

Full length article



Full-color multiplexed reflection hologram of diffusing objects recorded by using simultaneous exposure with different times in photopolymer Bayfol® HX

Irene Vázquez-Martín^a, Julia Marín-Sáez^{a,b}, Marina Gómez-Climente^a, Daniel Chemisana^{b,*}, María-Victoria Collados^a, Jesús Atencia^a

^a Applied Physics Department, Aragon Institute of Engineering Research (I3A), Faculty of Science, University of Zaragoza, Pedro Cerbuna 12, 50009 Zaragoza, Spain

^b Applied Physics Section of the Environmental Science Department, Polytechnic School, University of Lleida, Jaume II 69, 25001 Lleida, Spain

ARTICLE INFO

Keywords:

Full-color holography
Volume hologram
Denisyuk hologram
Reflection hologram
Photopolymer

ABSTRACT

An optimized wavelength multiplexing procedure for color reflection holograms of diffusing objects recorded in Bayfol® HX photopolymer is proposed. It is based on simultaneous initial illumination of all the wavelengths and sequential shuttering down of the laser beams involved. Three lasers of 442 nm, 532 nm and 633 nm were selected, as a tradeoff solution between setup complexity and color reproduction accuracy. The obtained results prove that the presented method improves the simultaneous and sequential exposure methods by increasing the mean efficiency and reducing the standard deviation (increasing uniformity) of the involved wavelengths. For a Spectralon® diffusing object, the assessed method reports a mean efficiency of 25.0% with a standard deviation of 2.9%, while simultaneous and sequential iterative exposures result in mean efficiencies of 12.1% and 20.6% with standard deviations of 3.4% and 5.4%, respectively.

1. Introduction

Among different applications of holography, color reproduction represents one of the most relevant. Several fields such as art [1,2], security holography [3], sensors [4,5], displays [6,7] and LEDs [8] demonstrate the successful application of color holograms [9]. Panchromatic holograms can be either transmission-type or reflection-type. The last ones are suitable for color reproduction when reconstructed with white light and are therefore the most commonly used for this kind of application. On the contrary, transmission color holograms need to be illuminated with the wavelengths and directions used in the recording stage.

First color reflection holograms were obtained by recording with single wavelengths and then stacking them to be reconstructed under color conditions. In an analogous way, a sandwich of monochromatic sensitive materials before recording was assembled in some cases to avoid subsequent stacking. An example of the monochromatic recording with subsequent stacking is the one reported by Kubota who combined silver halide (red component) and dichromated gelatin (green and blue components) holograms to obtain a successful reflection color hologram

[10]. This methodology reports high-efficiency results since the photosensitive material index modulation is not shared for different wavelengths. More recently, Shelkovnikov et al. analyzed the performance of a polymer under several sandwich configurations (as it was previously indicated, the sandwich is prepared before recording) for two-color holograms, obtaining diffraction efficiencies above 70% [11]. Another representative study about multiple photosensitive layer architectures in photopolymers is the one by Kawabata et al. in which multilayer films containing different spectral-sensitive photopolymer layers were assessed, achieving efficiencies of 60% at each color [12]. Similarly, several configurations of monochromatic grouped layers were studied by Mukawa et al. to obtain full color displays using waveguides [13].

In the 90s, when panchromatic recording materials were available, Bjelkhagen et al. [14] demonstrated the possibility of recording color holograms in a single-layer silver halide emulsion by wavelength multiplexing. The proper multiplexing conditions (exposure times, intensities, etc.) are highly dependent on the photosensitive material involved. In this sense, panchromatic photopolymers present important advantages such as grain absence and, therefore, avoidance of diffusion in the blue region in comparison to silver halides. In the literature about

* Corresponding author.

E-mail address: daniel.chemisana@macs.udl.cat (D. Chemisana).

<https://doi.org/10.1016/j.optlastec.2021.107303>

Received 14 January 2021; Received in revised form 10 May 2021; Accepted 1 June 2021

Available online 11 June 2021

0030-3992/© 2021 The Author(s).

Published by Elsevier Ltd.

This is an open access article under the CC BY-NC-ND license

(<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

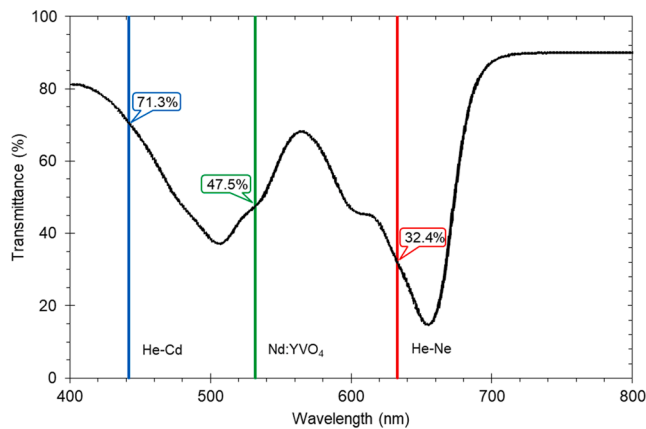


Fig. 1. Spectral transmittance of Bayfol® HX photopolymer and wavelengths of the lasers used in the experimental setup.

photopolymers, two main multiplexing techniques have been reported: simultaneous and sequential exposure. Simultaneous exposure is based on illuminating the recording material with different wavelengths at a time and during the same time interval. The dosage of the individual wavelengths is controlled by the exposure time (the same for all) and the intensity of the laser. In the case of simultaneous exposure, several problems associated with the lack of accurate control of the recording intensity may arise. Lasers with variable power, continuous density filters or two combined linear polarizers would be needed to obtain the required recording intensity values. Resulting efficiencies of the wavelengths involved are then difficult to be controlled and, thus, the color management [15]. On the other hand, by using sequential exposure, the lasers act on the recording material one after the other and during a certain time period, depending on the adequate dosage for every wavelength employed. Piao et al. [16,17] presented a variation of the sequential exposure founded on time-scheduled iterative exposure. This means to repeat an illumination sequence for a certain number of cycles achieving high diffraction efficiencies (over 50%), but for transmission-type multiplexed gratings.

It should be noted that in the studies described above (from other authors) either holographic gratings are recorded or the calibration has been carried out using gratings. In none of them the calibration was conducted by means of diffusing objects. Diffusing objects perform quite differently than gratings or any type of direct beam interference since, in that case, the light is scattered in the object and multiple interferences are produced between scattered waves. This phenomenon produces speckle on the recording material, increasing the mean refractive index and, thus, decreasing the effective modulation of the refractive index in comparison to plane-wave holograms [18].

Besides the multiplexing technique, a key aspect is the number of wavelengths necessary for reliable color recording. Bjelkhagen et al. [19] stated that, although using four or more wavelengths provides a more precise color reconstruction, with three lasers with wavelengths around 466, 545 and 610 nm, the error in color reproduction is tolerable. While increasing the number of lasers improves color reproduction, it also increases the complexity and cost of the recording set-up. Consequently, a trade-off solution should be considered to balance complexity and accurate color reproduction.

Bearing in mind the issues mentioned above, the present research proposes an optimized exposure method for registering Denisyuk-type reflection holograms [20] of color diffusing objects in photopolymers improving the state-of-the-art. The technique is founded on simultaneous exposure with different times for the lasers involved, with the goal of obtaining uniform and high efficiencies for the wavelengths involved. The procedure regards the polychromatic calibration of the photosensitive material by directly including the diffusing object (Spectralon® target). To our knowledge, there are no previous works, from other

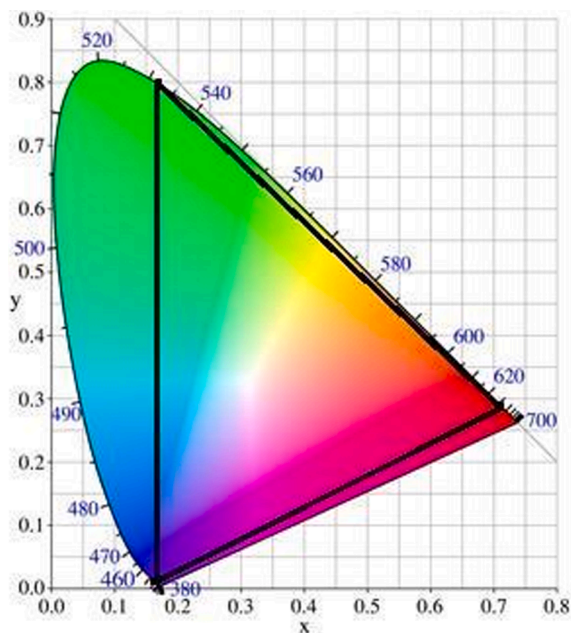


Fig. 2. Main characteristics of the lasers and their representation in the CIE diagram.

authors, performing a polychromatic calibration of the photosensitive material with a diffusing object in reflection holograms. The photosensitive recording material utilized is the panchromatic Bayfol® HX photopolymer, and the number of lasers selected is three. The manuscript is divided into five main sections. In the two first parts, the recording material and the experimental setup used are described. The methodologies of the monochromatic and polychromatic calibrations are then explained. The following section includes the results and discussion. Finally, the main conclusions are stated.

2. Photosensitive recording material

As indicated in the introduction, the photosensitive recording material utilized is the panchromatic self-processing photopolymer Bayfol® HX [21,22]. Fig. 1 illustrates the bandwidth where the polymer is more sensitive (minimum transmittance zones) and the transmittances at the wavelengths of the lasers used in the present research, which will be introduced in the following Section 3. The photopolymer layer lies on a flexible plastic substrate and its thickness is 16 μm .

The minimum transmission appears at approximately 654 nm, thus the maximum absorption occurs at this point. The second minimum is located at around 502 nm. Contrarily, the transmission of the photopolymer is quite high in the yellow region ($\sim 68\%$ at ~ 565 nm), therefore the sensitivity is low. Based on this fact, a 590 nm LuxeonRebel LT1012 LED security light was used during the experiments, located at a distance of at least 50 cm from the material, according to manufacturer's recommendations.

The material initiates a light-induced polymerization in the recording that is considered to finalize in about 5 min. Afterwards, a photocuring process is needed for fixing the hologram (polymerization of the remaining monomer and bleaching of the dye). This is done by illuminating the hologram with a white light led lamp of 50 W for 25 min [23]. The maximum index modulation attained with this material for reflection gratings is $\Delta n \cong 0.033$ [22]. Nonetheless, much smaller effective modulation is expected for the recording of diffusing objects due to the intermodulation noise produced by the interference between the object points. Before recording, the samples were adhered, by direct contact, to a 4-mm thick soda-lime glass thanks to the viscosity of the photopolymer to avoid bending since the substrate of the photopolymer

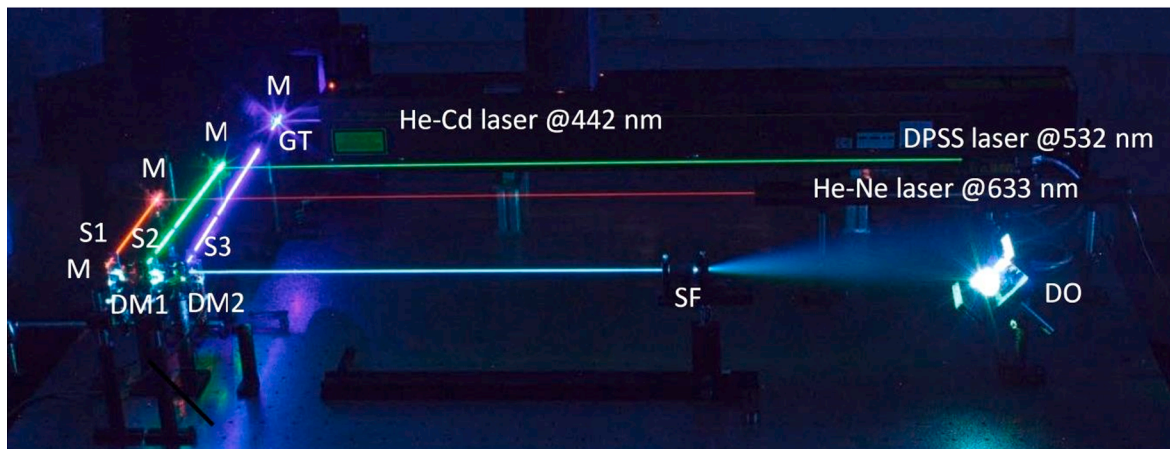
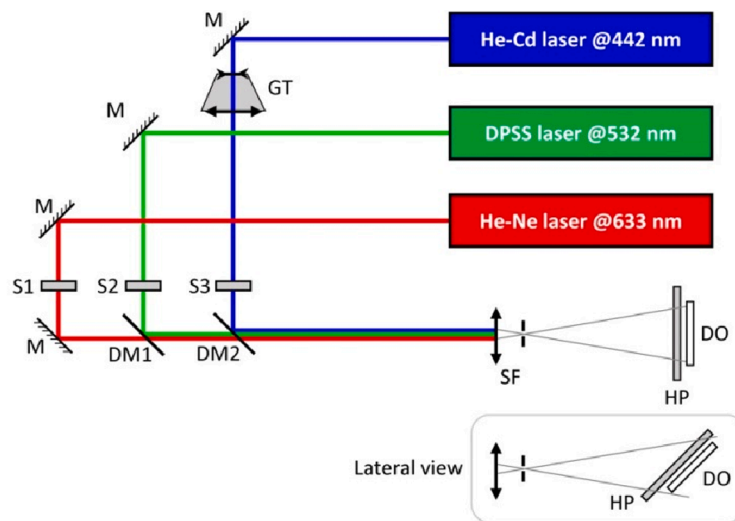


Fig. 3. Top: Recording setup diagram. GT is the Galileo telescope, M are the broadband mirrors, S1–S3 are the shutters for each laser, DM1 and DM2 are the dichroic mirrors, SF is the spatial filtering stage (microscope objective lens + pinhole), HP stands for the photopolymer and DO represents the diffusing object. Bottom: photograph of the experimental setup.

was flexible. In addition, the assembly had the rigidity to ease manipulation, holding and fixing in the recording and characterization steps.

3. Experimental setup

In the experiments conducted, the number of lasers selected was three. As it was pointed out in the introduction, three wavelengths balance the complexity of the setup while keeping the error in color reproduction at tolerable values. Taking into consideration the sensitivity of the photopolymer, the optimal values indicated in [4] for three-laser setup and the available lasers in the laboratory, two gas lasers for 442 nm and 633 nm (He-Cd and He-Ne respectively) and a Nd:YVO4 diode-pumped solid-state laser (DPSS) for 532 nm were chosen. In Fig. 2, the main parameters of the lasers jointly with their representation in the CIE diagram are summed up. The lines marked in black enclose the colors that can be reproduced with the three wavelengths used in the present investigation.

Fig. 3 depicts the experimental setup for recording the Denisyuk-type volume holograms. The He-Cd laser beam diameter is about one third the DPSS and He-Ne ones, and, therefore, a 3x Galileo telescope was added to the He-Cd beam to achieve similar illuminated spot for all the three lasers (sufficient for covering the entire object). The three beams exposure time was controlled by three independent shutters just before those beams were combined using two dichroic mirrors. Finally, the

combined laser beams are spatially filtered (microscope objective lens + pin-hole) to avoid possible noise due to dust or other effects. The lasers had linear polarization in the direction perpendicular to the plane of the figure (s-polarization).

The object used in the calibration stage is a Spectralon® lambertian-like diffusing reflector. This is perfectly attached to the glass where the photopolymer is fixed. The object and photopolymer are positioned at Brewster's angle ($\psi_B \cong 56.7^\circ$) with respect to the reference beam to eliminate additional reflections and, thus, minimizing the possibility of undesired interferences that may cause spurious holograms. The diffusing object scatters isotropically and fully depolarized the incident light; hence a big intensity loss in the object beam is experienced because only the fraction with the proper polarization interferes with the reference beam. In addition, since the photopolymer absorbs part of the blue, green and red wavelengths before reaching the object, an extra loss occurs (see Fig. 1). Therefore, in the calibration, only the reference beam intensity was considered for the exposure time calculation since this intensity was at least 10 times higher than the intensity provided by the diffusing object beam. The lambertian-like diffusing object represents the worst possible scenario in terms of efficiency and refractive index modulation. Any other less diffusing object would provide better efficiency results.

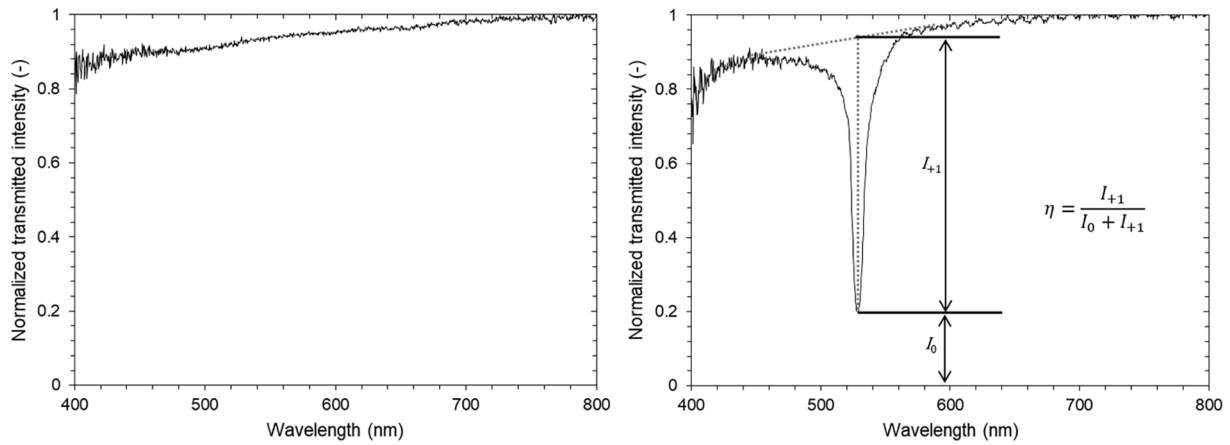


Fig. 4. Determination of the diffraction efficiency in the chromatic selectivity curve. Left: Unexposed bleached sample normalized transmitted intensity; Right: chromatic selectivity for a monochromatic green grating.

4. Methodology

4.1. Monochromatic calibration

The monochromatic calibration included recording of single-wavelength holograms to determine the response of the photosensitive material for each of the three selected colors and the recording parameters to maximize efficiency. Efficiency dependence on exposure energy was examined.

The holograms were evaluated by measuring their chromatic selectivity through the transmitted spectrum when illuminated with white collimated light (Ocean Optics, HL-2000) at the Bragg angle condition with a spectrophotometer (Ocean Optics, USB2000). The measurement was normalized to an unrecorded cured (bleached) photopolymer (Fig. 4-left) spectral transmittance curve in order to eliminate absorption and reflection losses. Additional absorption is observed in the blue bandwidth when the gratings are recorded in the photopolymer. This phenomenon is repeated in all the measurements carried out.

Fig. 4-right shows a typical transmitted spectrum of the holograms studied in this work. This spectrum corresponds to the 0 order of diffraction and, since it is a volume hologram reconstructed under Bragg conditions, only the 0 and the +1 orders of diffraction are presented. Therefore, minimum transmittance at the 0 order means maximum diffraction at the +1 order. In the same figure, the expression for obtaining diffraction relative efficiency is included.

Once the efficiency is determined, the index modulation can be derived. Typically, the refractive index modulation increases with exposure energy above a certain threshold value. In the case of low exposure energies, the increase follows a linear pattern. Moreover, it saturates for a given exposure energy value [15,16]. This performance is theoretically modelled with an exponential function:

$$n_1 = n_{1,max} (1 - e^{-\alpha(E-E_0)}), \quad (1)$$

where E_0 is the minimum exposure energy value for the index modulation to start growing and is determined experimentally, $n_{1,max}$ is the maximum achievable index modulation and α is a fitting parameter that corresponds to the slope of the linear zone of the curve.

On the other hand, the relative theoretical efficiency for a volume reflection hologram [24] that meets the Bragg condition in the reconstruction is described in Eq. (2):

$$\eta = \tanh^2 \left(\frac{\pi d n_1}{\lambda_a \sqrt{c_r c_s}} \right) \quad (2)$$

where d is the thickness of the photopolymer, λ_a is the reconstruction wavelength, c_r is the direction cosine of the angle of incidence ($c_r =$

$\cos \varphi_B \cong 0.549$) and c_s is the direction cosine of the angle of the reflected beam ($c_s = 1$).

Although volume reflection holograms efficiency could achieve a relative efficiency of 100%, in the case of diffusing object the maximum efficiency is lower since the spherical waves scattered from the different object points interfere between them, inducing speckle effect. As reported by Upatnieks et al. [18], this effect decreases the effective modulation of the refractive index compared to that obtained in the case of holograms with plane waves, and therefore an efficiency of 100% cannot be achieved.

Taking into account the exponential dependence of n_1 with the exposure energy, Eq. (1), the efficiency can be reformulated to:

$$\eta = \tanh^2 \left(\frac{\pi n_{1,max} d}{\lambda_a \sqrt{c_r c_s}} (1 - e^{-\alpha(E-E_0)}) \right) \quad (3)$$

It can be noted that the maximum relative efficiency occurs when the exposure energy tends to infinite, describing an asymptote.

Mechanical or thermal instabilities that may occur during hologram recording cause movement of the fringes. This reduces index modulation and, as a consequence, there is also a decrease in the efficiency. The theoretical curve is considered to correspond to the envelope of the points with the highest efficiency.

4.2. Polychromatic calibration

The present section deals with the characterization of the photopolymer under wavelength multiplexing conditions. In the polychromatic recording, each of the three wavelengths used aims at recording an independent hologram. The efficiency of each of these holograms is lower than that achieved with monochromatic exposure because the effective index modulation is distributed among the 3 wavelengths.

The goal of the polychromatic calibration is to accomplish similar efficiencies for every wavelength hologram, keeping them at the highest possible value. The maximum attainable efficiency value is estimated to be that reached in the monochromatic calibration.

In the introduction section, two main methods for wavelength multiplexing have been pointed out: simultaneous and sequential exposures. Besides, it has been mentioned that polychromatic calibration for reflection holograms recorded in photopolymer with diffusing object has not been reported in the literature. In this regard, the authors in the frame of a previous study reported a comparison of the simultaneous and sequential iterative exposure methods, including diffusing object [15]. Results indicated that the sequential iterative method was the best, reaching efficiencies of 22.5% for 442 nm, 14.5% for 532 nm and 24.7% for 633 nm (mean efficiency, $\bar{\eta} = 20.6\%$; standard deviation, $\sigma = 5.4\%$).

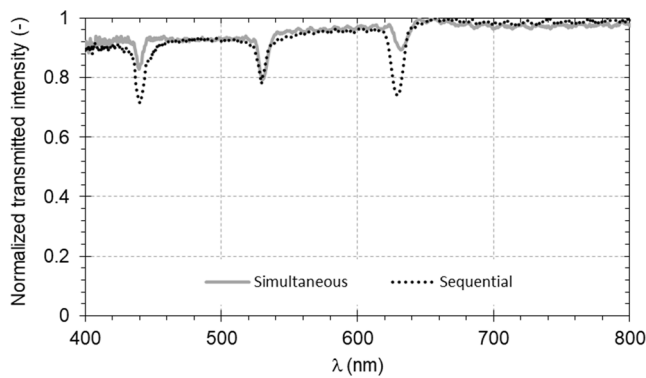


Fig. 5. Chromatic selectivity curves obtained for simultaneous and sequential exposures. The efficiencies can be derived as introduced in Fig. 4.

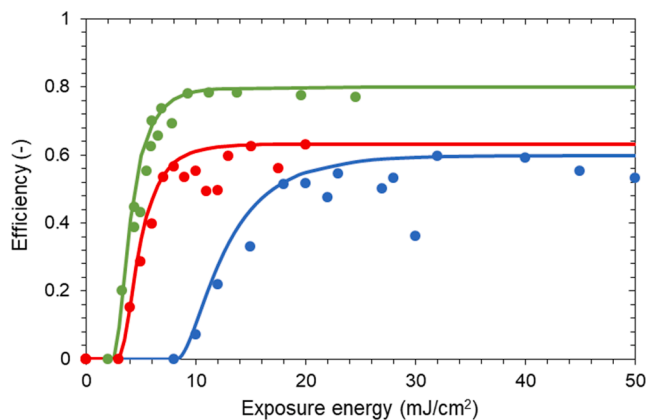


Fig. 6. Efficiency vs exposure energy for the three wavelengths of the monochromatic calibration. Blue color corresponds to 442 nm laser and the green and red colors to 532 nm and 633 nm, respectively. The lines represent the theoretical tendency (Eq. (3)).

The timings were 8, 2 and 9 s, which corresponded to 3 mJ/cm², 1 mJ/cm² and 1 mJ/cm², for 442 nm, 532 nm and 633 nm, respectively, with an optimum number of cycles of 10 to obtain the maximum and more uniform efficiency pattern. On the other hand, the reported efficiencies for simultaneous exposure were 10.9% at 442 nm, 15.9% at 532 nm and 9.6% at 633 nm ($\bar{\eta} = 12.1\%$ and $\sigma = 3.4\%$). Fig. 5 charts the spectral transmittance curves from which the diffraction efficiency for the simultaneous and sequential methods can be derived.

In spite of the fact that sequential iterative method produced better results, the uniformity in the efficiency for the different studied wavelengths was not considered to be satisfactory. With the aim of improving this uniformity and increasing the efficiency values, several method variations were analyzed, concluding that the optimal is the simultaneous exposure with different times for every wavelength. Neutral density filters were included in the setup to control the intensity of the laser beams and independent shutters did allow to operate all the beams simultaneously but to close each one at the desirable time for achieving the adequate exposure energy.

The measurements performed in this case are the same as those described in the previous subsection for the monochromatic calibration, but taking into account that the refractive index modulation is shared between the three wavelengths involved, so

$$n_{1,max} = n_{1,max,blue} + n_{1,max,green} + n_{1,max,red}. \quad (4)$$

Based on Eq. (3), to equalize the efficiency for each wavelength, the maximum index modulation values are related as

$$n_{1,max,blue}/\lambda_{blue} = n_{1,max,green}/\lambda_{green} = n_{1,max,red}/\lambda_{red}. \quad (5)$$

Table 1

Recording wavelength, measured incident intensity, maximum efficiency, exposure energy threshold, exposure energy for achieving 99% of the theoretical maximum efficiency, constant of the theoretical exponential of Eqs. (1)–(3) and maximum refractive index modulation obtained in each monochromatic calibration.

λ (nm)	442	532	633
I_i (mW/cm ²)	0.37	0.52	0.11
η_{max} (%)	60	80	63
E_0 (mJ/cm ²)	8.2	2.5	3
$E_{ 0,99\eta_{max}}$ (mJ/cm ²)	25.6	8.4	8.8
α	0.22	0.5	0.5
$n_{1,max}$	0.0067	0.0113	0.0101

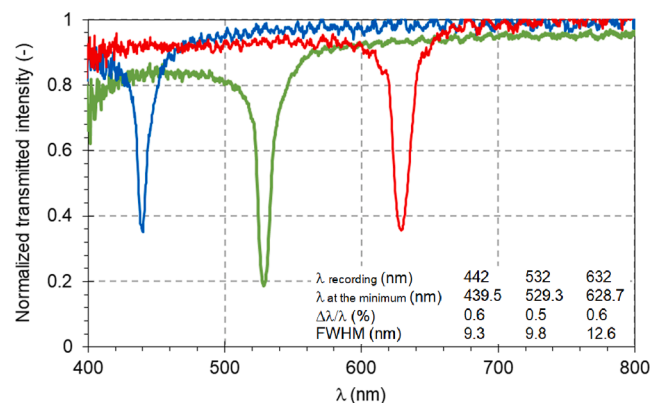


Fig. 7. Chromatic selectivity curves for the three wavelengths recorded.

5. Results and discussion

5.1. Monochromatic calibration

A series of samples were recorded under different exposure energies. Each exposure energy is represented by a point in the charts shown in Fig. 6. Moreover, the theoretical results are also included, indicating very good agreement between model and experiments.

It can be noted that for all the three wavelengths and by the exponential fitting from Eq. (3) a threshold value (E_0) is clearly identified, below which the hologram is not efficient. In the case of green and red wavelengths, the E_0 values are similar and around three times (2.5 and 3 mJ/cm², respectively) lower than for the blue one (8.2 mJ/cm²). More details about the polymerization process and material properties of Bayfol®HX film that could lead to this performance are described in [25]. In the case of the blue wavelength, experimental points are observed to experience bigger dispersion. This effect is attributed to the vibrations produced by the He-Cd laser cooling systems. From the E_0 point, the efficiency increases rapidly till the saturation zone. Since the asymptotic value that represents the maximum efficiency is achieved for exposure energies tending to infinity, it is assumed as the optimal exposure energies the one reporting maximum efficiency values at the 99% of the theoretical maximum value. In Table 1, these data, jointly with maximum efficiency, refractive index modulation and alpha parameter of the exponential fitting are included. Measured intensities for each wavelength are also shown.

Fig. 7 shows the chromatic selectivity curves for each of the individual holograms recorded with the parameters indicated in Table 1. It can be seen that the red hologram is the one achieving less chromatic selectivity (12.6 nm FWHM) while the blue and the green ones present similar values around 9.5 nm FWHM. A slight shift of the peak wavelengths is observed in Fig. 7, with a relative displacement below 1%.

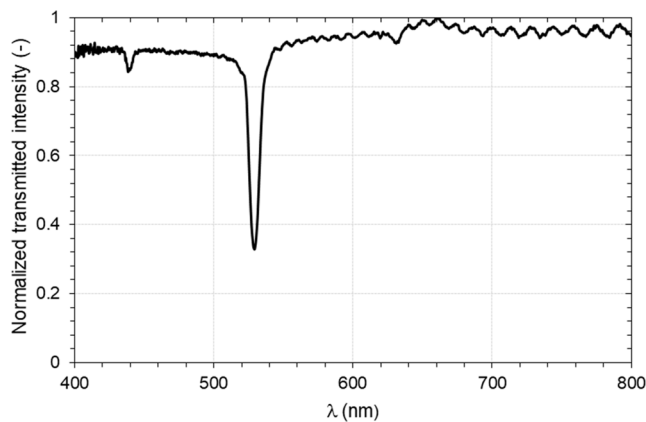


Fig. 8. Chromatic selectivity curve of an intermediate stage of the proposed model.

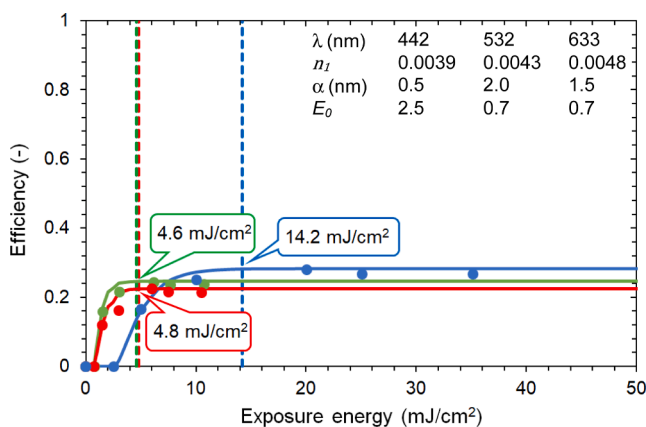


Fig. 9. Efficiency vs exposure energy for the three multiplexed wavelengths. Blue color corresponds to 442 nm laser and the green and red colors to 532 nm and 633 nm, respectively. The lines represent the theoretical tendency (Eq. (3)). For each wavelength, the optimal exposure energy values are indicated.

This effect is attributed to emulsion shrinkage. By using Eq. (2) and the values provided in Table 1, the emulsion layer thickness after processing is calculated, finding shrinkage values below 0.7% for the three wavelengths.

5.2. Polychromatic calibration

At the first stage of the polychromatic calibration, several tests were performed with different exposure times for every laser, keeping proportionality with the exposure energy values previously found in the monochromatic calibration for reaching 99% of the maximum theoretical efficiency (see Table 1). The lasers beam intensities were measured in the beginning of the polychromatic calibration experiments resulting in 0.33 mW/cm² for the blue, 0.52 mW/cm² for the green and 0.12 mW/cm² for the red. In all the tests performed, chromatic selectivity results revealed that the green was clearly dominant over the rest. This occurs because the green consumes the major part of the refractive index modulation, since the higher intensity promotes a faster polymerization and the main part of the monomer is involved. Fig. 8 illustrates this effect for exposure times of 57.5, 11.8 and 53.7 s for the blue, green and red lasers, respectively, and corresponding exposure energies of 19.0 mJ/cm², 6.1 mJ/cm² and 6.4 mJ/cm². In the case of the green laser, the diffraction efficiency achieved is 64% with respect to 2% for the blue and red cases.

This effect is mitigated by including a neutral density filter in the green optical path allowing the blue and red wavelengths to modulate

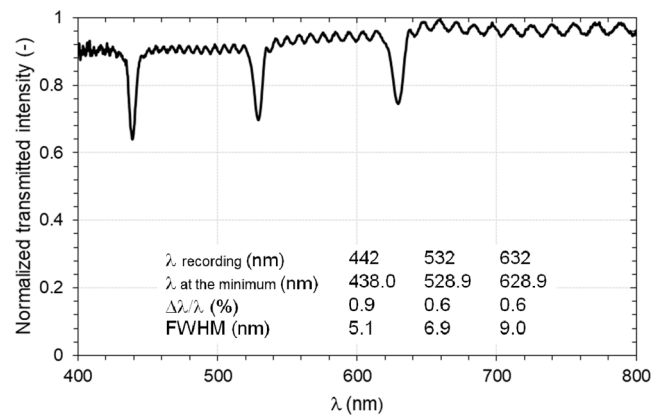


Fig. 10. Chromatic selectivity curve of the three multiplexed wavelengths.

the corresponding refractive index properly. After several tests, the optimal solution was to add a reflective neutral filter of density 0.7 to the green optical path in the experimental setup. Fig. 9 represents the efficiencies as a function of the exposure energy. The exposure times achieving the best and most uniform efficiencies were 43.1, 44.3 and 40.3 s for the blue, green and red (corresponding exposure energies of 14.2 mJ/cm², 4.6 mJ/cm² and 4.8 mJ/cm², respectively). As mentioned in Section 4, it can be seen that since the index modulation is shared between the recording wavelengths, the efficiency is also reduced accordingly. The figure also contains the modelled efficiency curves, demonstrating great agreement. In the embedded table (Fig. 9), the index modulation, the constant parameter of the exponential in the theoretical model and the exposure energy threshold are included.

Fig. 10 illustrates the chromatic selectivity of the most efficient multiplexed hologram. The efficiencies for the three wavelengths are quite similar: 28.1% for the blue, 24.4% for the green and 22.5% for the red ($\bar{\eta} = 25.0\%$ and $\sigma = 2.9\%$). The index modulation values for each wavelength (included in Fig. 9) are found to be very close to the optimal ones resulting from Eqs. (4) and (5), taking into account the maximum index modulation obtained in monochromatic recording (Table 1). This fact indicates that the proposed method performs satisfactory. The FWHM (Full Width at Half of the Maximum) for the three wavelengths is lower than 10 nm, which is considered enough for adequate color reproduction. Similarly to the monochromatic recording, the relative displacement in wavelength reported is lower than 1%, with a negligible effect on the full-color reproduction.

The fact that the optimal times (43.1, 44.3 and 40.3 s for blue, green and red lasers) are similar confirms that the competition between the gratings is a critical aspect to take into account in this type of recording panchromatic material. The polymerization begins at the moment when the threshold exposure is exceeded, and, as the results of the present research demonstrate, the kinetics of polymerization is strongly dependent on intensity, even though this dependence is different for each wavelength.

5.3. Example of a color hologram

Finally, the calibration data obtained in the previous section has been assessed recording a hologram of a colored object. A Playmobil® clown featuring all the basic colors has been chosen as the object. In this case, the object is not a perfect diffuser, which in principle can favor registration, but it does not have the same reflectance for all colors, which diminishes the relationship between the object and reference beams by lowering the contrast.

Two different methods have been used to record the hologram. Firstly, separate holograms for each color have been recorded on different Bayfol® HX sheets. All holograms will be recorded using the exposure energy necessary to reach 99% of maximum efficiency, data

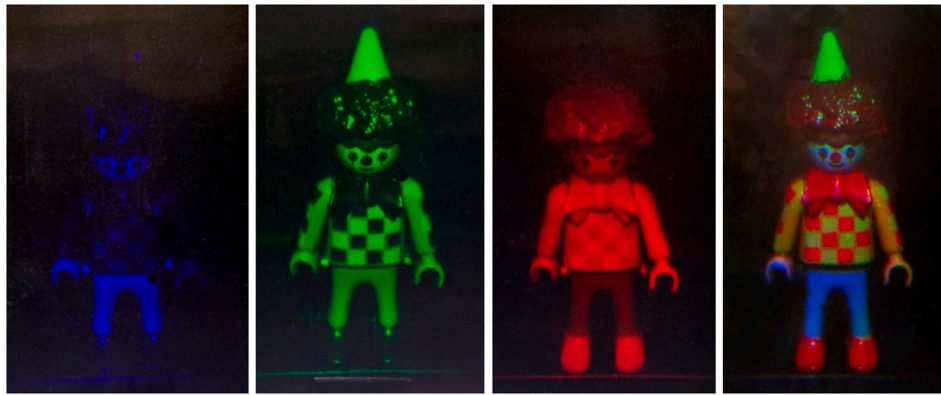


Fig. 11. From left to right, individual monochromatic holograms and stacking of the three.



Fig. 12. Comparison of the wavelength multiplexed hologram (left) and the real object (right).

given in Section 5.1. Once the individual holograms are made, they are overlapped trying to be perfectly aligned, obtaining the result shown in Fig. 11.

The main difficulty of this method is that three holograms need to be recorded and fine alignment is required to achieve a correct result. Colors appear highly saturated, with an unnatural appearance.

The hologram has also been recorded using the simultaneous exposure method with different times (Section 5.2). In Fig. 12, the result is shown, compared to a photograph of the actual object.

As it can be seen in Fig. 12, when using the proposed multiplexing method the color reproduction of the figure is almost perfect, except for a slight reddish tone that can be easily noticed in the clown's hands. This problem could be solved by a finer adjusting of the exposure times of each laser.

Comparing the results obtained, it is verified that with the superposition method a greater efficiency of the hologram is achieved, but the color reproduction is more faithful with the simultaneous exposure method.

6. Conclusions

An optimized wavelength multiplexing method for recording Denisyuk-type color reflection holograms of diffusing objects in Bayfol® HX photopolymer has been proposed and assessed. The procedure consists in a simultaneous opening of all the shutters but with different exposure time, depending on the wavelength, thus sequential shutters'

closing. The goal of the method is to maximize efficiencies while keeping them as uniform as possible.

Based on the literature and previous experience from the authors, three wavelengths were selected to adequately reproduce color: 442 nm (He-Cd laser), 532 nm (DPSS laser) and 633 nm (He-Ne laser). The photopolymer has been initially calibrated for monochromatic illumination and diffusing object (Spectralon® diffuser) resulting in diffraction efficiencies of 60%, 80% and 63% at 442 nm, 532 nm and 633 nm, respectively.

Once the maximum efficiencies and the associated refractive index modulation values were determined, the polychromatic calibration has been carried out. After several setup configurations, including neutral density reflective filters to adequate intensities to the photopolymer absorption characteristics, the best setup configuration was including a neutral density filter of 0.7 density in the green optical path and recording exposure energies of 14.2, 4.6 and 4.8 mJ/cm² for the blue, green and red colors, respectively. The efficiencies obtained for that setting are 28.1% for the blue, 24.4% for the green and 22.5% for the red. These values are close to maximum achievable efficiencies for a diffusing object and the uniformity between efficiencies is considered to be satisfactory. In the case that a better equalization of efficiencies is required for a specific application, finer control of exposure time or laser intensities would be needed. In addition, the FWHM of the diffracted spectra is very low (<10 nm), confirming high chromatic selectivity of the used setup and leading to a later reconstruction with negligible chromatic aberration.

Experimental values in the monochromatic and polychromatic calibrations have been compared to the corresponding theoretically modelled ones, achieving a very good agreement between both.

The maximum efficiencies obtained, for both monochromatic and polychromatic exposures, are centered at wavelength values almost identical to those recorded (less than 1% relative displacement). This confirms that the Bayfol® HX photopolymer experiences a negligible change in thickness during hologram recording.

Finally, a hologram of a Playmobil® clown has been recorded by the proposed method and also with monochromatic recording and subsequent stacking of the three holograms. In the case of monochromatic recording, the efficiencies obtained are higher but three holograms have to be recorded and a fine alignment between them is necessary. On the other hand, although the efficiency is lower, the multiplexed hologram demonstrates a higher level of color reproduction of the real object.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

The authors would like to thank Covestro Deutschland AG for supplying the recording photopolymer material. This research has been supported by the “Diputación General de Aragón-Fondo Social Europeo” (TOL research group, E44.17R), the “Generalitat de Catalunya” (2017FI_B2_00127 and 2017 SGR 1276) and the “Ministerio de Ciencia e Innovación” (PID2019-108598GB-I00). Daniel Chemisana thanks ICREA for the ICREA Academia award.

References

- [1] A. Sarakinos, A. Lembessis, N. Zervos, A transportable system for the in situ recording of color Denisyuk holograms of Greek cultural heritage artifacts in silver halide panchromatic emulsions and an optimized illuminating device for the finished holograms, *J. Phys.: Conf. Ser.* 415 (2013).
- [2] A. Sarakinos, A. Lembessis, Color holography for the documentation and dissemination of cultural heritage: Optoclones™ from four museums in two countries, *J. Imaging* 5 (2019).
- [3] H. Ohtaki, M. Watanabe, D. Kodama, F. Noujima, K. Ueda, Development of peripheral materials for color graphic arts holograms, *Proc. SPIE – Int. Soc. Optical Eng.* 3956 (2000) 245–252.
- [4] I. Naydenova, Holographic Sensors, *Opt. Hologr. Theory Appl.* 165–190 (2020).
- [5] D. Cody, S.-E. Gul, T. Mikulchik, M. Irfan, A. Kharchenko, K. Goldyn, S. Martin, S. Mintova, J. Cassidy, I. Naydenova, Self-processing photopolymer materials for versatile design and fabrication of holographic sensors and interactive holograms, *Appl. Opt.* 57 (2018) E173–E183.
- [6] Y. Zhao, K.-C. Kwon, M.-U. Erdenebat, S.-H. Jeon, M.-L. Piao, N. Kim, Implementation of full-color holographic system using non-uniformly sampled 2D images and compressed point cloud gridding, *Opt. Express* 27 (2019) 29746–29758.
- [7] P. Gentet, Y. Gentet, L.-H. Kim, K.-J. Kim, S.-H. Lee, Recording ultra-realistic full-color analog holograms for use in a moving hologram display, *J. Vis. Exp.* 2020 (2019).
- [8] S. Keshri, J. Marín-Sáez, I. Naydenova, K. Murphy, J. Atencia, D. Chemisana, S. Garner, M.V. Collados, S. Martin, Stacked volume holographic gratings for extending the operational wavelength range in LED and solar applications, *Appl. Opt.* 59 (2020) 2569–2579.
- [9] H. Bjelkhagen, D. Brotherton-Ratcliffe, *Ultra-Realistic Imaging: Advanced Techniques in Analogue and Digital Colour Holography*, 2016.
- [10] T. Kubota, Recording of high quality color holograms, *Appl. Opt.* 25 (1986) 4141–4145.
- [11] V.V. Shelkovich, E.V. Vasil'ev, V.V. Russkikh, L.V. Ektova, V.N. Berezhnaya, E.F. Pen, Monochrome and two-color holograms in layered photopolymer materials, *Optoelectron. Instrum. Data Process.* 52, 404–412 (2016).
- [12] M. Kawabata, A. Sato, I. Sumiyoshi, T. Kubota, Photopolymer system and its application to a color hologram, *Appl. Opt.* 33 (1994) 2152–2156.
- [13] H. Mukawa, K. Akutsu, I. Matsumura, S. Nakano, T. Yoshida, M. Kuwahara, K. Aiki, A full-color eyewear display using planar waveguides with reflection volume holograms, *J. Soc. Inf. Disp.* 17 (2009) 185–193.
- [14] H.I. Bjelkhagen, T.H. Jeong, D. Vukičević, Color reflection holograms recorded in a panchromatic ultrahigh-resolution single-layer silver halide emulsion, *J. Imaging Sci. Technol.* 40 (1996) 134–146.
- [15] I. Vázquez-Martín, M. Gómez-Climente, J. Marín-Sáez, M.V. Collados, J. Atencia, True colour Denisyuk-Type hologram recording in Bayfol HX self-developing photopolymer. *Proceedings of SPIE - The International Society for Optical Engineering*, 2017.
- [16] M.-L. Piao, N. Kim, Achieving high levels of color uniformity and optical efficiency for a wedge-shaped waveguide head-mounted display using a photopolymer, *Appl. Opt.* 2180–2186 (2014).
- [17] M.-L. Piao, K.-C. Kwon, H.-J. Kang, K.-Y. Lee, N. Kim, Full-color holographic diffuser using time-scheduled iterative exposure, *Appl. Opt.* 54 (2015) 5252–5259.
- [18] J. Upatnieks, C. Leonard, Efficiency and Image Contrast of Dielectric Holograms, *J. Opt. Soc. Am.* 60 (1970) 297–305.
- [19] H.I. Bjelkhagen, E. Mirlis, Color holography to produce highly realistic three-dimensional images, *Appl. Opt.* 47 (2008).
- [20] Y.N. Denisyuk, On the Reproduction of the Optical Properties of an Object by the Wave Field of Its Scattered Radiation. II, *Opt. Spectrosc.* 18 (1965) 152.
- [21] D. Jurbergs, F.-K. Bruder, F. Deuber, T. Fäcke, R. Hagen, D. Hönel, T. Rölle, M.-S. Weiser, A. Volkov, New recording materials for the holographic industry. *Proceedings of SPIE - The International Society for Optical Engineering*, 2009.
- [22] H. Berneth, F.K. Bruder, T. Fäcke, R. Hagen, D. Hönel, D. Jurbergs, T. Rölle, M.-S. Weiser, Holographic recording aspects of high-resolution Bayfol® HX photopolymer. *Proceedings of SPIE - The International Society for Optical Engineering*, 2011.
- [23] J. Marín-Sáez, J. Atencia, D. Chemisana, M.-V. Collados, Characterization of volume holographic optical elements recorded in Bayfol HX photopolymer for solar photovoltaic applications, *Opt. Express* 24 (2016) A720–A730.
- [24] H. Kogelnik, Coupled Wave Theory for Thick Hologram Gratings, *Bell Syst. Tech. J.* 48 (1969) 2909–2947.
- [25] F.-K. Bruder, T. Fäcke, T. Rölle, The chemistry and physics of Bayfol® HX film holographic photopolymer, *Polym. (Basel)*. 9 (2017).