the production of polymer chains ³³⁻³⁷. Inhibition typically leads to a dead band at the start of exposure ²². In order to improve our visualisation of the entire photoreaction, the process is summarized using the flow chart in Fig. 1.



Figure 1 Flow chart illustrating photopolymerization in a free-radical material ⁴¹⁻⁴⁷.

3. MODULATION METHOD

In the photopolymerizable materials, light propagation can be described using a paraxial wave equation approximation as discussed, for example in ^{8,48-51}. Using such models SWWs formation can be examined by 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 ω is considered ⁵². In the bulk geometry it can be given as:

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$$\boldsymbol{E}(x, y, z, t) = \boldsymbol{E}_0(x, y, z, t) \exp[i(n_0 k_0 z - \omega t)]\hat{\boldsymbol{x}},$$
(5)

where $E_0(x, y, z, t)$ 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 ^{11, 53, 54},

$$\nabla^2 \boldsymbol{E} + \left(k_0^2 n^2 - i\omega\mu\sigma\right)\boldsymbol{E} = 0., \tag{6}$$

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 Δn is the change in the refractive index produced by the exposure. μ is the permeability of the medium which is assumed to be equal to that of free space. The conductivity is denoted by σ , and in this analysis it appears in the form of an absorption parameter $\alpha = \mu c \sigma/n_0$. Substituting the expression in Eq. (5) into Eq. (6) we get that:

$$\frac{\partial^2 E}{\partial z^2} + 2ik_0 n_0 \frac{\partial E}{\partial z} + \nabla_{\perp}^2 E + 2k_0^2 n_0 \Delta n E + k_0^2 \Delta n^2 E + ik_0 n_0 \alpha E = 0$$
(7)

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

$$\frac{\partial E}{\partial z} = \frac{i}{2k_0 n_0} \nabla_{\perp}^2 E + ik_0 \Delta n E - \frac{1}{2} \alpha E.$$
(8)

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 use our model, which includes the effects of saturation, to describe refractive index formation including both photosensitivity and photopolymerization effects ^{5, 48, 49, 55-57}. 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 photo-polymerization ¹¹. In this model, the refractive index evolution during the self-writing process is approximated using a simple phenomenological model ^{11, 48, 55, 58}:

$$\frac{\partial \Delta n(x, y, z, t)}{\partial t} = AI(x, y, z, t)^{p} \left(1 - \frac{\Delta n(x, y, z, t)}{\Delta n_{s}} \right), \tag{9}$$

where *t* is the exposure time, Δn_s is the fixed saturation value of the refractive index change, and I(x, y, z, t) is the local light intensity ^{11,48,55,58} where $I(x, y, z, t) = |E(x, y, z, t)|^2$. 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 photo-polymerization process) ^{11,48}. Eq. (9) typically employed to calculate the index change because it can be relatively easily combined with the paraxial wave equation to give useful predictions. It is clear that Eq. (9) 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 ⁵⁹. 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 ^{11,60}. We note that Eq. (8) is a nonlinear partial differential equation and generally no exact analytic solution will exist except for some specific cases ⁶¹. Therefore numerical methods are necessary to solve it. Most such method can be classified into two categories, i.e., finite-difference ⁶²⁻⁶⁴ and pseudo-spectral methods ⁶⁵⁻⁶⁷. Generally pseudo-spectral methods are faster by up to an order of magnitude while still achieving satisfactory accuracy ⁶⁸.

4. EXPERIMENTAL AND NUMERICAL RESULTS

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*-direction. 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. 2. In this setup, the green light beam ($\lambda = 532 \text{ nm}$) divided into two beams using 50:50 beam splitter (BS) splits the exposing beam to produce two mutually coherent plane waves of equal intensity, i.e. $I_1=I_2$. As Fig. 2 the two beams are focused onto the front face of the mask (contains holes on a straight line), which touches with front face of the photopolymer bulk. The generation of the waveguiding structures was detected by a CCD-Camera placed as shown parallel to the sample.

The photopolymer used, AA/PVA, was prepared as previously described ^{2, 3, 11-17, 60}. In this case, Eosin-Yellowish (*EY*) ($C_{20}H_6Br_4Na_2O_5$) 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 ^{2,3, 11-17, 60}.



Figure 2. Schematic diagram of the setup used to monitor self-written net-waveguide.

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 ^{11, 48, 55, 58}. Such additional complexity requires the development of a more detailed theoretical model. Experimentally, we used tow Gaussian writing beam incidents on mask include holes that to produce W-shaped channel waveguide in a photopolymer bulk, see fig. 3. Fig. 3 (a) and (b) shows the experimental observations for Gaussian beam passed throughout two holes that propagated along *z* (of wavelength λ = 532 *nm* and power 0.5 *mW*), under room conditions. Additionally, the observation for net-SWW of Gaussian beam passed throughout four holes is shown in fig. 3(c). In this study the dye used is Eosin-Yellowish (*EY*), and the thickness of photopolymer bulk (AA/PVA) is 8 *mm*.



Figure 3 Experimental observation for net-Gaussian beams evolution inside photopolymer material during SWW process: (a) and (b) two holes, (c) four holes.

The numerical simulation results of the self-written waveguide in the dry photopolymer material is shown in the fig. 4. We obtained that the Gaussian beam has self-written the optical waveguide channel along the full length of the sample, i.e., $0 \le z \le 8$ mm. Recalling the experimental results presented in fig. 3 and the simulation results in fig. 4, 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. As can be seen from fig. 4 (a) and (b), the numerical simulation results of W-shaped of net-SWW using two holes are shown. In order to improve our visualization of SWW evolution, the process is designed by increasing holes numbers, see fig. 4 (c) and (d). In this way several waveguides can be fabricated simultaneously in the material. In general, the beam trajectories are more complex with photopolymerization and index self-modulation, both act simultaneously and dynamically to dominate the formation process $^{2,3,11-17,60}$.



Figure 4 Numerical prediction (light intensity distribution) for interactions of net-waveguides propagating through photopolymer media: (a) and (b) two holes with different exposure incident light angles, (b) four holes, and (c) six holes.

These results are observed by the complex waveguides created during propagation Gaussian beams in the AA/PVA photopolymer bulk. We note that there is good agreement between the observed experimental and theoretical results,

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which supports the validity of the model and procedure used. 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.

5. CONCLUSION

The procedures of formation the self-writing waveguide has been demonstrated in photopolymerizable materials. The phenomenon of self-writing arises because of the balance of the optical wave diffraction via dispersion and confinement due to the increase in index of refraction along the path of light beam because of photopolymerization. Using photopolymerizable materials, self-writing has been applied to create light trajectories. During formation the beam of light writes its own waveguide, and then it is guided by the resulting trajectories generated. Beam self-trapping occurs along the propagation axis. However, non-linear material response can lead to the structure produced having irregular shape characteristics. Such effects constrain photopolymer materials applications.

Through this work the optical properties required for a material to host SWWs can be identified. The formation of such net-SWW structures is investigated and explored both numerically and experimentally. The input Gaussian light beams are exposed in front of the photopolymer. During this exposing the emerging beam is imaged from the top of the sample using a CCD camera. In this way, the evolution of the index change is monitored by observing the beam shape while creating the waveguide. Then by studying the self-formation of light trajectories, during the creation of SWWs process in the dry photopolymer bulk. Results are observed the complex net-waveguides that created during propagation Gaussian beams in the AA/PVA photopolymer bulk.

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