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Title	Holography and photopolymer recording materials
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Publication date	2010-04-11
Publication information	Digital Holography and Three-Dimensional Imaging, OSA Technical Digest (CD) (Optical Society of America, 2010)
Conference details	Paper presented at Fundamental Advances in Holography II (DMB), Digital Holography and Three-Dimensional Imaging (DH) conference, Miami, Florida, April 11, 2010
Publisher	Optical Society of America
Link to online version	http://www.opticsinfobase.org/abstract.cfm?URI=DH-2010-DMB1
Item record/more information	http://hdl.handle.net/10197/3377
Publisher's statement	This paper was published in Digital Holography and Three-Dimensional Imaging and is made available as an electronic reprint with the permission of OSA. The paper can be found at the following URL on the OSA website: http://www.opticsinfobase.org/abstract.cfm?uri=DH-2010-DMB1 . Systematic or multiple reproduction or distribution to multiple locations via electronic or other means is prohibited and is subject to penalties under law.

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Holography and Photopolymer Recording Materials

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Abstract: Photopolymers act as drivers and enabler of fabrication technologies for applications including refractive/diffractive optical elements, hybrid 3D optoelectronic circuitry, data storage recoding media and self-trapping. The need for and development of modeling tools is discussed.

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OCIS codes: 090.7330, 090.2900, 050.1940, 160.5335, 160.5470, 300.1030.

1. Introduction

For those of us who started our holographic research careers working with Silver Halide and Dichromated Gelatin, (or even photoresist and photorefractives), our first encounter with photopolymers comes in general as a very pleasant surprise. They are simply very easy to work with. A first year graduate student working in an average optics laboratory can make such materials relatively easily, producing fairly sensitive stable thick layers, and then go on to record high efficiency holograms. Therefore with relatively little effort students can make holograms and impress their friends, and with slightly more effort professors get to present papers at conferences in Miami.

As usual however things happen to spoil the fun. First the professors cannot stop themselves from asking awkward questions like ‘How does the stuff really work?’, and second those who have to sell things in order to make a living want to know ‘How can I make practical use of it?’. It should be emphasised that usually these people manage to do an awful lot of interesting things before the boring professor shows up and complicates things. However we also note that given a chance the professor might be able to save such people a great deal of time, energy and money if the right questions can be formulated and answered.

Over the past 2 years I have been lucky enough to work closely with some very clever people in academia and industry. This paper which is informed by these experiences indicates why I think the modelling of photopolymers is necessary and of practical value. The condensed review of our work below, and the related references [1-20], will allow those interested to access the background information and technical detail impossible to present here.

2. Photopolymers Performance

As stated above photopolymers are relatively convenient to work with, however they are very difficult to describe (model) in a useful, flexible and accessible way. The reasons are their complexity and what we mean when we talk about understanding or describing them. In order to be able to say I understand them I want access to a model, derived from some consistent set of principles, which provides quantitative outputs (predictions) in agreement with reproducible experimental results. In a typical photopolymer material any change in chemical composition may alter several of the parameters governing the photochemistry, polymerisation process or mass transport effects inside the layer volume during and/or post exposure. As discussed in the literature, [1-20], the simplest of these materials exhibit non-latent, non-linear and nonlocal optical responses. Furthermore any change in the form of exposure, the form of the optical replay, the ambient environmental conditions or the cover-plate/layer/substrate combination, may change the optical performance of the device recorded in the exposed material.

So we have a physical problem which must be modelled using a great many physical constants and whose complete behaviours are governed by a large number of physical effects (optical, thermal, chemical, thermodynamic, etc.). Given sufficient motivation we know that we can deal with such problems, albeit with difficulty. Simplifications of the problem should be possible when specific applications, pure constituent materials and highly controlled laboratory conditions are available during layer fabrication and use.

A great deal of effort has over the past 10 years been made in relation to material production control and processing. As noted several practical applications (pictorial holography, data storage, self trapping, photo-embossing, and hybrid 3D optoelectronics circuits) have been identified and motivate this work. There have also been significant advances in developing material models. However to my knowledge no quantitative design of a photopolymer material layer optimised for a particular application has ever taken place. I must again emphasise

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that there are an ever increasing number of well engineered material available, but they have been primarily arrived at through a process of professionally informed development allied with intense iterative experimental trial and error. Until very recently no commercial photopolymer material design process, involving any significant use of a quantitative photopolymer material model had ever taken place simply because no such model existed.

Let me try to make clear what I am saying by directing a few simple questions to those of you who work with photopolymers:

1. If you take the material currently being used and vary the concentration of any constituent component by x%, by what percentage will the diffraction efficiency of a simple holographic grating recorded in that material change?
2. How will changing the exposing intensity affect the percentage change?
3. Which of the following is most important in terms of material performance for your application: The rates of dye recovery, dye bleaching, inhibition, polymer initiation, propagation or termination?
4. How many components diffuse in your material?
5. How do viscosity changes in your material affect the resulting application?

If you have performed many experiments using your particular material you may, at least qualitatively, know the answer to one or two of these questions. However given the very large number of variables involved it is extremely unlikely that you can answer most of them quantitatively, (most certainly not to the satisfaction of an awkward old Irish professor).

Below I highlight some of the problems tackled by us in this area and indicate how we are attempting, step by step, to carefully approach a fuller understanding of these fascinating and highly useful materials.

3. The Modelling Process

The Non-local Polymerization Driven Diffusion model, (NPDD), [1], was proposed by us and is being developed within our group to describe the material behavior of polymer layers which upon illumination undergo free radical based photo-polymerisation [2, 3]. The NPDD was originally developed in order to take into account both spatial and temporal non-local polymer chain formation and growth [1,4]. It includes the effects of monomer diffusion, [1], i.e., the diffusion of monomer inside the dry layer from dark to brightly illuminated regions. It has been shown to be capable of predicting both the low and high spatial frequency drop-offs in the material response, (including reciprocity failure), and the non-linear formation of higher grating harmonics (loss of fidelity) [5]. By including a careful analysis of the contributions to the over all refractive index of the background, monomer and optically generated polymer in the exposed layer the temporal evolution of the gratings, formed both during and post-exposure, in several different materials have been successfully predicted [5, 6, 7]. Material shrinkage has also been modelled by introducing the concept of holes [4,8]. Using the NPDD three dimensional analysis analyses have been developed [9] as have proposals how to chemical improvement material spatial frequency response using a chain transfer agents [10].

The availability of this model, combined with rigorous electromagnetic theory, permits the theoretical study of applications, for by example examining existing scaling laws and empirical figures of merit, placing them on a sounder basis [11-14]. Another is the ability to generate new approximate short exposure (low or pulsed light) models in order to rank the significance of different material parameters for particular applications [15]. Knowledge of the appropriate model (and most significant physical parameters) also drives the development of simple but accurate experiments allowing efficient material layer characterization. In this way we have been able to study different materials, containing different dyes, including commercial products.

Given the overall complexity of the full material system our approach requires that different aspects of the material be examined at different times. In our earliest work [1,3] we assumed a very simple model could be used to describe the absorption of light and the resulting initiation of polymer chains. However we have shown how idealized photo-kinetics break down in relation to inhibition [16], the steady state approximation and dye recovery and bleaching [17-20]. Combining [17-20] with the results in [3,6,7] will allow further generalization and improvements in the physical accuracy of the NPDD model.

Much remains to be done. A listing of many of the current limitations of our models and several proposed avenues of future work are given in [3,7,8]. One such topic, for example, is the development of physically consistent multi-component models. Clearly photopolymer modeling remains a topical area of research. I hope I have supplied sufficient information to demonstrate its' importance and use. Only through detailed analysis will the processes taking place in these materials be understood and thus photopolymers full potential be achieved.

4. Acknowledgements: We acknowledge the support of Enterprise Ireland, Science Foundation Ireland, and the Irish Research Council for Science Engineering, and Technology.

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