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Non-local polymerization driven diffusion based model: general dependence of the polymerization rate to the exposure intensity

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Abstract: The nonlocal diffusion model proposed by Sheridan and coworkers has provided a useful interpretation of the nature of grating formation inside photopolymer materials. This model accounts for some important experimental facts, such as the cut-off of diffraction efficiency for high spatial frequencies. In this article we examine the predictions of the model in the case of a general dependence of the polymerisation rate with respect to the intensity pattern. The effects of this dependence on the different harmonic components of the polymerisation concentration will be investigated. The influence of the visibility on the different harmonic components will also be studied. These effects are compared to the effects of varying R_D and σ_D .

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1. Introduction

Photopolymers are attractive materials for the production of high quality holograms [1-3]. Recently several models have been used to describe the mechanism of hologram formation inside photopolymer materials [4-14]. Since the first paper by Zhao and Mouroulis [5] it has been clear that not only polymerization but also diffusion play an important role in the formation of the diffraction gratings. This first article proposed by Zhao *et al.* provides the basis for polymerization driven diffusion (PDD) models of hologram formation in photopolymers. The monomer concentration was expanded into its Fourier series and a set of coupled differential equations was solved for the different harmonic components of the monomer concentration. The different harmonic components of the polymer concentration were obtained by integrating with time the harmonic components of the monomer concentration. Finally, the harmonic components of the refractive index were assumed to be proportional to the harmonic components of the polymer concentration. The general procedure developed by Zhao *et al.* has been used by different authors, by introducing a Fick's law diffusion term in the equation describing the variation with time of the monomer concentration. The use of a Fourier expansion of the monomer concentration has also been a feature several models proposed in the field.

Although the original model proposed by Zhao *et al.* was useful, there were some observed effects that the model did not include. For instance, although for high times of exposure the harmonic components of the monomer concentration tend to zero, these components have non-negligible values for low times of exposure. Since the refractive index of the monomer is different to that of the polymer, the existence of monomer in the material for low exposure times influence the dynamical refractive index of the medium. Thus, for low exposure times the contribution of the monomer concentration to the refractive index should be included. This was studied by Aubrecht *et al.* [8], who proposed a modified version of Zhao's model. This contribution was also later included in a first harmonic PDD model by Neipp *et al.* [14]

Another important fact that the Zhao models had failed to explain was the cut-off of the diffraction efficiency for high spatial frequencies. This was successfully achieved by Sheridan and co-workers, who implemented a non-local polymerization driven diffusion (NPDD) [11-13]. The main feature of this model is that it includes a non-local response function that takes into account the growth of polymer chains inside the photopolymer material. The model has also the particularity that for the limiting case in which the non-local effects are disregarded the model reduces exactly to the PDD proposed by Zhao *et al.*

Previous models assumed that the polymerization rate, that is the rate of conversion of monomer in polymer by photopolymerization, had a linear dependence to the intensity pattern stored in the photopolymer material. In an article by Kwon *et al.* [9] this assumption was modified and a dependence on the square root of the interference pattern was proposed. For instance, Sheridan and co-workers modified their previous model to include a general

dependence [13] and in a very recent paper by S. Wu *et al.* [15], in which the non-local diffusion equation is solved by using a finite-difference method, a dependence of the polymerisation rate on the square root of the intensity pattern was assumed.

Zhao *et al.* also examined a more general dependence [16]:

$$F(x, t) = F_0 [1 + V \cos(Kx)]^\gamma \quad (1)$$

They studied the influence of γ on the different harmonic components of the polymer concentration. In this work we present a study of the effect of γ on the NPDD model proposed by Sheridan *et al.* The influence of the fringe visibility on the different harmonics of the polymerisation concentration will also be analysed and compared to the effect of R_D and σ_D .

2. The general non-local diffusion model

The one-dimensional non-local response diffusion equation, which governs the change of monomer concentration, $u(x, t)$, with time is [11]:

$$\frac{\partial u(x, t)}{\partial t} = \frac{\partial}{\partial x} \left[D(x, t) \frac{\partial u(x, t)}{\partial x} \right] - \int_{-\infty}^{+\infty} G(x, x') F(x', t) u(x', t) dx' \quad (2)$$

In this equation $D(x, t)$ is the diffusion constant, $G(x, x')$ is the non-local response function and $F(x', t')$ represents the rate of polymerisation at point x' and time t' .

The monomer concentration is assumed to be of the form:

$$u(x, t) = \sum_{i=0}^{\infty} u_i(t) \cos(iKx) \quad (3)$$

while the non-local response function $G(x, x')$ will be supposed to have a Gaussian form:

$$G(x, x') = \frac{\exp[-(x - x')^2 / 2\sigma]}{\sqrt{2\pi\sigma}} \quad (4)$$

The square root of the variance $\sqrt{\sigma}$ characterizes the length scale over which the non-local effect is significant.

In this work we will assume the following general dependence for the polymerization rate:

$$F(x, t) = F_0 [1 + V \cos(Kx)]^\gamma \quad (5)$$

where

$$F_0 = \kappa I_0^\gamma \quad (6)$$

And κ is a fixed constant.

The rate of polymerization will now be supposed to have the following series expansion:

$$F(x, t) = F_0 \sum_{i=0}^{\infty} f_i \cos(iKx) \quad (7)$$

The diffusion constant will also be expanded in a Fourier series as:

$$D(x, t) = \sum_{i=0}^{\infty} D_i(t) \cos(iKx) \quad (8)$$

As it has been done in previous papers [5, 11-13] we will initially only retained the first two harmonic components of the Fourier series.

$$D(x, t) = D_0(t) + D_1(t) \cos(Kx) \quad (9)$$

To calculate the two harmonic components of this expansion we will determine the values of the maximum and minimum diffusion coefficients in the polymer, D_{max} and D_{min} respectively. Therefore D_0 and D_1 can be calculated as [5]:

$$D_0 = (D_{max} + D_{min}) / 2 \quad (10)$$

$$D_1 = (D_{max} - D_{min}) / 2 \quad (11)$$

Since the diffusion coefficient will also be considered to decay with time it has the following dependence:

$$D(x,t) = D \exp[-\alpha F_0 t \{1 + V \cos(Kx)\}^\gamma] \quad (12)$$

So

$$D_{\max} = D \exp[-\alpha F_0 t (1 - V)^\gamma] \quad (13)$$

and

$$D_{\min} = D \exp[-\alpha F_0 t (1 + V)^\gamma] \quad (14)$$

By using Eqs. (13, 14) and (10, 11) the final expression of the diffusion coefficient can be calculated as:

$$D(x,t) = D \exp[-\alpha F_0 t \{ (1 - V)^\gamma + (1 + V)^\gamma \} / 2] \\ \times \left\{ \cosh[\alpha F_0 t \{ (1 + V)^\gamma - (1 - V)^\gamma \} / 2] \right. \\ \left. - \sinh[\alpha F_0 t \{ (1 + V)^\gamma - (1 - V)^\gamma \} / 2] \cos(Kx) \right\} \quad (15)$$

If Eqs. (7) and (15) are substituted in Eq. (2) the more general NPDD model will be obtained.

3. Dimension-less equations

As has been done in a previous paper by S. Wu et al. [15] it is interesting to set Eq. (2) in dimension-less form.

The same definitions as those in Ref. [15] will be taken:

$$x_D = Kx \quad (16)$$

$$t_D = F_0 t \quad (17)$$

$$R_D = DK^2 / F_0 \quad (18)$$

$$\sigma_D = K^2 \sigma \quad (19)$$

R_D is a parameter that measures the relative strength between the mechanisms of diffusion and polymerization. If we define τ_D as $\tau_D = 1/DK^2$ and τ_p as $1/F_0$. τ_D and τ_p express the characteristic time of the mechanisms of diffusion and polymerization, respectively. Therefore for values of $R_D > 1$ diffusion dominates over polymerization, whereas for values of $R_D < 1$ polymerization is the dominant process. On the other hand σ_D is a parameter that controls the non-locality effects in the dimensionless non-local response function $G_D(x_D, x_D')$ defined as:

$$G_D(x_D, x_D') = \frac{\exp[-(x_D - x_D')^2 / 2\sigma_D]}{\sqrt{2\pi\sigma_D}} \quad (20)$$

In dimension-less variables, Eq. (2) takes the form:

$$\frac{\partial u(x_D, t_D)}{\partial t_D} = R_D \frac{\partial}{\partial x_D} \left[D_D(x_D, t_D) \frac{\partial u(x_D, t_D)}{\partial x_D} \right] \\ - \int_{-\infty}^{+\infty} G_D(x_D, x_D') F_D(x_D', t_D) u(x_D', t_D) dx_D' \quad (21)$$

The non-dimensional polymerization rate is expressed as:

$$F_D(x_D, t_D) = \sum_{i=0}^{\infty} f_i \cos(ix_D) \quad (22)$$

The non-dimensional diffusion constant now has the form:

$$D_D(x_D, t_D) = \exp[-\alpha_D \{ (1 - V)^\gamma + (1 + V)^\gamma \} / 2] \\ \times \left\{ \cosh[\alpha_D \{ (1 + V)^\gamma - (1 - V)^\gamma \} / 2] \right. \\ \left. - \sinh[\alpha_D \{ (1 + V)^\gamma - (1 - V)^\gamma \} / 2] \cos(x_D) \right\} \quad (23)$$

Finally, by substituting the Fourier expansion of Eq. (22) in Eq. (21) and retaining only, the first four harmonic terms the following four dimensionless coupled monomer rate equations can be found as:

$$\frac{du_0(t_D)}{dt_D} = -f_0 u_0(t_D) - \frac{1}{2} [f_1 u_1(t_D) + f_2 u_2(t_D) + f_3 u_3(t_D)] \quad (24)$$

$$\begin{aligned} \frac{du_1(t_D)}{dt_D} = & -R\text{Ch}[t_D]u_1(t_D) - R\text{Sh}[t_D]u_2(t_D) - S_1 \left[f_1 u_0(t_D) + \left(f_0 + \frac{f_2}{2} \right) u_1(t_D) \right. \\ & \left. + \frac{1}{2} (f_1 + f_3) u_2(t_D) + \frac{1}{2} (f_2 + f_4) u_3(t_D) \right] \end{aligned} \quad (25)$$

$$\begin{aligned} \frac{du_2(t_D)}{dt_D} = & -4R\text{Ch}[t_D]u_2(t_D) + R\text{Sh}[t_D][u_1(t_D) + 3u_3(t_D)] \\ & - S_2 \left[f_2 u_0(t_D) + \frac{1}{2} (f_0 + f_2) u_1(t_D) + \left(f_0 + \frac{f_4}{2} \right) u_2(t_D) + \frac{1}{2} (f_1 + f_5) u_3(t_D) \right] \end{aligned} \quad (26)$$

$$\begin{aligned} \frac{du_3(t_D)}{dt_D} = & -9R\text{Ch}[t_D]u_3(t_D) + 3R\text{Sh}[t_D]u_2(t_D) - S_3 \left[f_3 u_0(t_D) + \frac{1}{2} (f_2 + f_4) u_1(t_D) \right. \\ & \left. + \frac{1}{2} (f_1 + f_5) u_2(t_D) + \left(f_0 + \frac{f_6}{2} \right) u_3(t_D) \right] \end{aligned} \quad (27)$$

With the following definitions:

$$\text{Ch}[t_D] = \exp[-\alpha t_D \{(1-V)^{\gamma} + (1+V)^{\gamma}\}/2] \times \cosh[\alpha t_D \{(1-V)^{\gamma} + (1+V)^{\gamma}\}/2] \quad (28)$$

$$\text{Sh}[t_D] = \exp[-\alpha t_D \{(1-V)^{\gamma} + (1+V)^{\gamma}\}/2] \times \sinh[\alpha t_D \{(1-V)^{\gamma} + (1+V)^{\gamma}\}/2] \quad (29)$$

$$S_i = \exp(-i^2 \sigma_D / 2) \quad (30)$$

The resulting concentration of polymerised monomer can be obtained as:

$$N(x_D, t_D) = \int_{-\infty}^{\infty} G_D(x_D, x_D') F_D(x_D', t_D) u(x_D', t_D) dx_D' \quad (31)$$

therefore the four orders of the polymerisation spatial-harmonic components are:

$$N_0(t_D) = \int_0^{t_D} [f_0 u_0(t_D) + \frac{1}{2} \{f_1 u_1(t_D) + f_2 u_2(t_D) + f_3 u_3(t_D)\}] dt_D \quad (32)$$

$$\begin{aligned} N_1(t_D) = & \int_0^{t_D} S_1 [f_1 u_0(t_D) + (f_0 + f_2 / 2) u_1(t_D) \\ & + 1/2 (f_1 + f_3) u_2(t_D) + 1/2 (f_2 + f_4) u_3(t_D)] dt_D \end{aligned} \quad (33)$$

$$\begin{aligned} N_2(t_D) = & \int_0^{t_D} S_2 [f_2 u_0(t_D) + 1/2 (f_0 + f_2) u_1(t_D) \\ & + (f_0 + f_4 / 2) u_2(t_D) + 1/2 (f_1 + f_5) u_3(t_D)] dt_D \end{aligned} \quad (34)$$

$$\begin{aligned} N_3(t_D) = & \int_0^{t_D} S_3 [f_3 u_0(t_D) + 1/2 (f_2 + f_4) u_1(t_D) \\ & + 1/2 (f_1 + f_5) u_2(t_D) + (f_0 + f_6 / 2) u_3(t_D)] dt_D \end{aligned} \quad (35)$$

Finally the refractive index of the polymeric material can be obtained by taking into account the contributions of the polymer and monomer concentrations in the following way:

$$n(x_D, t_D) = n_{av} + C_P \sum_{i=0}^3 N_i(t_D) \cos(ixD) + C_M \sum_{i=0}^3 u_i(t_D) \cos(ixD) \quad (36)$$

4. Numerical results and discussion

In this section, numerical results about the influence of the dimensionless variables on the polymerisation characteristics of the recorded holograms are presented.

Figure 1 shows the first order of the polymer and monomer concentrations as functions of the dimensionless time, t_D , for different values of the visibility, V , and γ . It can be seen that the effect of decreasing the values of γ is to diminish the value of the first harmonic components of the polymer and monomer concentration. This is due to the fact that for lower values of the parameter γ , the nonlinearity of the pattern stored in the hologram increases. This is more clearly seen in Fig. 2, where the second and third harmonic components of the polymer concentration are represented as a function of the dimension-less time t_D for different values of the visibility, V , and γ . The importance of these terms increases as γ decreases. The effect of the visibility on the harmonics in the Fourier expansion of the polymer concentration is different. In general, a decrease in the visibility implies a decrease in the interference pattern stored in the hologram. Therefore, a decrease of V is accompanied by a decrease in the values of the harmonic components higher than zero of the polymerisation and monomer concentration.

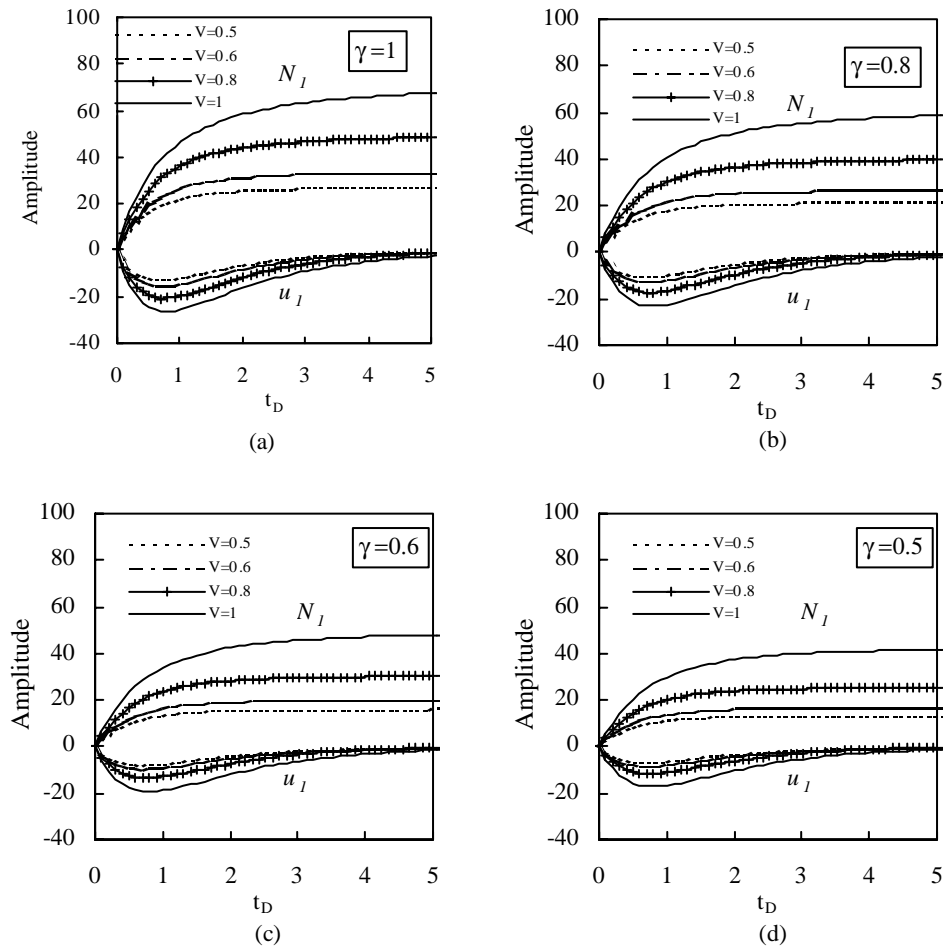


Fig. 1. First harmonic components of the polymer, N_I and monomer, u_I , concentrations as a function of the non-dimensional time, t_D for different values of γ 0.5, 0.6, 0.8, 1 and for different values of the visibility, V : 0.5, 0.6, 0.8, 1.

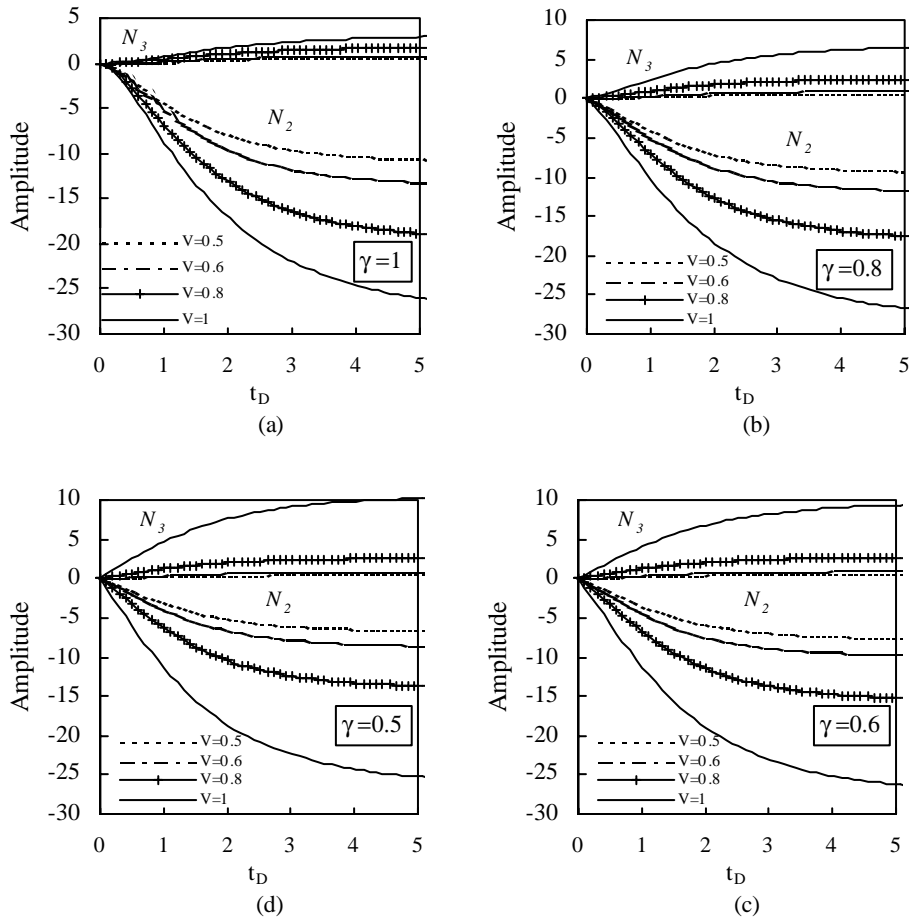


Fig. 2. Second and third harmonic components of the polymer concentration, N_i as a function of the non-dimensional time, t_D for different values of γ 0.5, 0.6, 0.8, 1 and for different values of the visibility, V : 0.5, 0.6, 0.8, 1.

It is also interesting to study the final patterns stored in the holograms. To do this Eqs. (24)-(27) and (32)-(35) were solved for a high value of the parameter t_D to allow the steady states of all the harmonic components of the polymer and monomer concentration to be reached. From Figs. 1 and 2 it is reasonable that a value of $t_D=20$ is sufficient for this purpose.

Figure 3 shows the polymer distribution as a function of the normalized distance, x_D , for different values of γ and R_D . As can be seen in the figure, the effect of R_D in the final pattern stored is clear. The higher the values of R_D , the more the polymer distribution resembles a sinusoidal pattern. This result holds for the different values of γ although the effect of a decreasing γ contributes to increase the nonlinearity of the pattern. It is noticeable that the influence of the parameter R_D on the characteristics of the final pattern of polymer distribution inside the polymer is more critical than the effect of γ . To more clearly support this conclusion in Fig. 4 the absolute value of ratio of the second to the first harmonic component of the polymer concentration as a function of R_D is represented for different values of σ_D for a fixed value of $\gamma=1$. For high values of R_D ($R_D > 1$) the influence of the second harmonic component of the polymer concentration decreases, which is in agreement with references [13,15]. It can also be appreciated that higher values of σ_D also imply that the nonlinearity is reduced, an effect which is also noted in [15]. The same effects may be observed if the absolute value of

the ratio of the third to the first harmonic component of the polymer concentration as a function of R_D is represented, Fig. 5. Again, high values of R_D are accompanied by very low values of the ratio N_3/N_1 . And even for a value of the parameter $\sigma_D=1$ the value of the third harmonic component can be disregarded with respect to the value of N_1 .

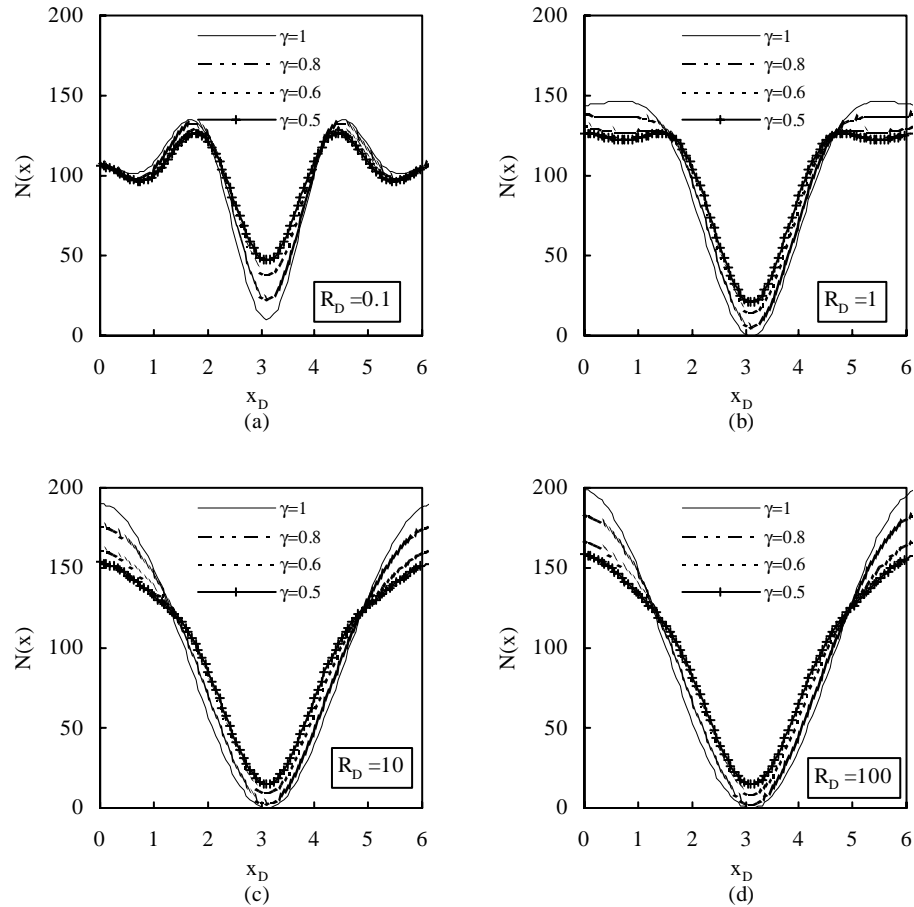


Fig. 3. Polymer concentration as a function of the non-dimensional space, x_D , for different values of the parameter R_D : 0.1, 1, 10, 100 and for different values of γ : 0.5, 0.6, 0.8, 1. $t_D=20$.

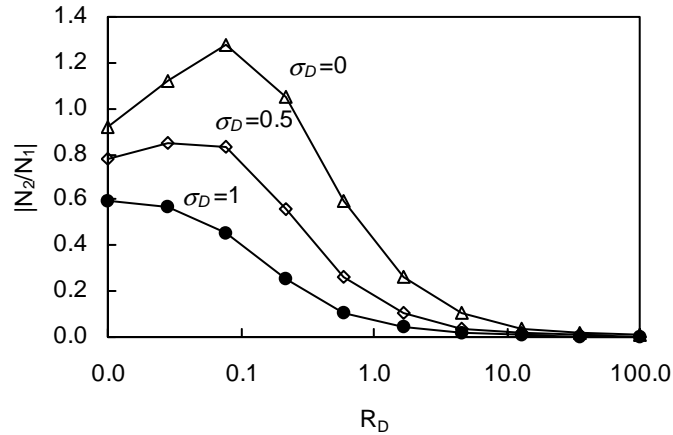


Fig. 4. Ratio of the second to the first harmonic components of the polymer concentration as a function of R_D for three different values of σ_D : 0, 0.5, 0.1. $\gamma=1$, $\alpha=0$, $t_D=20$.

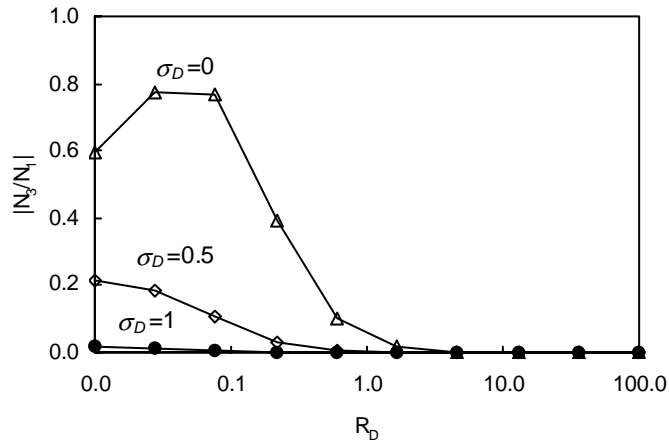


Fig. 5. Ratio of the third to the first harmonic components of the polymer concentration as a function of R_D for three different values of σ_D : 0, 0.5, 0.1. $\gamma=1$, $\alpha=0$, $t_D=20$.

The influence of γ on the ratios N_2/N_1 and N_3/N_1 has also been investigated. Figure 6 shows the absolute value of ratio N_2/N_1 as a function of the parameter γ for different values of σ_D with a fixed value of $R_D=1$. In this case, the dependence of the ratio N_2/N_1 resembles straight lines for all values of σ_D . Although it can be appreciated that for low values of the parameter γ the nonlinearity is increased, the effect of varying γ in the range of $0.5 \leq \gamma \leq 1$ is not as important as is the effect of R_D in the range $0.01 \leq R_D \leq 100$, these ranges assumed for real materials [17]. This can also be seen in Fig. 7 where the absolute value of ratio N_3/N_1 is represented as a function of γ . Nonetheless, in this case, the slope of the curve corresponding to a value of $\sigma_D=0$ is higher than that of Fig. 6. For values of σ_D equal to 1 and 0.5 the value of N_3 can be disregarded with respect to the value of N_1 .

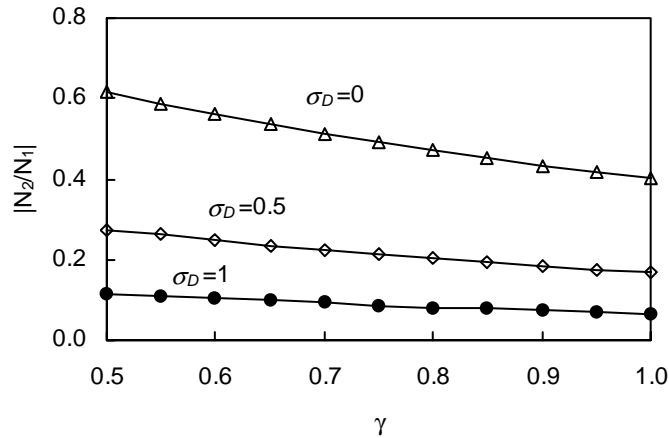


Fig. 6. Ratio of the second to the first harmonic components of the polymer concentration as a function of γ for three different values of σ_D : 0, 0.5, 0.1. $R_D=1$, $\alpha=0$, $t_D=20$.

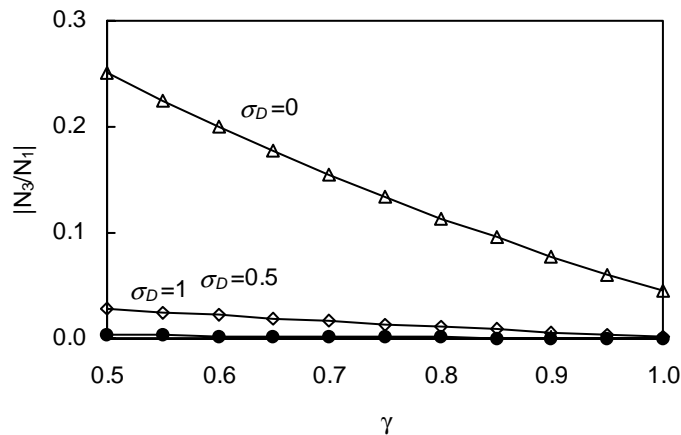


Fig. 7. Ratio of the third to the first harmonic components of the polymer concentration as a function of γ for three different values of σ_D : 0, 0.5, 0.1. $R_D=1$, $\alpha=0$, $t_D=20$.

5. Conclusions

The influence of the parameter γ , which quantifies the nonlinear dependence of the polymerisation rate on the exposure intensity, upon the harmonic components of the polymerisation and monomer concentration has been investigated. Furthermore the effect of the visibility on the different harmonic components of the polymer concentration has also been examined. The effect of decreasing the visibility is basically to reduce the pattern stored in the hologram, with a consequent diminution of the values of all the harmonic components of the polymer concentration higher than zero. It has also been demonstrated that a decreasing value of γ contributes to a decrease in the values of the first harmonic components of the polymer and monomer concentration. In addition the effect of R_D on the distribution profiles of the final polymer concentration stored in the hologram has been examined. It was found that the higher the values of R_D , the more the final polymer distribution resembles a sinusoidal pattern.

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