

Optical formation of stable waveguiding structures from a photopolymerisable composition with a nonpolymerisable component

S.N. Mensov, Yu.V. Polushtaitsev

Abstract. We report formation of stable dielectric waveguiding structures from a photopolymerisable composition containing a nonpolymerisable component by optical radiation. A computer simulation has shown that the use of nonpolymerisable additives not only retains the self-trapping modes of incident radiation but also provides matching conditions for the synthesised waveguiding structure with standard optical fibres at telecommunication wavelengths. The efficiency of these nonlinear wave processes for connecting single-mode fibres SMF-28 is experimentally confirmed.

Keywords: fibre optical connection, stable waveguiding structures, multicomponent photopolymerisable compositions, nonpolymerisable component, nonlinear wave processes.

1. Introduction

Currently, the popularity of photopolymerisable compositions (PPCs) as a base of optical media with a nonuniformly distributed refractive index constantly increases [1–10]. Modern PPCs are transparent materials with a high photosensitivity: photopolymerisation readily occurs under visible light with an intensity of several mW mm^{-2} [11–13]. Nonlinear optical wave processes in these media make it possible to form nonexpanding channels by laser beams with a significant diffraction divergence [1–4], fabricate crooked and branched waveguiding structures [14], and connect even misaligned optical fibres with end faces spaced by rather large distances [5, 6, 14]. The refractive index distribution in these polymer structures is set by their exposure duration, which determines the monomer-to-polymer concentration ratio in a PPC. However, one can also implement a gradient refractive index in the polymer bulk by introducing a diffusant that is inert to photopolymerisation: a nonpolymerisable component (NC) with a refractive index significantly differing from that of the polymer [8–10]. At a limiting PPC conversion this structure does not contain a reactive monomer and, therefore, is stable. Hence, it can be used not only in the IR range but also in the visible range.

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In this study we considered the processes of optical formation and ‘fixation’ of waveguiding structures from an NC-containing PPC. The possibility of applying nonlinear wave processes to form stable connecting fibreoptic elements is investigated both by numerical simulation and experimentally.

2. Optical formation of waveguiding structures in a multicomponent PPC

The initial stage in the polymerisation of liquid compositions is always characterised by significant mobility of molecules of the polymer that is formed in the monomer bulk [15]. The presence of low-molecular organic solvents in PPC enhances these processes. Therefore, we studied the light propagation in a liquid photopolymerisable NC-containing medium based on the diffusion model [10], according to which violation of equilibrium in a system under consideration leads to the formation of diffusion fluxes of (i) nonpolymerisable component (j_N), (ii) monomer (j_M), and (iii) polymer (j_P), which are described by the expressions

$$\begin{aligned} j_N &= -(\alpha_{NM}M + \xi_{PN}P)\nabla N + \alpha_{NM}N\nabla M + \xi_{PN}N\nabla P, \\ j_M &= \alpha_{NM}M\nabla N - (\alpha_{NM}N + \eta_{MP}P)\nabla M + \eta_{MP}M\nabla P, \\ j_P &= \xi_{PN}P\nabla N + \eta_{MP}P\nabla M - (\xi_{PN}N + \eta_{MP}M)\nabla P, \end{aligned} \quad (1)$$

where M , P , and N are, respectively, the mass fractions of the monomer, polymer, and NC; the coefficients α_{NM} , $\xi_{PN} = \alpha_{NM}\exp(-PIP^*)$, and $\eta_{MP} = \eta_M\exp(-PIP^*)$ characterise the interdiffusion processes between the monomer and NC, and polymer and NC, and the polymer and monomer, respectively; η_M is the monomer self-diffusion coefficient; and P^* is the polymer mass fraction at which a characteristic change in its viscosity occurs ($P^* = 0.05–0.2$) [10]. During photopolymerisation the NC is redistributed over the composition volume and is not involved in the formation of polymer (the total NC content N_0 is constant), and the monomer not only participates in the interdiffusion processes but is also transformed into polymer molecules at a rate $V(M, N)$ [10]:

$$\begin{aligned} \frac{\partial N}{\partial t} + \text{div}j_N &= 0, \\ M + N + P &= 1, \\ \frac{\partial M}{\partial t} + \text{div}j_M &= -V(M, N). \end{aligned} \quad (2)$$

With allowance for this phenomenon, the analysis of the optical formation of a waveguiding channel under a mono-

chromatic Gaussian beam with $E(x, 0, H) = E_0 \exp[-x^2 \times (2a_0^2)^{-1}]$, propagating along the z axis in an initially homogeneous, weakly absorbing PPC, was based on numerical solution of the quasi-stationary parabolic equation for the complex field amplitude [7]:

$$2ik \frac{\partial E(x, z, H)}{\partial z} = \Delta_{\perp} E(x, z, H) + 2k^2 \frac{\Delta n(x, z, H)}{n_M} E(x, z, H), \quad (3)$$

in which the nonlinear additive to the PPC refractive index ($\Delta n = n - n_M$) during photopolymerisation was calculated from the system of material equations [10]

$$-\frac{\partial N}{\partial t} = \text{div} \mathbf{j}_N,$$

$$-\frac{\partial M}{\partial t} = V(I, M, N) + \text{div} \mathbf{j}_M, \quad (4)$$

$$V(I, M, N) = \gamma \frac{I}{H_0} M \left[-\ln \left(\frac{M}{1-N} \right) \right]^{1-1/\gamma},$$

$$n = n_M M + n_P P + n_N N.$$

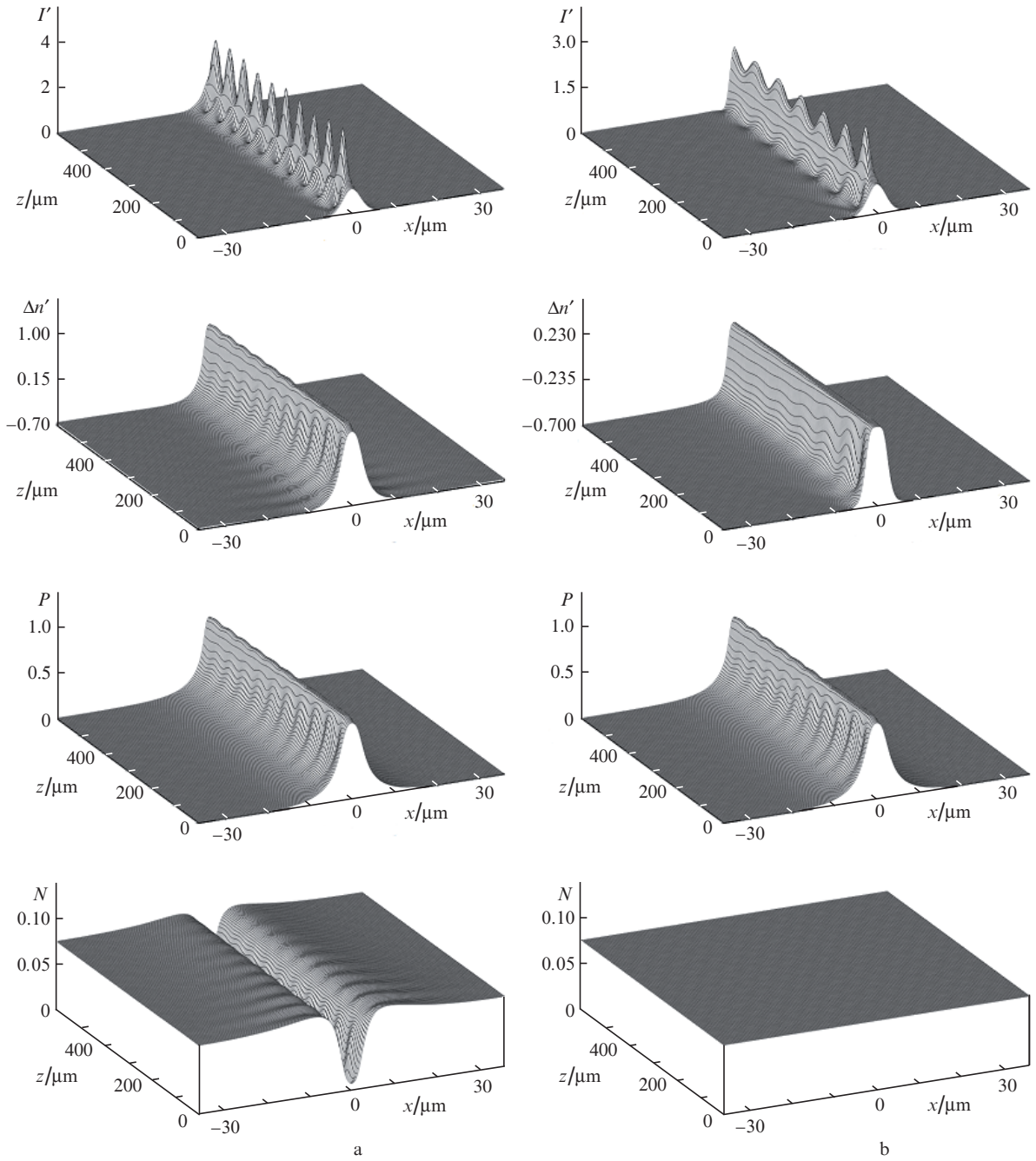


Figure 1. Distributions of the incident radiation intensity $I' = I/I_0$; nonlinear additive to the refractive index, $\Delta n' = \Delta n/\Delta n_{PM}$; and the polymer and NC mass fractions in a waveguiding channel formed in a PPC (a) taking into account and (b) neglecting the interdiffusion of the composition components ($t = t_p$).

Here, a_0 is the beam half-width; k is the wave number; $I(x, z, H) \sim |E(x, z, H)|^2$ is the incident radiation intensity (correspondingly, $I_0 \sim E_0^2$);

$$H(x, z, t) = \int_0^t I(x, z, \tau) d\tau$$

is the exposure; γ and H_0 are the parameters characterising the composition contrast; and n_M , n_P and n_N are the refractive indices of the monomer, polymer, and NC, respectively.

The simulation results showed that the use of the NC in the PPC composition does not violate the optical-radiation self-trapping modes [1–4]: the exposure to a light beam leads to the formation of a long nonexpanding waveguiding channel. As an example, Fig. 1a shows the results of modification by a Gaussian beam ($a_0 = 3 \mu\text{m}$, $\lambda = 0.63 \mu\text{m}$), which is used for optical synthesis of holographic structures in a multicomponent PPC [10] ($N_0 = 0.07$, $t_p = H_0/I_0 = 1 \text{ s}$, $t_p/t_d = t_p\eta_M/a_0^2 \approx 0.67$, $\alpha_{NM} = 10\eta_M$, $\gamma = 3$, $P^* = 0.13$, $\Delta n_{PM}/n_M = 0.01$, $\Delta n_{NM}/n_M = -0.1$, $\Delta n_{PM} = n_P - n_M$, $\Delta n_{NM} = n_N - n_M$).

One can easily see that, up to the end of channel formation, the channel periphery remains in the liquid state (a mixture of an unmodified monomer and NC). The refractive index n_0 at the periphery is proportional to the initial contents of these components: $n_0 = n_M(1 - N_0) + n_N N_0$, while in the paraxial region of the channel the refractive index is n_P . Therefore, the refractive index amplitude in the channel [the difference between the maximum (at the channel centre) and minimum (at the periphery) values of the refractive index] is determined by not only the nature of the oligomer (the composition material [11–13]) but also by the refractive index and content of the NC (Fig. 2):

$$\Delta n_c = \Delta n_{PM} - \Delta n_{NM} N_0. \quad (5)$$

An increase in the initial content of the NC with a refractive index larger than that of the monomer ($n_N > n_M$) leads to a decrease in the amplitude of the channel refractive index [Fig. 2, surface (1)] and even to inversion of its profile (at

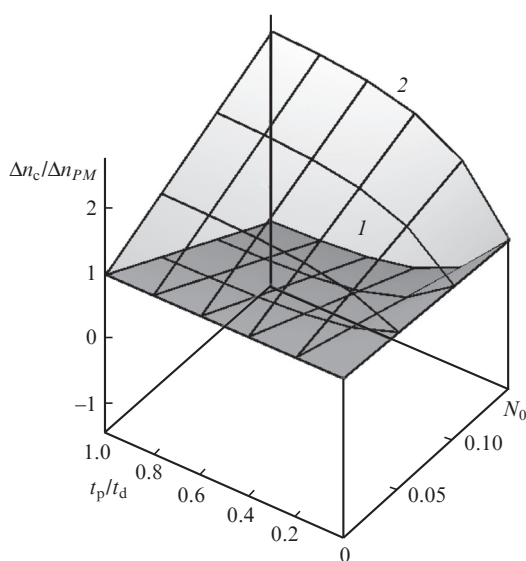


Figure 2. Dependences of the refractive index amplitude in the channel ($t = t_p$) on the ratio t_p/t_d and the NC content N_0 at (1) $n_N > n_M$ and (2) $n_N < n_M$.

$\Delta n_{PM} < \Delta n_{NM} N_0$) [8, 9]. However, a channel cannot be formed due to the radiation self-trapping in this defocusing medium. Vice versa, an introduction of the NC with $n_N < n_M$ into the composition allows one to increase significantly the amplitude of the refractive index profile in the channel: its gradient increases with an increase in the NC content [Fig. 2, surface (2)] [10].

A waveguiding channel can also be formed in a PPC with the NC in the absence of diffusion redistribution of components ($\eta_M = 0$, $\alpha_{NM} = 0$; Fig. 1b). However, in this case the polymer should contain the NC with $n_c = n_P(1 - N_0) + n_N N_0$ in its structure (in the paraxial region). Therefore, $\Delta n_c = \Delta n_{PM}(1 - N_0)$ (the line $t_p/t_d = 0$ in Fig. 2). However, to form optically gradient structures, one needs a nonuniform NC distribution in the polymerisable bulk [10]. Consideration of the diffusion processes in the PPC allows one to determine the polymerisation conditions under which the NC can be displaced from the paraxial region of the channel during its formation (Fig. 1a): the polymerisation time $t_p = H_0/I_0$ should exceed the characteristic diffusion time $t_d = a_0^2/\eta_M$ (Fig. 2).

3. Fixation of refractive index gradients

The gradient structure formed by a light beam remains sensitive to illumination, because the completely modified monomer preserves photopolymerisation activity. Nevertheless, the amplitude of the refractive index profile in the channel is sufficient for transferring IR radiation, which does not initiate polymerisation (for example, the radiation with $\lambda \sim 1.55 \mu\text{m}$, which is working in fibre systems) [5, 6]. At the same time, propagation of visible light causes polymerisation at the channel periphery, thus increasing its width [Fig. 3, curve (1)] and changing the waveguide parameter. In addition, a waveguiding polymer channel enclosed in a liquid coating may degrade in the course of time due to the diffusion [Fig. 3, curve (2)]: a part of mobile polymer and the NC cladding are distributed over the periphery, tending to concentration equilibrium.

The waveguiding channel can be stabilised using additional polymerisation of the monomer by radiation homogeneously exposing the entire PPC volume. In this case, the liquid monomer at the channel periphery is transformed into a solid polymer. Naturally, in the case of complete conversion

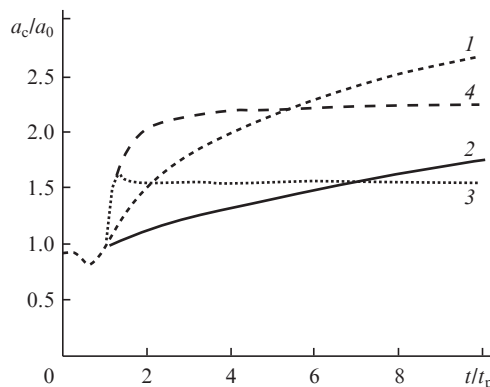


Figure 3. Change in the width a_c of optically formed waveguiding channel in the initial cross section ($z = 0$) at (1) exposure to a Gaussian light beam, (2) diffusion spread of unfixed channel, (3) fixing channel by 'homogeneous' radiation, and (4) incomplete additional polymerisation.

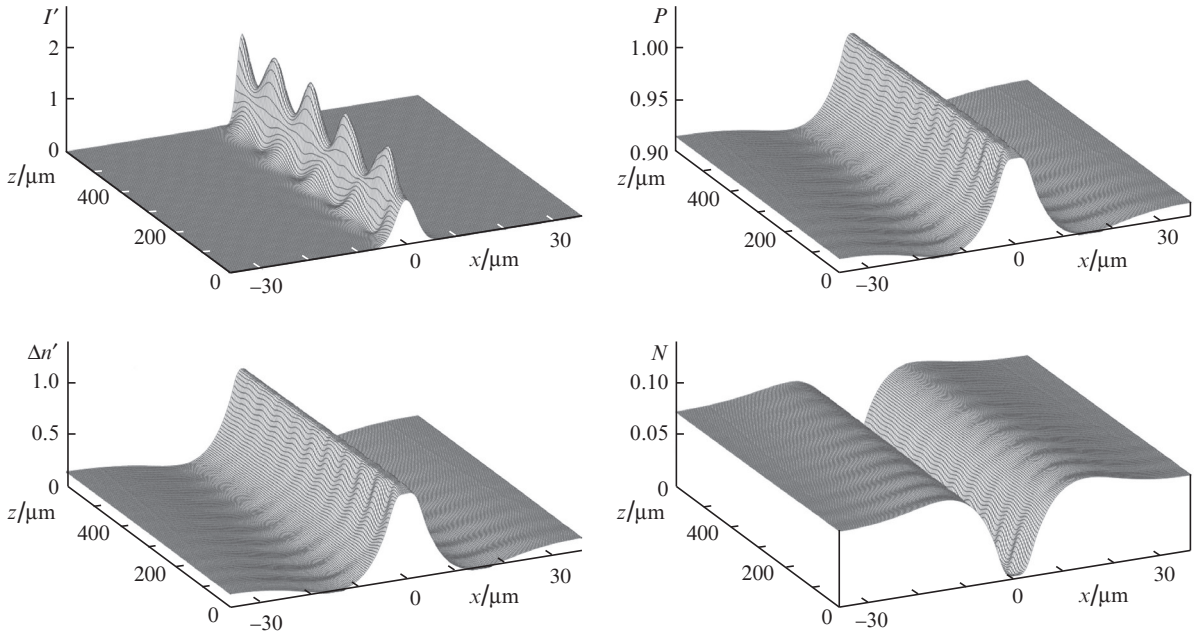


Figure 4. Distributions of the intensity I' of radiation with $\lambda = 0.63 \mu\text{m}$, nonlinear additive $\Delta n'$ to the refractive index, and the mass fractions of polymer and NC after fixing the polymer structure ($t = 10t_p$).

of a monomer into a PPC without the NC ($N_0 = 0$), the refractive index gradients induced by inhomogeneous light field during exposure are ‘erased’, and a homogeneous polymer is formed. The same result is obtained using the NC with $n_N = n_p$, despite the fact that the polymer and NC ($N_0 \neq 0$) concentrations become nonuniformly distributed in the channel at the ultimate PPC conversion. To form a stable waveguiding structure in this way, one must use the NC with a refractive index lower than that of the polymer (Fig. 4).

Another parameter affecting the resulting refractive index profile is the instant at which the fixing background illumination begins, t_b (Fig. 5). For example, at simultaneous ($t_b = 0$) initiation of polymerisation by a Gaussian light beam and by the radiation exposing the entire PPC volume, the refractive index amplitude for even the primary focusing inhomogeneity

in the input plane is so small that cannot focus the incident radiation. The radiation self-trapping conditions are violated in this case, because a long waveguiding channel can be formed in the PPC only provided that the diffraction divergence of narrow light beams is compensated for due to the nonlinear refraction of the medium. To this end, one needs significant refractive index gradients (the ranges $t^* > t_p$ and $t_b \geq t^*$ in Fig.5, to the right and above the $t^* = t_p$ and $t_b = t^*$ lines).

With an increase in the time interval between the instant t^* when the channel formation stops and the instant t_b when fixing begins (Fig. 5), the resulting-distribution width may increase (although insignificantly). Therefore, it is expedient to start fixing the refractive index gradients immediately after forming the structure [curve (3) in Fig. 3; $t_b = t^*$ line in Fig. 5] in order to prevent it from degradation. To avoid increasing

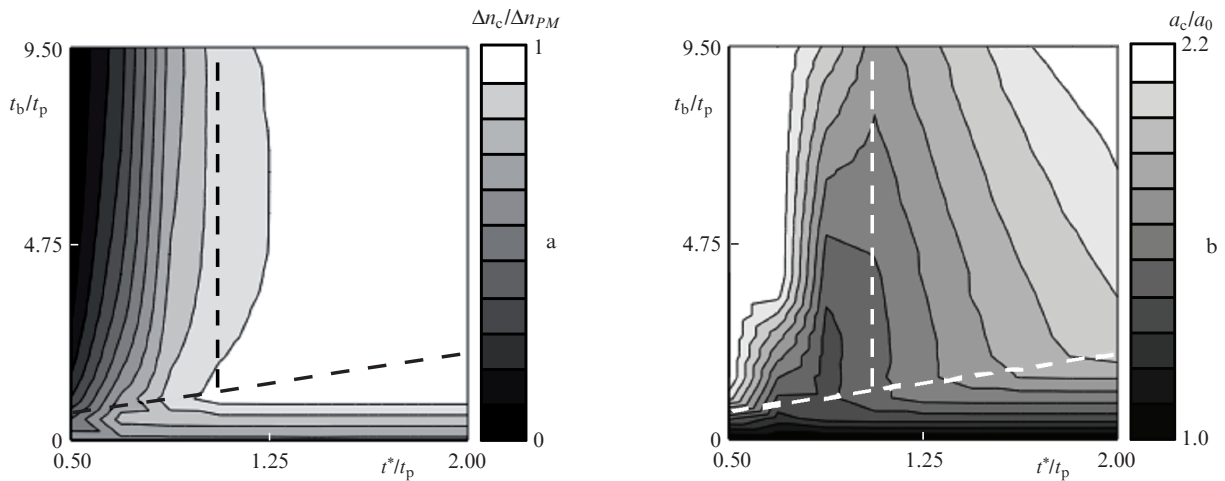


Figure 5. Dependences of the (a) refractive index amplitude Δn_c in the channel and (b) the channel width a_c on the exposure time t^* under a Gaussian light beam and the initial instant t_b of channel fixing.

the channel width [curve (4) in Fig. 3, where additional polymerisation by homogeneous radiation is stopped at 50% PPC conversion], fixing must be performed rapidly ($t_p < t_d$) and continued up to ultimate exhaustion of the monomer. For this fixing technique the resulting amplitude of the refractive index profile in the channel is determined by the difference in the polymer and NC refractive indices, with allowance for the NC amount (note that, when the difference $\Delta n_{PN} = n_P - n_N$ is negative, the refractive index profile in the channel is inverted during fixing):

$$\Delta n_c = (n_P - n_N) N_0. \quad (6)$$

In other words, one can set the refractive index amplitude in the channel by choosing an appropriate refractive index of the NC or changing its amount in the PPC. It is noteworthy that a homogeneous polymer (which does not scatter optical radiation) can be formed only at a limited NC concentration introduced into the PPC (no more than 10%–15%) [16]. Therefore, to obtain a necessary refractive index amplitude in the channel, the NC must have as low as possible refractive index in comparison with the polymer ($n_N < n_P$).

4. Optical connection of optical fibres in a multicomponent PPC

In accordance with the results of analysis of the formation of stable waveguiding structures at light-beam self-trapping in the NC-containing PPC, we considered the possibility of connecting optically standard telecommunication fibres SMF-28 with a core diameter of 8.2 μm in this medium. We investigated the formation of a polymer channel in the gap between the fibres by counterpropagating acting light beams emerging from their end faces and its fixing by ‘homogeneous’ radiation with an intensity $10I_0$. In numerical simulation the input portions of the fibres were taken to be fairly long ($\sim 650 \mu\text{m}$) to establish intrinsic mode regimes for propagating radiation [5, 6].

According to expression (6), it is necessary to introduce $\sim 3.4\%$ NC ($\Delta n_{NM}/n_M = -0.1$) to form a channel with a refractive index amplitude $\Delta n_c = 5.5 \times 10^{-3}$ (this value corresponds to the difference in the refractive indices of the SMF-28 core and active cladding) in the PPC with $\Delta n_{PM}/n_M = 0.01$. The computer simulation showed that, with the aforementioned parameters, the polymer connector formed between the fibre end faces can transfer not only incident radiation but also longer wavelength IR radiation to the fibre with counterpropagating beam (Fig. 6a). Note that an increase in the NC concentration in the PPC (Fig. 6b) does not lead to an increase in the light transfer efficiency.

The experimental studies on the formation of stable polymer connectors were performed with the PPC based on oligo(carbonate methacrylate) (OCM-2), which is characterised by a significant increase in the refractive index (by approximately 1%) at monomer conversion into polymer [11–13]. The individual PPC components (dimethacrylic oligomer, photoinitiating system) and the final polymer have a high transparency in the optical and IR ranges [11, 13]. Due to this, PPC-based fibre-optic connectors can be developed for telecommunication wavelengths (for example, in the ranges near 0.9, 1.1, 1.3, and 1.55 μm) [6]. Different alcohols and organic solvents with a low viscosity and a low (as compared with the polymer) refractive index can be used for this composition as an NC that forms a true solution with the monomer. We chose acetonitrile, which is transparent even in the IR range.

The exposure characteristic of the composition used has the following parameters in the intensity range of 5–50 mW mm^{-2} : $\gamma = 3$ and $H_0 = 30 \text{ mW s mm}^{-2}$. The red boundary of the composition photosensitivity is 0.65 μm . Therefore, a polymer channel was formed using radiation of standard semiconductor laser modules with a wavelength $\lambda = 0.63 \mu\text{m}$ and an output power of 5 mW. The lasers were used in the pulsed regime, due to which we could measure the visible light power transmitted through the polymer connection

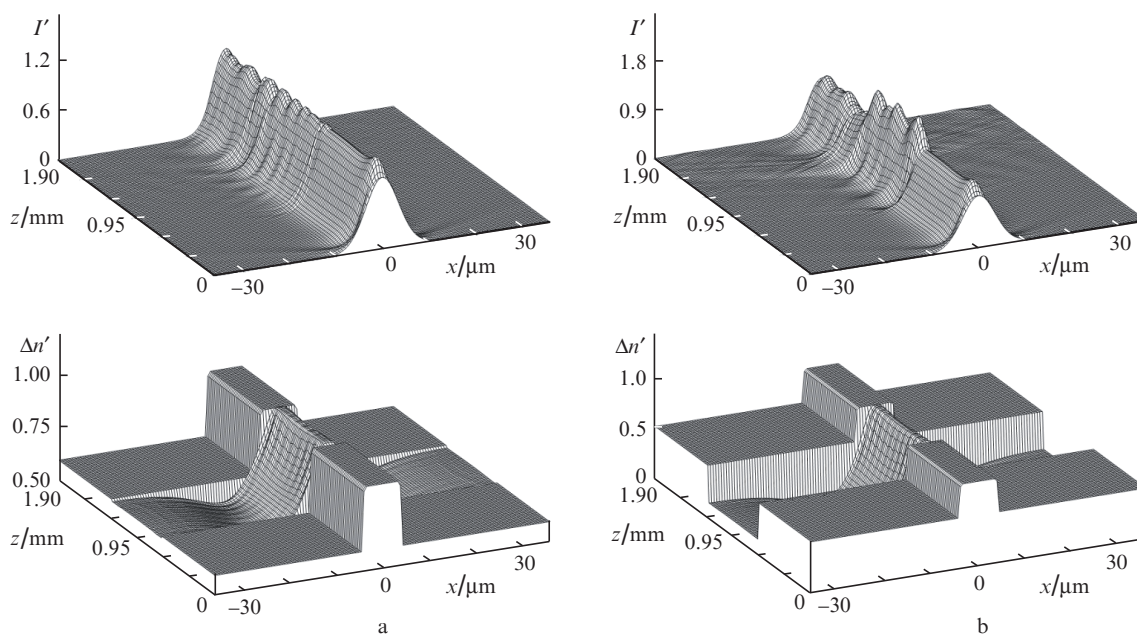


Figure 6. Distributions of the intensity I' of IR radiation with $\lambda = 1.55 \mu\text{m}$ and the nonlinear additive $\Delta n'$ to the refractive index for a polymer structure between the end faces of optical fibres at $N_0 =$ (a) 0.034 and (b) 0.07.

during the entire process of connector formation and fixing. To this end, a semitransparent plate was placed between the fibre end face and the laser source. This beam splitter was used to reflect the radiation emerging from the fibre to the surface of a PD-1180 photodetector. The experimental setup also makes it possible to measure the connector efficiency in the IR range (1.55 μm), according to the scheme similar to that described in [5].

The measurement results are in agreement with the computation data (Fig. 7): the light transfer efficiency increases during the connector formation. Intense additional polymerisation stabilises the radiation transfer coefficient, in contrast to the version where the PPC does not contain the NC; therefore, the connector efficiency decreases due to the disappearance of refractive index gradients. This decrease is significant at large distances between the fibre end faces [5, 6].

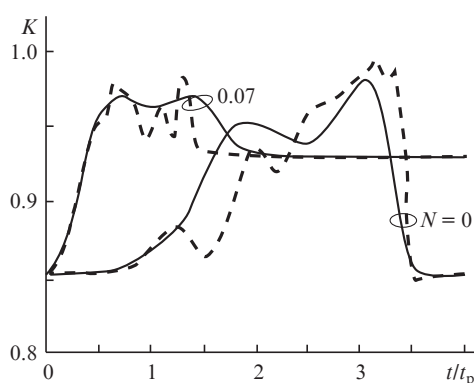


Figure 7. Dynamics of the radiation transfer coefficient K of the polymer connector formed in the PPC with different NC contents N_0 . The solid and dashed lines are the results of the experiment and numerical simulation, respectively.

As an example, Fig. 8 shows a photograph of a fixed polymer structure. Note that the possibility of fixing optically a gradient polymer structure, along with stabilisation of radiation transfer coefficient, solves simultaneously another important problem: mechanical fixation of fibre end faces.

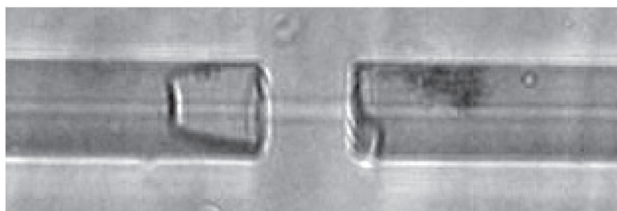


Figure 8. Polymer connector for fibres SMF-28.

5. Conclusions

Introduction of nonpolymerisable additives into the PPC makes it possible not only to implement nonlinear wave processes of light beam self-trapping but also to form stable gradients of polymer and NC concentrations by optical radiation at ultimate PPC conversion. An appropriate choice of the NC optical parameters allows one to synthesise structures with a nonuniform refractive-index distribution. Finally, stable

waveguiding structures can be formed for fibre optics, where desired refractive index gradients can be implemented by specifying the necessary NC amount in the PPC.

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