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16. Abstract

The task for this report is to perform the basic research and develop a prototype benchtop holographic optical storage system based on photochromic and/or photorefractive polymers so that both permanent and erasable images may be stored and retrieved in the same mixed polymer medium. The task consist of: assembly and setup of the benchtop holographic storage system, including lasers, optics, and other ancillary equipment in a laboratory setting; and research and development of a suitable polymner matrix that will allow practical storage and retrieval of digital data. This will necessitate molecular design of the matrices involved and subsequent physics test to verify the characteristics of the matrices provide practical storage and retrieval.

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Holographic Storage Using Photorefractive Polymers

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Undergraduate Research Assistants: Kristi D. Harris and Rajani Ayachitula

This is the annual report for work performed under contract NAS5-32337 Task 81, "Holographic storage using photorefractive polymers" at UMBC from 06/01/99 to 05/31/00. During this period, we continued to study the feasibility of using photorefractive (PR) and photochromic (PC) polymers as holographic storage media for applications useful to the Earth and Space Data Computing Division (ESCDC) of the NASA/Goddard Space Flight Center and to NASA in general. During the past year, we completed a detailed study of a series of PC materials useful for long term data storage. We also began to study the physical phenomena governing the response time and storage times of the PR materials. The results of the PC study are currently being written up for submission to *Applied Optics*.

The graduate student involved in the work and supported by a GSRP Fellowship, Shane Strutz, graduated with a PhD during the past year (July 1999) and took a position in the Optical Sciences Division at the Naval Research Laboratory. Two undergraduates, Rajani Ayachitula and Kristi Harris, have taken over some of the work that Shane started. To assist us in our studies of these materials and applications for NASA, Ms. Harris competed for and won the prestigious Provost's Undergraduate Research Award. Under this award, entitled, " Investigation of Photorefractive Polymer Composites for Holographic Storage Applications", Ms. Harris will extend our studies of the physical processes limiting the writing time response in PR materials and will also be investigating ways to extend the storage time in PR materials to a few hours so they will be useful for short term storage applications on satellites.

Presentations and Publications

During the past year we delivered/prepared the following publications relating to this work,

"Dual grating photorefractive polymer", L. Michael Hayden and Shane J. Strutz, *Organic Thin Films for Photonic Applications*, OSA Technical Digest Series (Optical Society of America, Washington, DC, 1999) p 93.

"Dual use chromophores for photorefractive and photochemical holographic applications", Kristi D. Harris, Rajani Ayachitula, Shane J. Strutz, Robert J. Twieg, and L. Michael Hayden, *in preparation for Applied Optics.*

We are also submitting the following paper to a conference this summer,

"Dual use chromophores for photorefractive and irreversible photochromic applications", R. Ayachitula, K. D. Harris, S. J. Strutz, R. J. Twieg and L. Michael Hayden, ACS Annual Mtg. *Organic Thin Films for Photonic Applications*, (Washington, DC, Aug. 20-25, 2000)

These papers and talks contain all of the major results accomplished in the past year. The results are included in the attached documents.

Major Milestones and Accomplishments

Our basic task during this contract has been to perform the basic research and develop a prototype benchtop holographic optical storage system based on photochromic and/or

photorefractive polymers so that both permanent and erasable images may be stored and retrieved in the same mixed polymer medium.

In order to put our efforts in perspective, I offer the following table of proposed objectives excerpted from the original proposal, their proposed date of accomplishment, and their actual date of accomplishment.

Major Objective	Proposed completion date	Actual completion date
Demonstrate storage of multiple images in PR/PC materials	1998-1999	Dec 1998
Demonstrate storage and retrieval of digital image pages in PR/PC materials	1999-2000	March 1999
Determine real world writing, access, and system throughput times.	1998-2000	Writing times - Dec 1998 Access and throughput times - ongoing Storage times - March 1999
Storage of multiple permanent and erasable holograms in the same material location	no specific date proposed	March 1999
Completion of PR/PC dual grating materials study	no specific date proposed	May 2000

The documents included in this report provide the technical details of our results this year. Again, we wish to thank you for your continued support of this work.

Sincerely,

L. Michael Hayden

Dual grating photorefractive polymer

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Photorefractive (PR) polymers are potential replacements for PR crystals. The rapid response times and high diffraction efficiency[1] (>86%) of these polymers, combined with their low cost, has lead to their study for various real-time holography applications, such as phase conjugation,[2] imaging through distorting media,[3] digital data storage,[4] and the coupling of light into waveguides.[5]

A subset of the PR crystals, $Bi_{12}SiO_{20}$,[6,7] $Bi_{12}TiO_{20}$,[8] and GaP,[9] have been shown to exhibit a dual grating behavior. In these crystals, photochromic gratings are formed during the photorefractive grating formation process. These dual grating materials are useful for applications which benefit from the interaction between light diffracted from two simultaneous gratings. These applications[10] include, wave front interferometry, image synthesis, logic operations, phase object detection, holographic interferometry, and novel spatial light modulation. Here we introduce a polymer composite capable of simultaneously storing both erasable, photorefractive and quasi-permanent, irreversible photochemical (PC) holograms. Gratings may be formed in the following combinations: PC only (at 675 nm with no external field); PC and PR simultaneously (at 675 nm under the application of an external field); or PR only (at wavelengths longer than 715 nm with an applied field).

Our composite consists of the photoconductor poly(N-vinylcarbazole) (PVK), the plasticizer butyl benzyl phthalate (BBP), the sensitizer/charge generator C_{60} and the chromophore, (3-(2-(4-(N,N-diethylamino)phenyl)ethenyl)-5,5-dimethyl-1,2-cyclohexenylidene)-propanedinitrile (Lemke-e).[11] Lemke-e performs two functions in our composite. First, it is a chromophore with a large PR molecular figure of merit,[12] allowing the formation of efficient PR gratings. Second, it takes part in certain photochemical reactions when triplet sensitized. These photochemical reactions can be used to write PC gratings. The photochemical reaction begins when a photon is absorbed by one of the singlet energy levels (675 nm) of C_{60}, followed by coupling of this energy into a long lived triplet state (795 nm)[13] through intersystem crossing. The C_{60} then transfers its energy to a Lemke-e molecule through triplet-triplet energy exchange. Finally, two Lemke-e molecules undergo a 2+2 cycloaddition reaction, resulting in a change in the local absorption of the composite.[14] Interference of two coherent beams of 675 nm light inside the composite results in a local absorption grating (hologram). Recent literature has referred to this reaction as photochromic;[14,15] however, we will refer to it as photochemical or irreversible photochromic (PC) since the reaction is irreversible. The composites studied in this work consisted of Lemke-e 7%/ C_{60} 1%/BBP 21-36%/PVK 72-56% (LCBP) by weight.

We verified the PR nature of the LCBP composites by performing asymmetric two-beam-coupling (TBC) experiments and measured the diffraction efficiency of the gratings using a degenerate four wave mixing (DFWM) technique. The PR experiments were performed with either a Ti-Sapphire laser, tunable between 715 and 820 nm or a 675 nm laser diode. An electric field of 82 V/μm applied to 75 μm thick samples yielded a TBC gain coefficient of, Γ_p=156 cm^{-1}, for *p*-polarized, 730 nm light. In the DFWM experiments, overmodulation occurred at 110 V/μm for 730 nm light with a maximum external diffraction efficiency of 53% (Figure 1a inset). As expected, the PR external diffraction efficiency, η_E, of the composite decreases as the glass transition temperature (T_g) of the polymer increases (Figure 1a inset).

The PC grating formation was probed using a DFWM configuration. For these experiments, the polymer was pressed between plain glass slides with the sample normal bisecting the two write beams. A slanted geometry was not required since no field was applied to the sample. A 675 nm laser diode was used to generate the coherent *s*-polarized write beams (1.8 W/cm^2), as well as the *p*-polarized read beam (0.05 W/cm^2). The initial slopes of the curves in Figure 1a yield the sensitivities of the different composites, defined here as the square root of the diffraction efficiency divided by the exposure. The highest sensitivity, S = 0.0006 cm^2/J (λ = 675 nm), was observed for the T_g = 26 °C system. The sensitivity of the material decreased as the T_g of the material increased. As a result, the time required to write a PC grating increased with the T_g. The increased write time (decreased sensitivity) is due to a decrease in the translational diffusion of the Lemke-e molecules which slows the photochemical process responsible for PC grating formation.

Dual Use Chromophores for Photorefractive and Irreversible Photochromic Applications

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Introduction

Photorefractive (PR) polymers are potential replacements for PR crystals. The rapid response times and high diffraction efficiency of these polymers, combined with their low cost, has lead to their study for various real-time holography applications, such as phase conjugation, imaging through distorting media, and digital data storage.

A subset of the PR crystals, $Bi_{12}SiO_{20}$, $Bi_{12}TiO_{20}$, and GaP, have been shown to exhibit a dual grating behavior. In these crystals, photochromic gratings are formed during the photorefractive grating formation process. These dual grating materials are useful for applications which benefit from the interaction between light diffracted from two simultaneous gratings. These applications[1] include, wave front interferometry, image synthesis, logic operations, phase object detection, holographic interferometry, and novel spatial light modulation. Here we present results on a polymer composite capable of simultaneously storing both erasable, photorefractive and long lasting, irreversible photochromic (PC) holograms. Gratings may be formed in the following combinations: PC only (at 675 nm with no external field); PC and PR simultaneously (at 675 nm under the application of an external field); or PR only (at wavelengths longer than 715 nm with an applied field).

Experimental Details

Our composites consist of the photoconductor poly(N-vinylcarbazole) (PVK), the plasticizer butyl benzyl phthalate (BBP) or tricresyl phosphate (TCP), the sensitizer/charge generator C_{60}, and a series of chromophores with electron donating groups on the phenyl ring of [2-(5,5-dimethyl-3-styrl-cyclohex-2-enylidene)-malononitrile] (DCPT). The chromophores (Figure 1) perform two functions in our composites. First, they are chromophores with large PR molecular figures of merit, allowing the formation of efficient PR gratings. Second, they take part in 2+2 photochemical reactions when triplet sensitized.[2] These photochemical reactions can be used to write long lasting PC gratings.

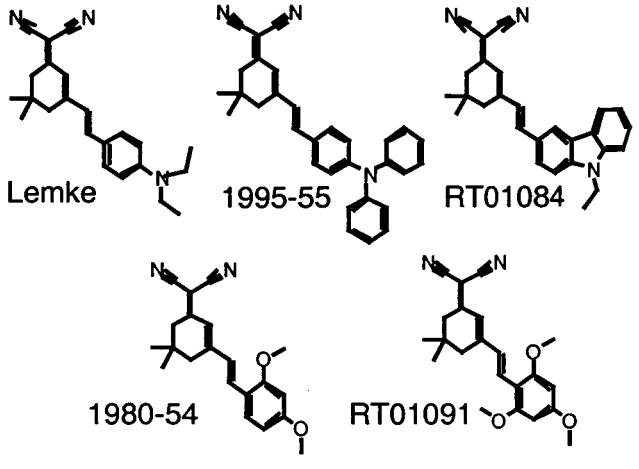

Figure 1. PR/PC chromophore structures.

Results and Discussion

We verified the PR nature of the composites by performing asymmetric two-beam-coupling (TBC) experiments. We then characterized the composites by measuring the diffraction efficiency of the gratings using a degenerate four wave mixing (DFWM) technique. The PR diffraction efficiency of the composite decreased as the glass transition temperature (T_g) of the polymer was increased.

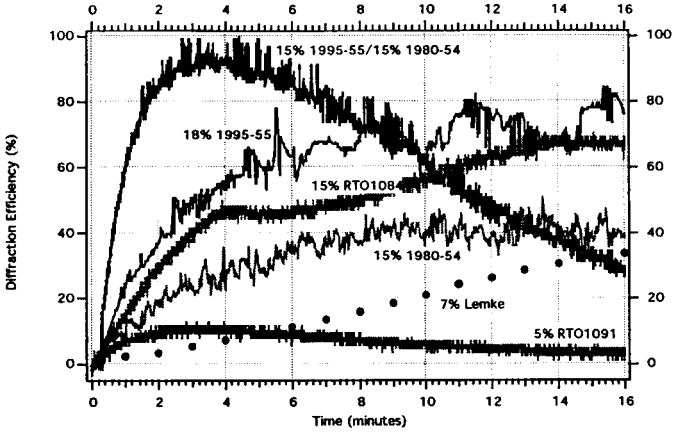

Figure 2. Transmitted (internal) PC diffraction efficiency growth rate. The films were 75 μm thick. A 675 nm laser diode was used to generate the coherent *s*-polarized write beams (1.8 W/cm^2), as well as the *p*-polarized read beam (0.05 W/cm^2).

The PC grating was also characterized by DFWM. For these experiments, the polymer was pressed between plain glass slides with the sample normal bisecting the two write beams. A slanted geometry was not required since no field was applied to the sample. The diffraction efficiency plotted in Figure 2 is the ratio of the diffracted intensity to the intensity transmitted through the sample before the grating was written. The initial slopes of the curves in Figure 2 yield the sensitivities S of the different composites, defined here as the square root of the diffraction efficiency divided by the exposure. The sensitivity of the material decreases as the T_g of the material is increased. The increased writing time (decreased sensitivity) is due to a decrease in the translational diffusion of the chromophore molecules at higher T_g, which slows the photochemical process responsible for PC grating formation.[3]

Table 1. PC Sensitivity of the Composites

Material	S (cm^2/J)
15% 1995-55/15% 1980-54/25% BBP/44% PVK/1% C_{60}	18×10^{-3}
18% 1995-55/24% BBP/57% PVK/1% C_{60}	13×10^{-3}
15% RTO1084/35% TCP/49% PVK/1% C_{60}	10×10^{-3}
15% 1980-54/23% BBP/61% PVK/1% C_{60}	5.6×10^{-3}
5% RTO1091/24% BBP/70% PVK/1% C_{60}	5.0×10^{-3}
7% Lemke/24% BBP/68% PVK/1% C_{60}	1.4×10^{-3}

Each of the composites listed in Table 1 exhibits both PR and PC activity and is a potential candidate for use in the applications mentioned in the introduction.

Acknowledgement. LMH would like to thank the Earth and Space Data Computing Division at NASA Goddard for supporting this work. SJS would like to thank the NASA Goddard Space Flight Center for a Graduate Student Research Fellowship. Research at Kent State University was supported by ALCOM (NSF DMR 89-20147)

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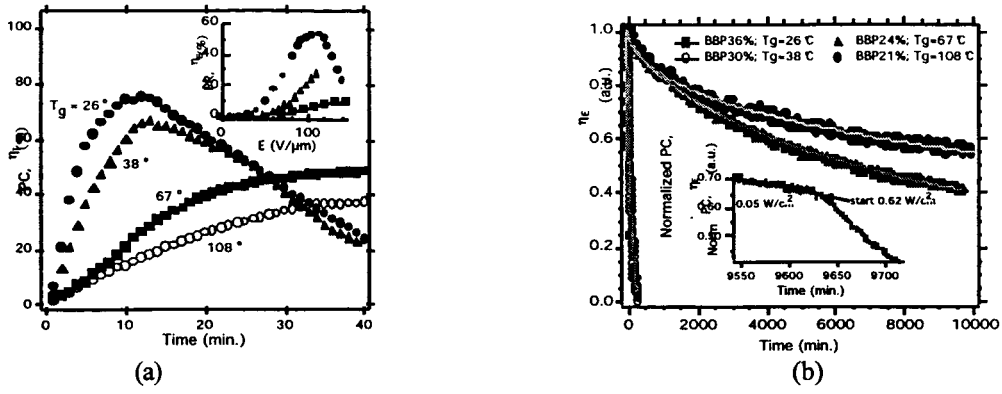

Figure 1 (a) PC write times and PR diffraction efficiencies versus T_g. The main figure shows the internal diffraction efficiency, η_I of the PC gratings. The inset figure shows the T_g dependence of the PR external diffraction efficiency, η_E; (b) Lifetime of PC holograms. The main figure shows grating decay during sparse reading; the inset during continuous reading.

A maximum internal diffraction efficiency, η_I = 74 % (η_E = 20%) was reached in the BBP 36% (T_g=26°C) system after writing for ten minutes. Since the photochemical grating formation process is irreversible, the decrease in η_I after reaching a maximum value in the low T_g systems is not due to overmodulation, as in the PR effect, but due entirely to the translational diffusion of the molecules that formed the grating. Thus, after a grating is formed, translational diffusion causes the grating to deteriorate in low T_g systems. The translational diffusion is reduced in the higher T_g systems. The effect of translational diffusion on the lifetime of the PC gratings is shown in Figure 1b. After writing a PC grating to its maximum efficiency ($\eta_E \approx 20\%$), the decay in the diffraction efficiency of the grating was periodically probed with a low power density (0.05 W/cm^2) read beam of the same wavelength. The *1/e* times were 24, 91, 11228 (7.7 days), and 26201 minutes (18.2 days) for the 26°C, 38°C, 67°C, and 108°C systems, respectively. Grating decay due to continuous reading can be alleviated by reading with a longer wavelength. For instance, 730 nm light reconstructs PC holograms but does not photochemically alter the material and hence will not cause erasure of the PC holograms. We are currently attempting to increase the dye concentration in our samples by either using dyes with lower absorbance at 675 nm, or by selecting a triplet sensitizer for Lemke-e that allows us to use longer wavelengths. Our early results (Figure 2) show that by using a low absorption chromophore and simply doubling the chromophore concentration, we can increase the sensitivity to S = 0.003 cm^2/J (sample T_g=67°C), a factor of 5 increase.

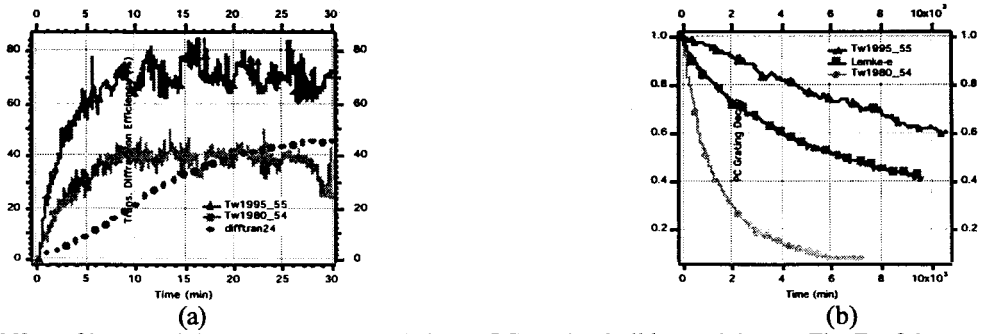

Figure 2. Effect of increased dye concentration and size on PC grating buildup and decay. The T_g of these systems was 67°C. Tw1995-55 and Tw1980-54 (synthesized by R. Twieg of Kent State Univ.) have blue shifted absorption maxima compared to Lemke-e, allowing higher concentrations, while Tw1995-55 is larger and Tw1980-54 is smaller than Lemke-e.

PC and PR holograms can also be stored in the same location. A LCBP (BBP 36%) composite was placed into a holographic storage setup which incorporated a spatial light modulator and multiple wavelength capability. We then stored and recovered holograms from the same location by illuminating the sample with the appropriate wavelength (Figure 3). Illumination by either 675 nm or 730 nm light reconstructed both holograms. However, when the PC hologram was reconstructed by 730 nm light or the PR grating was reconstructed with 675

nm light, the images were diffracted away from the camera. Besides the demonstration shown in Figure 3, we were also able to angle multiplex and recover 10 PC hologrames and an erasable, dynamic PR hologram in the same volume, simultaneously.

(a) (b)

Figure 3. PC and PR holographic storage in the same sample volume. With no electric field applied, the sample was exposed to 675 nm light for two minutes to write the PC image. Exposure for one second at 730 nm with 100 V/μm applied recorded the PR image. Subsequent illumination at 675 nm recovered the stored PC image (3a). After reading the PC image, switching wavelengths back to 730 nm retrieved the stored PR image (3b). The reading cycles could be repeated until the gratings were erased in a time similar to those seen in the FWM experiments. The vertical lines in the "H" are 4 pixels wide, each pixel is 15x15 μm.

The main advantage of our composite is that it is capable of storing both erasable and quasi-permanent holograms in the same location. In addition, the grating formation processes are wavelength selective, allowing for the formation of simultaneous PR and PC gratings or each grating type individually. Using 675 nm light and an external electric field, causes the formation of both grating types, allowing applications such as holographic interferometry, image synthesis, and logic operations to be carried out. Using longer wavelengths, purely PR holographic operations such as optical switching and imaging though distorting media are possible. Finally, long term storage applications like digital data storage may be possible by writing purely PC gratings at 675 nm with no external field.

Acknowledgments. We thank the Earth and Space Data Computing Division at the NASA Goddard Space Flight Center for its support.

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