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Material Kinetics during Fabrication of Holographic Gratings in Acrylamide-based Photopolymer

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Abstract: We describe holographic grating formation in Acrylamide-based photopolymer material using the Non-Local Diffusion Driven model & discuss radical suppression leading to an inhibition period before grating growth. Diffusion effects of monomer & polymer are discussed.

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1. Introduction
A more thorough understanding of the Acrylamide-based photopolymer material kinetics during and after holographic recording is necessary to further facilitate the evolution of holographic data storage systems. To this end, experiments are carried out and using the results obtained, values for the diffusion constant are calculated.

The diffusion effects in the photopolymer material during and post-exposure are modeled using the Nonlocal Polymer Driven Diffusion (NPDD) model. Using the value obtained for the diffusion constant, the evolution of the Acrylamide monomer and Polyacrylamide concentration are modeled.

The model accounts for the existence of an inhibition period prior to the onset of diffraction of the replay beam. This period is attributed to the presence of Oxygen within the material, which results in the termination of the Polyacrylamide chains before they grow large enough to cause a significant density variation in the material.

2. Diffusion Constant
The grating is recorded using a standard holographic setup. From the results obtained and using Kogelnik's coupled wave theory\(^1\), a value for \(\alpha\), the decay constant of the grating refractive index modulation is found. From this, using Fick's Law, a value for the diffusion constant is found. The impact of the crosslinker concentration affects the rate of decay of the grating and hence the diffusion constant.

3. NPDD Model
The NPDD model is based on a 1-D diffusion equation utilizing Fick's Law:

\[
\frac{\partial u(x,t)}{\partial t} = \frac{\partial}{\partial x} \left[ D(x,t) \frac{\partial u(x,t)}{\partial x} \right] - \int_{-\infty}^{0} \int_{0}^{\infty} R(x,x',t,t') F(x',t') \times \left[ u(x',t') \right]^{\beta} \, dt' \, dx' - \frac{u(x,t)}{u(x,t) + N(x,t)} \frac{\partial H(x,t)}{\partial t}
\]

where \(u(x,t)\) is the free-monomer concentration, \(D(x,t)\) is the diffusion constant, \(F(x,t)\) is the polymerization rate, \(N(x,t)\) is the polymer concentration, \(R(x,x',t,t')\) is the nonlocal material response function,\(^2\) \(H(x,t)\) is the hole concentration,\(^3\) and \(\beta\) is a factor introduced to specify the dominant chain termination mechanism, either bimolecular (\(\beta=1\)) or primary (\(\beta=2\)).\(^4\) The inclusion of the material response function accounts for the spatial and temporal effects associated with polymer chain growth. The model successfully predicts high spatial frequency cut-off experimentally evident in photopolymers and dark reactions following short exposures. More recently the model has been used to determine an optimum recording schedule for holographic data storage.\(^5\)

4. Inhibition Effects
The ‘dead-band’, which can be seen at the initial stages of grating growth, has been modeled by a step function. Experiments were carried out to observe the effects caused by inhibitors such as oxygen and the inclusion of these effects have been added to our model. Equation 2 shows the polymerization rate with the inclusion of the step-function.

\[
F(x,t) = f_0 \theta \left[ \frac{U_0}{l_d} - \rho(t) \right] \left[ 1 + V \cos(Kx) \right]
\]