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Characterisation of an Acrylamide-based Photopolymer for Data Storage Utilizing Holographic Angular Multiplexing

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Abstract

An acrylamide-based photopolymer formulated in the Centre for Industrial and Engineering Optics has been investigated with a view to further optimisation for holographic optical storage.

Series of 18 to 30 gratings were angularly multiplexed in a volume of photopolymer layer at a spatial frequency of 1500 lines/mm. Since the photopolymer is a saturable material, an exposure scheduling method was used to exploit the entire dynamic range of the material and allow equal strength holographic gratings to be recorded.

This investigation yielded the photopolymer M/# for moderately thin layers. Photopolymer temporal stability was also studied by measuring variations of material shrinkage, Bragg selectivity curve, and diffraction efficiency.

PACS codes**:** 42.40.i, 42.40.Lx, 42.70.Ln, 42.70.Jk, 42.70.Gi

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1. Introduction

With potential storage capacities of 1TB/cm^3 and data transfer rates of 10 GB/s, holographic data storage (HDS) is positioned to be a serious competitor in the next generation of optical storage devices [1-3]. HDS has the advantage that the entire volume of the recording medium is used. Data pages are multiplexed (superimposed) into the same volume allowing for the parallel recording and readout of data, thus achieving high data transfer rates and short access times. The advent of commercial HDS systems and photopolymer media developed by companies such as InPhase Technologies, Optware, and Aprilis, [4-5] indicates the technology is no longer merely confined to the optical bench. The emphasis is now on investigating different materials to find the optimum medium for this new technology.

To date, many photopolymers have been investigated for their suitability as HDS media. This article details certain properties of one such photopolymer. The dynamic range, the total response of the material when multiple gratings are multiplexed in the same volume was investigated. The temporal stability was also studied by measuring the photopolymer layer shrinkage, changes in the Bragg selectivity curve and diffraction efficiency. Until now, the material's chemistry and layer preparation has been optimised for applications other than HDS, such as real time holographic interferometry [6], electronic speckle pattern interferometry [7], and holographic optical elements [8].

The work presented below was carried out to assess the material's performance in its current formulation for moderately thin layers, with regard to the properties mentioned above. For this purpose an angular multiplexing system was developed. This system relies on angular Bragg selectivity [9] to record individual gratings in the same volume and is computer controlled by means of software written in LabVIEW, which, along with the self developing capabilities of the material, enables a high degree of automation in the experimental process. The system allows multiple gratings to be recorded in the same volume of material, with each grating being recorded at an angle that is Bragg mismatched from its neighbouring gratings.

The system was used to determine the photopolymer's M/# for holographic gratings with low diffraction efficiency, η, recorded at a spatial frequency of 1500 line/mm. The diffraction efficiency was taken to be the intensity of the diffracted beam divided by that of the input reference beam intensity. To determine the material's dynamic range, sets of 18 to 30 multiplexed holographic gratings were equalised by means of an iterative exposure scheduling method [10]. From this the M/# was calculated, indicating the square of the average diffraction efficiency for N number of holograms recorded using a particular recording geometry, this being obtained from the equation $\eta = (M/\#^2/N^2)$. In general materials with large values of M/# are more suitable for use as HDS media.

The material's temporal stability was also investigated with particular reference to material shrinkage, Bragg selectivity curve, and the diffraction efficiency. This was accomplished by monitoring a single slant grating during a 37 day period. It is necessary that there is no significant shrinkage or swelling of the material after recording, as a medium having high storage capacity, but suffering from mechanical distortion, will exhibit a decrease in signal-to-noise ratio and consequently an increase in bit error rate.

2. Photopolymer Chemistry

The acrylamide-based photopolymer is a self-developing dry layer. It consists of a polyvinylalcohol binder in which a monomer, an electron donor, and dye sensitizer (erythrosine B) are dissolved. When a dye molecule absorbs a photon in the presence of an electron donor, free radicals are produced that cause local polymerisation of the acrylamide. A corresponding variation occurs in the local refractive index of the material [11]. This variation is not only attributed to bond conversion in the polymerisation process, but also to the secondary effect, whereby the local density is modulated as mass transport occurs during the recording.

The photopolymerisation process starts when an erythrosine B molecule (XD) absorbs a photon of light and is promoted to an excited singlet state. It may then transfer to an excited triplet state through intersystem crossing and react with the electron donor (ED), triethanolamine, to produce a dye radical and a triethanolamine free radical.

$XD + h\nu = {}^1XD^*$ (absorption of light) (1)

${}^1XD^* = {}^3XD^*$ (intersystem crossing) (2)

${}^3XD^* + ED = XD\bullet + ED\bullet$ (3)
(dye reduction-electron transfer)

By itself, the dye radical is not normally reactive enough to initiate polymerisation. The polymerisation is usually due to the triethanolamine radical reacting with an acrylamide molecule (ACR):

$ED\bullet + ACR = ED\text{-}ACR\bullet$ (initiation) (4)

$ED\text{-}ACR\bullet + ACR = ED\text{-}(ACR)_2\bullet$ (propagation) (5)

3. Experimental

A plane wave recording system utilizing angular multiplexing was constructed, Fig. 1, having a rotational stage whose axis is at the intersection of the object and reference beams, and in the plane of the photosensitive layer. An Nd:YVO_4 (Verdi V-5) 532 nm laser beam was spatially filtered using a 10x objective lens and a 15 μm pinhole. The filtered beam was then collimated before being split into two equally intense beams of 10 mm diameter. The angle between the beams was set to approximately 47^0. This produced holographic gratings with a spatial frequency of 1500 lines/mm. Storing multiple gratings in the same volume entailed rotating the photopolymer layer by an angle of 1.5^0 between recordings. This angle was chosen so that the first order diffracted peak of each grating would not contribute to or detract from the diffraction efficiency of neighbouring gratings. A combined exposure intensity of 3.2 mW/cm^2 was used to record the individual gratings; the exposure energy varied for each grating due to the different scheduling times being used.

The stored holographic gratings were readout with the same wavelength laser as used to record them. The reference beam power was reduced to ≈ 128 μW/cm^2 so that additional photopolymerisation of the material would be negligible at readout.

4. Determination of M/#

Since the photopolymer is a saturable material, its dynamic range is not shared equally among gratings recorded with equal exposure energy within the photopolymer layer. As a result each additional grating recorded has less potential for modulating the photopolymer's refractive index. The diffraction efficiency of the multiplexed holographic gratings is expected to scale as $1/N^2$ [12], therefore as the number of holographic gratings stored in the photopolymer increases, appropriate exposure scheduling becomes increasingly important. For this reason an iterative exposure scheduling method was used [10], enabling the dynamic range to be equally shared between all the gratings.

To determine the material's M/# for layers ≈ 160 μm thick, series of 18 to 30 holographic gratings were multiplexed in the photopolymer layer. As more gratings are multiplexed in the volume the smaller the angle between the photopolymer layer and the bisector of the recording beams becomes; it is for this reason that only 30 gratings were recorded as more than this would result in high Fresnel reflection losses. The recording system is being modified so that it utilizes peristrophic as well as angular multiplexing; in future this should increase the number of gratings able to be recorded. To reduce the inhibition period, i.e. the time it takes for the concentration of an inhibitor molecule, oxygen in this case, to fall below a critical value whereby the polymerization process can begin [13], a preexposure of 480 μJ/cm^2 was used before the multiplexed gratings were recorded. This energy was delivered to the material through one beam. A delay of 100 seconds was then used to allow for all the preexposure processes within the material to come to a halt.

5. Exposure scheduling method

An exposure scheduling method [10] was used to equalize each set of multiplexed gratings. First, a plot of diffraction efficiency, η, against exposure energy for equal strength gratings was obtained. To determine how the gratings scale, a second graph of the cumulative grating strength was plotted. The cumulative grating strength was defined as being the integral of the square root of the recorded gratings' diffraction efficiencies.

$$A = a_0 + a_1E + a_2E^2 + a_3E^3 + \ldots + a_6E^6 \qquad (1)$$

where A is the cumulative grating strength and E is the total exposure energy. By differentiating the polynomial, the grating strength growth rate as a function of exposure energy is obtained. Fourth, fifth, and sixth-order polynomials were found to have sufficient degrees of freedom to fit the data,

$$\frac{dA}{dE} = a_1 + 2a_2E + 3a_3E^2 + \ldots + 6a_6E^5 \qquad (2)$$

Obtaining equal strength gratings is a matter of dividing the available dynamic range equally among the number of gratings to be multiplexed. Thus, the desired exposure schedule becomes,

$$\frac{A_{Sat}}{N} = \left.\frac{dA}{dE}\right|_{E=\Sigma_{i=1}^{n-1}E_i} \times E_n \qquad (3)$$

where N is the number of gratings to be recorded, A_{Sat} is the saturation grating strength, E_i is the amount of energy the photopolymer received to record the i^{th} grating, and E_n is the amount of energy required to record the n^{th} grating. Each grating is allocated *1/N* of the photopolymer's dynamic range.

It is essential that the combined exposure intensity used when plotting the cumulative grating strength, be used for subsequent recordings for the polynomial fit to remain valid. From the energy equation $E = I.t$ where I is the combined intensity of the two beams and t is the exposure time, the exposure schedule in terms of time becomes,

$$t_n = \frac{A_{Sat}}{N \times I\left[a_1 + 2a_2\left(\sum_{i=1}^{n-1} Ei\right)^1 + 3a_3\left(\sum_{i=1}^{n-1} Ei\right)^2 + \ldots + 6a_6\left(\sum_{i=1}^{n-1} Ei\right)^5\right]} \qquad (4)$$

where t_n is the exposure time of the n^{th} grating and I is the total incident intensity.

By recording a new set of gratings using the times generated from eqn. 4, the diffraction efficiencies of the newly recorded gratings were uniformly distributed. Increasing the uniformity further is simply a matter of repeating the exposure scheduling method outlined above as many times as is required.

Once each set of gratings was recorded with approximately uniform diffraction efficiencies, the new exposure times were used to repeat the experiment for several series of photopolymer layers each having a thickness ≈ 160 μm. The combined data from all these experiments was plotted on a graph of $\sqrt{\eta}$ against the reciprocal of the number of holograms recorded. The mean slope was then obtained, this being equal to the M/#.

6. Results and Discussion

As can be seen in Fig. 2 a constant two second exposure produces holographic gratings of highly differing strengths. The majority of the diffracted energy is concentrated in the first five gratings. The material's dynamic range is effectively consumed too early in the multiple grating recording process. This is not an ideal situation for holographic data storage considering that all gratings should have the same signal-to-noise ratio with each grating having a diffraction peak equally as strong as its neighbour. With this in mind, it becomes clear that a large series of weaker but equalised gratings is more desirable.

To increase the uniformity of multiplexed gratings it is necessary to progressively increase the exposure times used for recording in such a way as to compensate for the reduced dynamic range that may exist by the time a particular grating is recorded. A grating that, without scheduling, would have a large diffraction efficiency is therefore recorded using a shorter exposure time, and a grating that would otherwise have a low diffraction efficiency is exposed for longer. In this way, the diffraction efficiencies can be equalized for a large number of gratings.

This was done by plotting the dependence of the cumulative grating strength for gratings recorded with constant 2 s exposure on the cumulative exposure energy. The grating strength growth rate (equation (7)) was determined from this graph and substituted into equation (9). The times generated from this equation were used to record new gratings, these being partially equalized. The exposure scheduling method was iterated three or four times depending on the number in the set of holograms to be equalised. It was found that three or four iterations was optimal as further iteration led to overall decrease of the grating strength without improvement of the uniformity of the diffraction gratings' efficiencies. For example once scheduling was complete, the exposure times generated ranged from 0.6 to 7.9 s for 18 and 0.3 to 8.6 s for 30 gratings. After exposure scheduling times have been determined, new gratings are multiplexed in the same volume, this time with the new exposure times.

The reconstructed gratings exhibit a roughly uniform profile; see figure 3. Some anomalous grating peaks were seen to occur; this may be attributed to imperfections in the optical quality of some of the photopolymer layers that were used as well as the glass substrate, as well as inadequate pre-exposure.

The requirement for low Fresnel reflection losses also limits the amount by which the layer can be rotated with respect to the object and reference beam. As the angle between the bisector of the recording beams becomes greater with respect to the layer normal, the layer reflects more light than is transmitted into its interior. This means there is less available light energy for refractive index modulation of the material. Consequently, gratings of lower diffraction efficiency are recorded. It is noted that for the maximum recording angle (22.5^0 from the photopolymer normal) used during the course of this work the Fresnel losses incurred were approximately 4%. This is similar to the losses that would be expected if the beams were normal to the layer; this was true also for the temporal stability study.

Since the optical quality of the layers was reasonably good, the anomalous peaks are attributed to inadequate preexposure of the photopolymer before the gratings were multiplexed in the layer volume.

In Fig. 4 a plot of the square root of the mean diffraction efficiency as a function of the reciprocal of the number of gratings was made. The slope of this graph produces an M/# of 3.6 for layers ≈ 160 μm thick.

A photopolymer layer with thickness of 160 μm was found to have an M/# of 3.6. This compares favorably to the two-chemistry system (formulations 523530 and 323530) developed by InPhase Technology [4]. The quoted M/# values are taken based on 200 μm samples. In the case of 523530 the M/# is 6.7 and the shrinkage is 0.1% whereas 323530 has a higher M/# but also a higher shrinkage of 0.5%. Thicker samples of 1.5mm produce M/# of 50 and 45 respectively [4] The M/# for our material is still unknown at these higher layer thicknesses. It is reasonable to assume that they too will yield higher values. Work is currently being carried out to produce thicker photopolymer samples, which have good optical quality.

7. Temporal stability

Shrinking or swelling of a photopolymer layer during or after recording of a slanted grating, can result in a drop off in diffraction efficiency of the reconstructed grating or worse, no diffraction of the signal beam at the expected reconstruction angle. The reason for this is due to swelling or shrinkage of the layer causing the slant angle of the grating fringes to change, thus leading to a corresponding change in the fringe period. Playing back the grating with maximum diffraction efficiency now requires that either the wavelength or the reference beam angle be readjusted.

As well as mechanical changes compromising photopolymer layer stability, undesired diffusion and polymerisation of the acrylamide monomer after recording may also cause a reduction in diffraction efficiency or have an adverse effect on the Bragg selectivity curve.

7.1 Shrinkage

To determine the shrinkage of the photopolymer layer, a single slanted holographic grating was recorded. The layer itself was approximately 120 μm thick and was recorded with a slant angle of 20^0. This is taken to be the angle between the photopolymer layer normal and the bisector of the recording beams. The grating spatial frequency was set to be 1500 lines/mm and the exposure energy used to record the grating was 23 mJ/cm^2. This exposure energy was delivered to the grating by an Ar$^+$ 514 nm wavelength laser. After recording, the grating was probed with a wavelength of 514 nm and an intensity of 59 μJ/cm^2. To protect the layer it was sandwiched between two glass plates and sealed before recording. An angular scan was then made of the diffracted light intensity. The grating has a typical diffraction efficiency that would be expected if it were multiplexed in a layer with additional gratings.

As stated previously, the occurrence of swelling or shrinking of the photopolymer layer results in a change in the fringe spatial period [14]. It is therefore necessary to alter the readout beam angle for maximum diffraction efficiency.

The fractional change in material thinckness, Δd, can be obtained by knowing the initial slant angle of the grating ϕ_0 and the final slant angle ϕ_1.

$$\Delta d = d_0 \left[\frac{\tan \phi_1}{\tan \phi_0} - 1 \right] \tag{5}$$

7.2 Diffraction efficiency and Bragg selectivity curve

It was seen that during the 37 day period, the replay angle of the slanted grating had shifted from the angle at which it was recorded. This was attributed to a decrease in the slant angle because the overall photopolymer layer had shrunk. It can be seen in Fig. 6 that there was no significant change in the shape of the Bragg selectivity curve over this period of time, however the diffraction efficiency varied by 11% between the initial and the final angular scan.

8. Summary

A recording system utilising angular multiplexing, which was controlled by means of software written with LabVIEW, was used to determine certain characteristics of a novel acrylamide-based photopolymer system. Sets of 18 to 30 multiplexed gratings were equalized. From the results obtained the material's M/# in the particular geometry used was calculated to be 3.6 for photopolymer layers ≈ 160 μm thick.

Furthermore, the photopolymer's temporal stability was investigated; it was found that initially the material shrank by 1% after the first day, and reached a maximum shrinkage of 2% after 37 days. This is quite large compared with shrinkage values quoted for InPhase Technology's 523530 and 323530 photopolymers, these being 0.1% and 0.5% respectively. Work is currently being carried out to reduce the shrinkage of our material and so make it more conducive to holographic data storage. The diffraction efficiency of the slanted grating varied by 11% of its original diffraction efficiency between the initial and the final angular scan over the same time period while the change in the shape of the Bragg selectivity curve was seen to be negligible.

This is an ongoing investigation and work is currently being carried out to produce ≈ 1mm thick samples of high optical quality for further analysis.

References

[1] G. Berger, C. Denz, S. S. Orlov, B. Phillips, L. Hesselink, Dec. 2001, Appl. Phys. B 73, 839-845.

[2] S. S. Orlov, W. Phillips, E. Bjornson, Y. Takashima, P. Sundaram, L. Hesselink, R. Okas, D. Kwan, R. Snyder, Sept. 2004, App. Opt. Vol. 43, No. 25, 4902-4914.

[3] G. W. Burr, J. Ashley, H. Coufal, R. K. Grygier, J. A. Hoffnagle, C. M. Jefferson, B. Marcus, May 1997, Opt. Lett. , Vol. 22, No. 9, 639-641

[4] M. Schnoes, B. Ihas, A. Hil, L. Dhar, D. Michaels, S. Setthachayanon, G. Schomberger and W. L. Wilson "Holographic Data Storage Media for Practical Systems", www.inphase-technologies.com/technology/whitepapers

[5] R. T. Ingwall, D. A. Waldman, Dec. 2000, SPIE, Holography, Vol. 11, No. 2, 1, 1-7.

[6] E. M. Mihaylova, V. Toal, S. R. Guntaka, S. Martin, Nov 2003, Pub: Proc. SPIE Vol. 5226, 209-213, 12th International School on Quantum Electronics: Laser Physics and Applications; P. A. Atanasov, A. A. Serafetinides, I. N. Kolev; Eds:

[7] E. Mihaylova, I. Naydenova, H. Sherif, S. Martin, Vincent Toal, Feb. 2003, Proc. of SPIE, Vol. 5249, Optical Systems Design and Engineering, L. M. Philip R., R. Wartmann, Editors, Saint-Etienne, 318-326.

[8] S. R. Guntaka, V. Toal, S. Martin, Dec. 2002 App. Opt., 10, 41 (35), 7475-7479,

[9] H. Kogelnik, Nov. 1969, Bell Sys. Tech. J. Vol. 48, 2909-2947.

[10] A. Pu, K. Curtis, D. Psaltis, Oct. 1996, Opt. Eng. 35 (10) 2824-2829.

[11] S. Martin, P. Leclere, Y. Renotte, V. Toal, Y. Lion, Sept. 1994, Opt. Eng., Vol. 33 3942.

[12] G. W. Burr, D. Psaltis, June 1996, Opt. Letts. Vol. 21, No. 12, 896-898

[13] T. V Galstian, S. Harbour, A. V Galstyan, R. S Hakobyan, May 2004, electronic-Liquid Crystal Communications

http://www.e-lc.org/docs/2004_05_05_11_13_17

[14] F.T. O'Neill, J.R. Lawrence, J.T. Sheridan, April 2001, Opt. Eng. 40(4) 533-53.

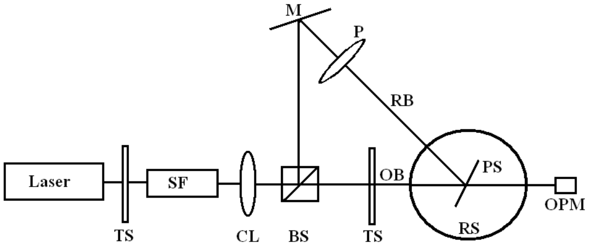

Fig. 1 Experimental set-up for characterisation of acrylamide based photopolymer, TS: shutter, SF: spatial filter, CL: collimating lens, BS: beam splitter, P: polarizer, RS: rotational stage, PS: photopolymer holder OPM: optical power meter, M: mirror, RB: reading beam, OB: object beam.

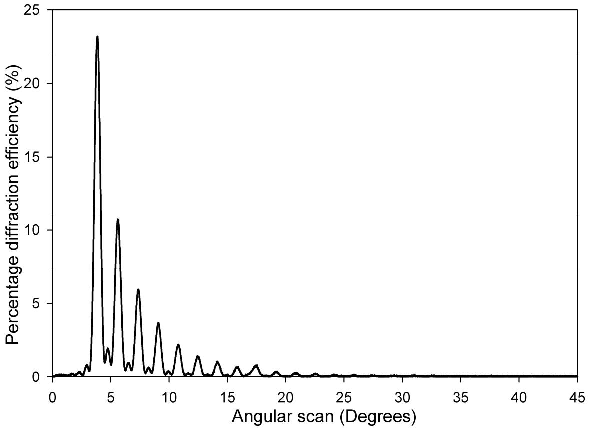

Fig. 2 Diffraction efficiency of 30 reconstructed holographic gratings. The first nine Bragg curves can easily be seen as these gratings consume the majority of the dynamic range.

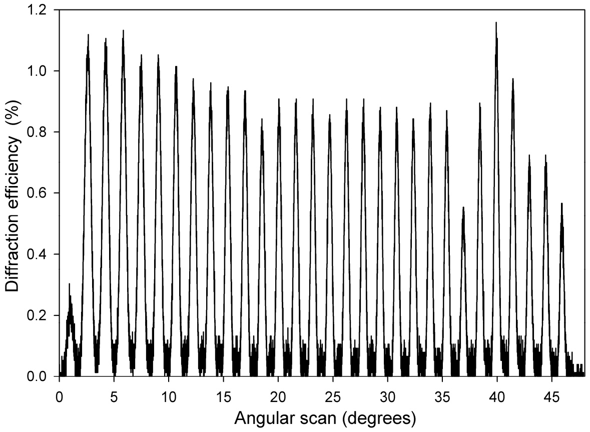

Fig. 3 Diffraction efficiency of 30 holographic gratings as a function of angular scan. An exposure schedule has been applied to equalise the gratings.

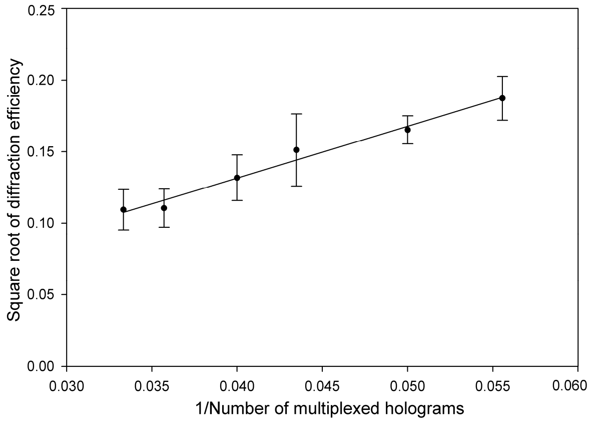

Fig. 4 The square root of diffraction efficiency as a function of the reciprocal of number of gratings multiplexed yields an M/# = 3.6 for layers ≈ 160 μm thick.

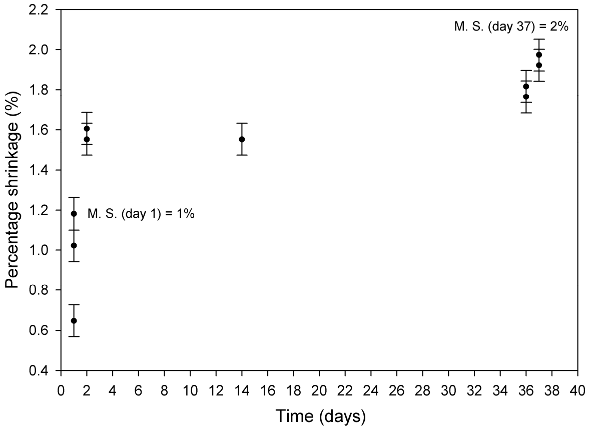

Fig. 5 Study of the percentage shrinkage change of a signal slant grating over a period of 37 days. M. S. = maximum shrinkage.

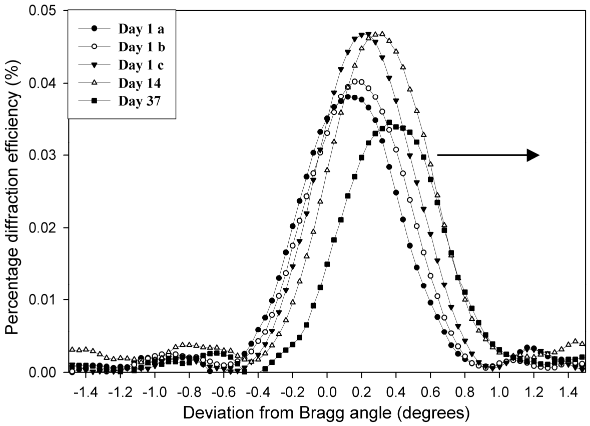

Fig. 6 Variation in the Bragg replay angle of the slanted grating during the period of investigation. The arrow indicates the direction of shrinkage.