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Comparison of holographic photopolymer materials by use of analytic nonlocal diffusion models

Feidhlim T. O'Neill, Justin R. Lawrence, and John T. Sheridan

The one-dimensional diffusion equation governing holographic grating formation in photopolymers, which includes both nonlocal material response and generalized dependence of the rate of polymerization on the illuminating intensity, has been previously solved under the two-harmonic expansion assumption. The resulting analytic expressions for the monomer and polymer concentrations have been derived and their ranges of validity tested in comparison with the more accurate numerical four-harmonic case. We used these analytic expressions to carry out a study of experimental results presented in the literature over a 30-year period. Automatic fitting of the data with these formulas allows material parameters, including the nonlocal chain-length variance σ , to be estimated. In this way, (i) a quantitative comparison of different materials can be made, and (ii) a standard form of experimental result presentation is proposed to facilitate such a procedure. © 2002 Optical Society of America

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1. Introduction

It has been proposed¹ that the physical situation governing the formation of holographic gratings in photopolymer can be accurately described by the introduction of a nonlocal response function into the one-dimensional diffusion equation governing grating formation.^{2,3} During exposure, monomer is polymerized, resulting in the formation of polymer chains that grow away from the point at which they are initiated. In adjoining regions the chains grow when the active monomer is removed, producing a smeared version of the recording exposure pattern. If we assume that the chain formation has a general Gaussian probability distribution characteristic,¹ then the extent of the nonlocality, that is, the effects of initiation at one point upon polymerization at another, is quantified by a nonlocal variance parameter σ . When $\sigma = 0^{1-3}$ (the local case), the diffusion-based models explain the existence of a low-frequency cutoff in the spatial-frequency response and the ex-

istence of reciprocity law failure.²⁻⁴ However, no high-frequency cutoff value is predicted to exist. In the nonlocal response regime, $\sigma > 0$, it is found that the larger the nonlocal response variance, the lower the visibility of the profile but also the more closely the profile recorded resembles the illuminating interference pattern. We refer to this model as model I.^{1,5}

A local model allowing for a general relationship between the illuminating intensity and the polymerization rate was developed.⁶ More recently⁷ it has been shown that the change in monomer concentration depends on the square root of the exposing intensity and not on the intensity.¹⁻³ We refer to the corresponding nonlocal model, generalized to incorporate this effect, as model II.⁸ In this formulation, assuming a Gaussian material response function, we derive coupled differential equations, assuming a Fourier-series relationship between the rate of polymerization and the illumination intensity.

For both model I and model II, two-harmonic analytic expressions were compared with the more rigorous four-harmonic numerical results, indicating the ranges of physical parameters for which they are accurate.^{7,8} We compared the analytic expressions by hand with experimental growth curves produced when recording sinusoidal gratings, with different grating spatial frequencies, in an acrylamide-based material.⁹⁻¹² The diffraction intensities measured (corrected to allow for Fresnel reflections¹³ during replay) were converted by use of Kogelnik's first-

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order two-wave coupled-wave theory^{14–16} into the corresponding refractive-index modulation values.

This fitting procedure involved parameter variation and repeated comparison by eye of the predictions of the analytic formulas (corrected to allow for Fresnel reflections during recording) with these growth curves. In this way physical parameters could be estimated. These parameters include the monomer diffusion constant D , the rate of polymerization F_0 , the constant of proportionality between the exposing intensity (or its square root) and the rate of polymerization κ , the constant of proportionality between the polymer concentration and the resulting refractive index C , the dimensionless reaction rate R , and the nonlocal polymer chain-length distribution variance σ .^{1,5,8}

In previous studies we reported that model II appears to provide better fits to the experimental data than model I. Although the predicted values of R and σ that were produced seemed physically reasonable, the procedure was hampered by the inaccuracy of the experimental data and the qualitative nature of the fitting process.

In this paper we carry out the fitting procedure using an automatic numerical data fitting package¹⁷ with the intent of placing the fitting procedure on a quantitative basis. Furthermore, the results of the procedure (both for model I and for model II) applied to experimental data appearing in the literature over a 30-year period are presented. We do this to (a) demonstrate the usefulness of the method and (b) provide a previously unavailable quantitative comparison of the performance of photopolymer-based holographic recording materials.

Several important issues arose during our attempt to identify sufficiently detailed experimental data in the literature to carry out such a comparison. In many cases the form of the published results did not lend themselves to our fitting methodology. Typically, the data presented were either too sparse or necessary technical information was not available. We present a review of some of the papers that we examined, listing the difficulties we encountered. Where possible we present the resulting estimates of the material bulk parameters. We compare the characteristics of these various materials and discuss how photopolymer material performance might be improved. We conclude that only when experimental diffraction efficiency measurements (or refractive-index modulation) are provided as a function of both the spatial frequency and the exposure energy can a meaningful quantitative comparison of material performance be carried out. Using our own experimental data of acrylamide-based material provided in this form, we then reestimate the value of our bulk parameters and compare them with our previous results.

2. Theory

For a detailed derivation of the expressions presented here, we draw the reader's attention to Refs. 1, 5, and 8. The predictions of two distinct models, models I

and II, are examined here, and it is important therefore to distinguish between them. In model I it is assumed that the rate of polymerization is a function of the exposing intensity spatial distribution within the material and is not a function of the exposure time t :

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

where V is the interference fringe visibility inside the recording material and $K = 2\pi/\Lambda$, with Λ the interference fringe period and thus the resulting grating period. In the case of model I, $F_0 \propto I_0$. In model II the rate of polymerization is a function of the square root of the exposing intensity:

$$F(x, t) = F_0[1 + V \cos(Kx)]^{1/2}. \quad (2)$$

In model II, $F_0 \propto I_0^{1/2}$. In this case we represent $F(x, t)$ as a Fourier series:

$$F(x, t) = F_0 \sum_{i=0}^{\infty} f_i \cos(iKx). \quad (3)$$

When $V = 1$,

$$f_0 = \frac{2\sqrt{2}}{\pi}, \quad f_m = \frac{4\sqrt{2}}{\pi} \frac{(-1)^{m+1}}{(-1 + 4m^2)}.$$

The analytic expressions for monomer and polymer concentration are derived on the assumption that the concentrations are well approximated by the 0th and 1st harmonics alone, (u_0, u_1) and (N_1, N_2) . The following first-order coupled differential equations governing monomer concentration can be derived as⁸

$$\frac{du_0(\xi)}{d\xi} = -f_0 u_0(\xi) - f_1 u_1(\xi)/2, \quad (4a)$$

$$\frac{du_1(\xi)}{d\xi} = -S f_1 u_0(\xi) - W u_1(\xi), \quad (4b)$$

with $S = \exp(-K^2\sigma/2)$, $R = DK^2/F_0 = R_0/\Lambda^2$, $W = S(f_0 + f_2/2) + R$, and $\xi = F_0 t$. These differential equations are solved with the initial conditions $u_0(0) = 100$ and $u_1(0) = 0$. The equations for the first harmonic of the polymerization concentration spatial distribution can then be found:

$$N_1(\xi) = \frac{400S}{B^2 - (W + f_0)^2} \left\{ R + \exp\left[-\frac{(W + f_0)\xi}{2}\right] \left[\left(\frac{L}{B}\right) \sinh\left(\frac{B\xi}{2}\right) - R \cosh\left(\frac{B\xi}{2}\right) \right] \right\}, \quad (5)$$

where

$$B = [(W - f_0)^2 + 2f_1^2 S]^{1/2} \text{ and } L = (f_0 - R)R + [-f_1^2 + (f_0 + f_2)(2f_0 - R)]S.$$

It is assumed that the modulation of the refractive index induced during recording is linearly related to the polymer concentration. Therefore the ampli-

tude of the refractive-index modulation recorded in the material is

$$n_1 = CN_1(\xi). \quad (6)$$

Furthermore, we assume that the on-Bragg diffraction efficiency of the unslanted volume transmission gratings examined is well approximated by Kogelnik's two-wave coupled-wave model^{14–16}:

$$\eta = \sin^2\left(\frac{\pi n_1 d}{\lambda_B \cos \theta_B}\right), \quad (7)$$

where d is the grating thickness, θ_B is the Bragg angle of incidence, λ_B is the corresponding Bragg replay wavelength, and TE replay polarization is assumed. Substituting for n_1 from Eq. (6), the diffraction efficiency of a thick volume grating can then be predicted.

3. Review of the Literature

The results discussed here include studies appearing in both reviewed journals and conference proceedings over a 30-year period. Although our literature search is certainly not complete, we do believe that the papers discussed are sufficiently representative for our purposes.

Unfortunately, our attempts to fit the data presented in the literature were hindered by a lack of sufficiently detailed technical information. If the information was available, or if relatively few simple assumptions were made, the fitting procedure itself proceeded with only minor problems. Common difficulties encountered when we attempted to fit data from the literature include the following:

- (1) No spatial-frequency response curves were available.
- (2) Spatial-frequency response data were provided for high spatial frequencies but no low-spatial-frequency data were presented.
- (3) Spatial-frequency response data were provided for low spatial frequencies but no high-spatial-frequency data were presented.
- (4) No growth curve data were presented.
- (5) Not stated whether data were corrected to allow for optical power losses that are due to Fresnel reflections from the material boundaries.
- (6) Not stated whether the value of the maximum diffraction efficiency given is for an undermodulated or an overmodulated grating.
- (7) Normalized diffraction efficiency values were given without the maximum diffraction efficiency and the corresponding exposure energy being provided.
- (8) It is not explicitly stated which normalized form of efficiency, diffraction selectivity, or diffraction efficiency is being used.
- (9) Polarization of replay beams was not specifically given.
- (10) The values of grating thickness are not available (shrinking and swelling were not discussed).

(11) The recording and replay wavelengths and angles of incidence were not available.

(12) The recording and replay optical powers were not available.

(13) The visibility of the exposure fringe pattern was not defined.

(14) No estimates of experimental errors (error bars) were provided with the data.

It is necessary to have unambiguous experimental information to use Eqs. (5), (6), and (7) to fit the experimental data. For example, σ and R are both functions of the recording spatial frequency. Examination of the models^{1,5,8} have shown that R effectively controls the low-spatial-frequency cutoff value, whereas σ controls the high-spatial-frequency cutoff value. Should the information referred to in points (1), (2), or (3) above be unavailable, increased difficulty in the estimation of R or σ will occur. The availability of only a few data points over a restricted range of parameters also reduces the chance of one extracting a meaningful fit. Furthermore, any uncertainty in the exposure energy used to produce a particular diffraction efficiency (or in the value of the diffraction efficiency itself) will lead to uncertainty in the values of n_1 and the rate of polymerization F_0 (and so ξ). Points (4)–(8) all lead to such ambiguity. If the exact recording and replay conditions are not specified, further difficulties arise in the interpretation of the data; see points (9)–(13). Finally, the reproducibility of the experimental data (errors) determines the meaningfulness of the fit achieved and ultimately determines the range of validity of the parameter estimates, which provide satisfactory fits to the data. In fitting the experimental data from the literature, in general, we had to make some assumptions when interpreting the data, for example, in the simplest cases, assuming use of TE polarization during replay or unit visibility during recording.

In Table 1 we provide a chronological listing of some of the papers examined. This subset was chosen to provide an overview of the materials reported on and also to illustrate some of the common difficulties encountered. Table 1 is divided into three sections, each containing three columns. In the first section we identify the paper, the photopolymer material, and the particular difficulties we identified in attempting to analyze the results presented. In the second and third sections our estimates of the bulk parameters found using model I and model II are presented where possible. The fitting procedure and results are discussed in Section 4.

4. Results

To illustrate our typical parameter extraction procedure, we start with data, extracted by hand, from Fig. 11 in Ref. 19. This is one of the earliest and most complete sets of data available of which we are aware. Significantly, the paper reports research on a recent ancestor of the popular DuPont family of commercial materials. Unfortunately little detailed information regarding the chemical composition of the material is

Table 1. Papers Examined and the Resulting Predictions

Reference	Material	Difficulty	Model 1			Model 2		
			R	$\sqrt{\sigma}$ (nm)	C (10^{-5}) (l/mol)	R ($\times 10^{-12}$)	$\sqrt{\sigma}$ (nm)	C (10^{-5}) (l/mol)
Colburn and Haines ¹⁸ (1971)	DuPont photopolymer	(10), (13), (14)	12.237	84.1	2.844	8.926	84.6	2.058
Booth ¹⁹ (1975)	DuPont photopolymer	(5), (14)	140.582	47.6	4.517	184.657	47.6	6.777
Calixto ²⁰ (1987)	Acrylamide-based photopolymer for red region	(1), (5), (8), (9), (11), (14)	—	—	—	—	—	—
Carre <i>et al.</i> ²¹ (1992)	Photopolymer for green region	(2), (10), (14)	3332.772 ^a	57.6	3.389	33,332.597 ^a	57.6	5.083
Loungnot and Turck ²² (1992)	Photopolymer for green region	(1), (5), (14)	—	—	—	—	—	—
Fimia <i>et al.</i> ²³ (1993)	Photopolymer based on a mixture of (HEMA ^c) and (EGDMA ^c)	(2), (5), (8), (14)	25.700	95.4	4.178	33.796	95.4	6.268
Gambogi <i>et al.</i> ²⁴ (1993)	DuPont photopolymer	(3), (5), (6), (8), (13), (14)	1.037	0	65.403	1.374	0	97.969
Martin <i>et al.</i> ²⁵ (1997)	Acrylamide based	(5), (6), (8), (14)	5.624	86.5	4.703	7.466	86.4	7.045
Aubrecht <i>et al.</i> ²⁶ (1998)	DuPont photopolymer HRS-150	(1), (5), (14)	—	—	—	—	—	—
Blaya <i>et al.</i> ²⁷ (1998)	Acrylamide-based photopolymer	(1), (5), (14)	—	—	—	—	—	—
Zhao <i>et al.</i> ²⁸ (1998)	Photopolymer for red region	(1), (5), (8), (14)	—	—	—	—	—	—
Kim <i>et al.</i> ²⁹ (1999)	Polyacrylamide/PVA ^d film for red region	(1), (5), (9), (13), (14)	—	—	—	—	—	—
Lawrence <i>et al.</i> ¹² (2001), see Table 3	Acrylamide including inhibitor	(2)	3333.268 ^a	68.0	1.960	3333.175 ^a	68.0	2.941
Cheben and Calvo ³⁰ (2001)	Photopolymerizable glass	(1), (5), (14)	—	—	—	—	—	—
Garcia <i>et al.</i> ³¹ (2001)	Acrylamide-based photopolymer	(1), (5), (8), (14)	—	—	—	—	—	—

^aIndicates values of R_0 that are too large (unphysical).

^bHydroxyethylmethacrylate.

^cEthylenglycoldimethacrylate.

^dPolyvinyl alcohol.

Table 2. Numerical Search and Predicted Parameter Fit Values from Ref. 19

Ranges Examined	Model I	Model II
$10^{-4} \leq C \leq 10^{-2}$	$C = 4.517 \times 10^{-5} \text{ l/mol}$	$C = 6.776 \times 10^{-5} \text{ l/mol}$
$10^{-20} \leq \sigma \leq 10^{-18}$	$\sigma = 2.273 \times 10^{-15} \text{ m}^2$	$\sigma = 2.273 \times 10^{-15} \text{ m}^2$
$10^{-13} \leq R_0 \leq 10^{-8}$	$R_0 = 140.581 \times 10^{-12} \text{ m}^2$	$R_0 = 184.657 \times 10^{-12} \text{ m}^2$
$10^{-5} \leq C \leq 10^{-1}$	$C = 4.517 \times 10^{-5} \text{ l/mol}$	$C = 6.777 \times 10^{-5} \text{ l/mol}$
$10^{-18} \leq \sigma \leq 10^{-14}$	$\sigma = 2.273 \times 10^{-15} \text{ m}^2$	$\sigma = 2.273 \times 10^{-15} \text{ m}^2$
$10^{-14} \leq R_0 \leq 10^{-11}$	$R_0 = 140.582 \times 10^{-12} \text{ m}^2$	$R_0 = 184.657 \times 10^{-12} \text{ m}^2$
$10^{-15} \leq C \leq 10^{-3}$	$C = 4.517 \times 10^{-5} \text{ l/mol}$	$C = 6.777 \times 10^{-5} \text{ l/mol}$
$10^{-22} \leq \sigma \leq 10^{-15}$	$\sigma = 2.27317 \times 10^{-15} \text{ m}^2$	$\sigma = 2.273 \times 10^{-15} \text{ m}^2$
$10^{-14} \leq R_0 \leq 10^{-9}$	$R_0 = 140.582 \times 10^{-12} \text{ m}^2$	$R_0 = 184.657 \times 10^{-12} \text{ m}^2$

available in the literature. In fitting the diffraction efficiency data, we assumed it has been corrected for boundary reflections. The author states that a recording wavelength of 514.5 nm was used, the fringe visibility was $V = 1$, and the replay wavelength was 633 nm. The replay polarization is not specified. The grating thickness is given as 50 μm ; shrinking or swelling effects are not discussed.²⁶ We assume, because it is not specified, that the diffraction efficiency values given are for long exposures sufficient to remove all monomer. Therefore, based on our understanding of the models,^{1,5,8} we assume $\xi = 10$. It should be noted that, if larger values of ξ are used, no significant changes in either the parameter estimates or the estimated variance of the fits were observed. When we varied the parameter ranges over which the numerical search for fits was carried out, three comparably good fits to the data (having similar and low

estimated fit variances) for both of the nonlocal models were found. We note use of the spatial-frequency independent parameter $R_0 = R \Lambda^2$.

In Table 2, results for this material, which are typical of those found for the materials discussed here, are presented. Column 1 gives the ranges specified for the searches. Columns 2 and 3 detail the resulting model I and model II estimates. In all six cases, the estimated fit variances¹⁷ were close to a value of 1.081×10^{-7} . The six fits to the data are shown in Fig. 1. We note that it is impossible to differentiate the fits by eye and that they match the data well. Furthermore, we note that (i) the predicted standard deviation of the chain length is $\sqrt{\sigma} = 47 \text{ nm}$ and (ii) a larger value of both C and R_0 is predicted by model II than by model I. The larger R_0 value indicates that a higher diffusion constant D or a lower rate of polymerization F_0 is being predicted by model II. The larger R_0 value implies that the grating that is formed will more closely resemble the exposing pattern.

In previous papers we hand fitted the growth curve data for an acrylamide-based material using both model I^{1,5} and model II.⁸ The results are summarized in Table 3. We now fit the identical growth curve data using the automatic fitting procedure.¹⁷ The recording geometry is that of an unslanted transmission grating with a spatial frequency of 1250 lines/mm. Numerical search ranges for the parameter values were used during the fitting routine that were similar to those listed in the first column of Table 2. The estimated variance of the fit with model I was typically 2.36×10^{-10} , whereas for model

Fig. 1. Six almost identically good fits to the data presented in Ref. 19 with model I and model II; see Table 1.

Table 3. Acrylamide-Based Material^{7,8}: Predictions of Fits to Growth Curves and Three-Dimensional Data Curves

Model	Fit	C (mol/l)	R_0 (m^2)	$\sqrt{\sigma}$ (nm)	I: $\frac{\kappa}{[\text{m}^2(\text{W/s})]}$ II: $\frac{\kappa}{[\text{m}(\text{W}^{1/2}/\text{s})]}$
I	By hand ⁷	2.1×10^{-5}	3×10^{-12}	80 ± 25	0.06
	Numeric	2.1×10^{-5}	1.25×10^{-12}	105	0.04
II	By hand ⁸	1.9×10^{-5}	3×10^{-12}	76 ± 25	0.035
	Numeric	3.1×10^{-5}	1.2×10^{-12}	91	0.04
I, 3-D fit	Numeric	2.1×10^{-5}	2.4×10^{-12}	91	0.04
II, 3-D fit	Numeric	3.1×10^{-5}	2.9×10^{-12}	90	0.035

Fig. 2. Two fits to the growth curve data discussed in Refs. 7 and 8 with both model I and model II; see Table 3. Spatial frequency is 1250 lines/mm.

II it was 2.76×10^{-10} . The experimental growth curve and the corresponding fits are shown in Fig. 2.

Now we combine a set of growth curves measured at different spatial frequencies to form the three-dimensional (3-D) set of experimental data shown in Fig. 3. We then fit this 3-D data set using the model I and model II analytic expressions and MATHEMATICA.¹⁷ The results are also summarized in Table 3. The resulting parameter predictions, when we use the different forms of data, the two different models, and the different fitting procedures, all appear to have values that differ by a factor of less than 2. A further improvement in the accuracy of the predictions might be expected given the availability of more detailed lower- and higher-spatial-frequency growth curves.

Substituting the parameter value estimates into the model I and model II analytic expressions results in the graphs shown in Figs. 4 and 5, respectively. In both cases, reciprocity law failure²⁻⁴ is clearly predicted at the low spatial frequencies, the drop-off in refractive-index modulation strength occurring with increased exposure (increasing ξ). We can also see

Fig. 3. Schematic 3-D representation of experimental data. Refractive-index modulations are a function of spatial frequency and exposure energy $I_0 t$.

Fig. 4. Model I predicted refractive-index modulation as a function of spatial frequency and exposure energy $I_0 t$.

the drop-off in n_1 as the material response decreases at higher spatial frequencies. Comparing Figs. 4 and 5, it can be seen that the two predictions are not identical. For example, a higher maximum refractive-index modulation is predicted in the model II case despite the fact that a material response to the square root of the illumination is assumed. This difference could be used to compare the qualities of the fits provided by the two models.

In general, a material with both a large R_0 and small σ (localized behavior) provides a good holographic recording medium. This is because such a material can reproduce the exposure pattern more accurately because it provides higher fidelity, recording both low and high spatial frequencies. Comparing our results in Table 3 with those given in Table 2,¹⁹ it can be seen that in these terms the DuPont material¹⁹ is superior to the acrylamide material.^{5,8} Clearly, other criteria, including material cost, availability, sensitivity, exposure wavelength, and stability, would also be of considerable practical significance.

Returning to Table 1, we note that a similar fitting procedure was used in all cases. In several cases (see footnote *a* in Table 1) the R_0 value predicted was large, ~ 3333 . This occurred when insufficient low-spatial-frequency data were available to meaningfully fit the data. We note, however, that in most cases when a fit was attempted, sufficient high-spatial-frequency data were available (or could be

Fig. 5. Model II predicted refractive-index modulation as a function of spatial frequency and exposure energy $I_0 t$.

inferred) to allow an apparently meaningful estimation of σ .

It would appear from the results presented in Table 1 that, because the meaningful estimates of R_0 vary by almost 2 orders of magnitude, much effort has been spent to vary the rate of polymerization and the diffusion constant of various materials. Much less effort appears to have been spent on attempts to control the polymer chain length. If a controlled reduction of σ can be achieved, the potential improvements in high-spatial-frequency response would be significant. We note that, for practical holographic applications, high-spatial-frequency material response, which determines material resolution and data-storage densities, is in general of greater importance than low-spatial-frequency response. To date an unsuccessful attempt to control the chain length, involving use of inhibitors, has been reported by us,¹² and this research is proceeding. The physical significance of the chain-length sizes predicted are discussed elsewhere.^{5,8}

5. Conclusion

In this paper the nonlocal model^{1,5} and its extensions⁸ have been reviewed. On the basis of this model, the derivation of analytic solutions has also been briefly reviewed. Despite the assumptions we made in deriving the model, it appears to offer a simple and physically intuitive explanation of the major characteristics of photopolymer materials.

In Table 1 a partial review of the literature in this area over a 30-year period has been presented. We discussed the difficulties in interpreting the data presented and put forth, where possible, estimates of material parameters. It became clear during this procedure that more detailed data must be provided in the literature if meaningful independent comparisons of materials are to be carried out. In particular, we draw the reader's attention to points (1)–(14) listed in Section 3. Two distinct types of curve fitting to experimental data were carried out: (1) fits to refractive-index modulation growth curves, $n_1(\xi)$, at a single spatial frequency and (2) fits to 3-D refractive-index modulation growth curve data, $n_1(1/\Lambda, \xi)$. In this paper we noted that the 3-D data, when presented in detail, appear to provide sufficient information to estimate the values of the material physical parameters.

In this paper we placed emphasis on examining the DuPont material discussed in Ref. 19 and the acrylamide-based material discussed in Refs. 5 and 8. We indicated that the DuPont material appears to have both shorter polymer chains (and a resulting higher-spatial-frequency material response) and a greater diffusion constant and lower rate of polymerization. It has been noted that, because the chain length governs the high-spatial-frequency material response, further effort should be made to reduce σ .

In an attempt to examine the role of the form of the response function, several other types have been examined elsewhere,³² including power law combined with Gaussian and Chernov–Debye-like responses.

The fundamental properties of the model appear unchanged. Furthermore, extended multicomponent models³³ are being developed that describe photopolymer systems with more than one diffusing molecule.

Material systems based on high-diffraction-efficient photopolymerizable glass³⁰ have been recently examined, to which it would appear that the model may be applicable. Unfortunately, to our knowledge, no detailed spatial-frequency response data are currently available. However, we have been informed³⁴ that the response is flat between 50 and 2000 lines/mm, indicating a large R_0 value.

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