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Review

Employing Glycerol for Improving Diffraction Efficiency, Photosensitivity and Pressure Sensitivity in Holographic Recording Layers

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Abstract

The aim of this review is to explore the improvement in diffraction efficiency, photosensitivity and pressure sensitivity in holographic materials containing glycerol. Glycerol is a well-known, non-toxic, water-soluble polyol compound. Glycerol polymers have attracted increased attention recently due to the diversity of the available compositions. Glycerol provides access to a range of monomers for subsequent polymerizations. Various glycerol containing polymers, including polyvinyl alcohol films, polyesters, polyethers and polycarbonates, have been investigated for different applications. It was discovered in 2009 that the addition of glycerol to the composition of water-soluble holographic photopolymers facilitates the faster formation of holograms due to greater photosensitivity. It was also discovered that the presence of glycerol in holographic photopolymers makes them highly pressure-sensitive. A new family of holographic photopolymer materials, containing glycerol and capable of recording holograms with bright reflections, was reported. The novel photopolymers are composed of glycerol, a polymeric binder, a crosslinking monomer, an initiation system, and sensitising dyes. No wet-processing of holograms is necessary. Each holographic photopolymer film contains bis-acrylamide (BA) monomer in polyvinyl alcohol matrix, triethanolamine and methylene blue dye solution, glycerol and water. It was shown that the new holographic material is capable of reaching a refractive index modulation matching that of the well-known acrylamide photopolymer material, but more quickly. The new holographic photopolymer materials are cheap and environmentally friendly. The use of glycerol to improve diffraction efficiency, photosensitivity and pressure sensitivity in holographic recording layers continues to expand. This review describes the development and applications of glycerol-containing photopolymer materials. An environmentally friendly diacetone-based photopolymer was developed. The positive effect of glycerol on N-vinylpyrrolidone photopolymer was investigated. Finally, potential opportunities for future research in the area of glycerol-containing photopolymers are outlined.

Keywords: holographic technologies; glycerol-containing photopolymers; low-toxicity holographic photopolymers; water-soluble polymers; bright interactive holograms

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1. Introduction

Holographic photopolymers are very attractive media for recording holograms. These photopolymers are low-cost and self-processing materials and easy to produce on an industrial scale.

The objective of this work is to present a review of glycerol-containing holographic photopolymers. Glycerol is well-known as a plasticizer for photopolymers because it reduces polymer–polymer chain bonding. This plasticizing effect allows for the faster diffusion of monomers during the photopolymerization process, which is a key factor in achieving high-quality holographic recording. To the best of our knowledge, this is the first review of holographic photopolymers containing glycerol.

In 1987, Calixto [1] developed an acrylamide-based (AA-based) holographic recording material, based on acrylamide (AA) monomer, polyvinyl alcohol (PVA), triethanolamine (TEA) and the photosensitive dye methylene blue. Fimia et al. [2,3] developed a method for increasing the sensitivity of acrylamide-based holographic photopolymers and reported an increase in diffraction efficiency (DE) up to 40% for diffraction gratings with a spatial frequency of 1000 lines/mm [2,3]. The photopolymer composition reported by Calixto [1] was optimised by Martin et al. [4] for recording at a wavelength of 514 nm: a crosslinking monomer bisacrylamide was included in the photopolymer composition, and a xanthene dye was used as a photosensitiser. Diffraction efficiencies of 80% and above were obtained with an exposure energy of 80 mJ/cm^2. Blaya et al. [5] continued the development of the acrylamide photopolymer for recording at 633 nm by using a different crosslinking monomer: N,N′-dihydroxiethylenbisacrylamide [5,6].

Interest in the optical properties of novel materials for sensing applications has grown considerably during the last decade because of their possible applications in laser technologies, eco-technologies, medicine and other areas [7–10]. There is a considerable interest in developing photopolymers [11–16] since they have many industrial applications [17–19]. The development of holographic photopolymers with high diffraction efficiency is of great importance. That is why much of the recent research on holographic photopolymers is focused on novel photopolymer compositions with high diffraction efficiency, and suitability for industrial applications, such as pressure, humidity, temperature and other types of sensors.

Photo-polymerization is the reaction in which monomer is converted to polymer upon exposure to light. Photo-polymerization occurs in regions where constructive interference takes place between two coherent light beams. Areas where there is destructive interference remain unchanged. The refractive index of the material changes where photopolymerization has occurred, producing a hologram. When the hologram is illuminated with a laser beam, diffraction of the beam occurs due to the modulation in the refractive index of the polymer material. Figure 1 shows a schematic diagram of the holographic recording mechanism.

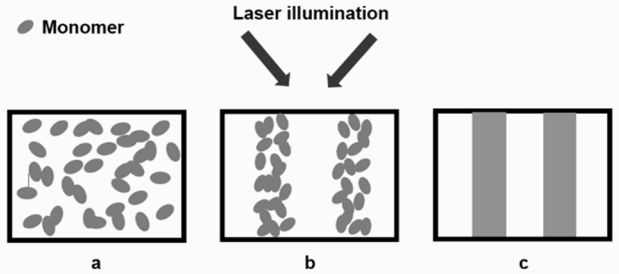


Figure 1. Holographic recording mechanism in a photopolymer layer. (**a**) homogeneous photopolymer mixture; (**b**) monomer diffusion and crosslinking in the illuminated areas; (**c**) refractive index modulation.

The most important characteristics of a holographic material are the diffraction efficiency (η) and refractive-index modulation Δn.

The diffraction efficiency (η) is a physical quantity which characterises the quality of the hologram. It is obtained by illuminating the hologram/grating with the laser beam and measuring the intensity of the first-order diffracted beam using a power metre. The following formula is then used:

$$\eta(\%) = \frac{\text{Intensity of Diffracted Beam}}{\text{Intensity of Probe Beam}} \quad (1)$$

The Refractive Index Modulation (RIM) is the difference between the refractive index of the polymer in areas where it has been exposed to light and in areas in which it has not. RIM is the most important parameter for comparing the ability of different photopolymers to record bright holograms with high diffraction efficiency. The application of Kogelnik's coupled-wave theory for volume, thick gratings [20], gives the refractive index modulation Δn, as follows:

$$\Delta n = \frac{\lambda \cos\theta \sin^{-1}(\sqrt{\eta})}{\pi d} \quad (2)$$

λ = wavelength of laser.
θ = reading beam incidence angle.
η = diffraction efficiency.
d = sample thickness.

It was discovered in 2009 that the addition of glycerol to the composition of water-soluble holographic photopolymers facilitates the faster formation of holograms due to their greater photosensitivity [21]. We will review the state of development of glycerol-containing holographic-recording layers until the present. We will also focus on some important practical applications of glycerol-containing photopolymers.

2. Photopolymer Materials Containing Glycerol

2.1. Bisacrylamide (BA)-Based Holographic Photopolymer

2.1.1. Composition of the BA-Based Holographic Photopolymer

A family of holographic photopolymer materials capable of recording of bright transmission and reflection holograms is presented. It is shown that the photopolymer composition has holographic recording characteristics like those of the well-known acrylamide-based photopolymer [4], but with reduced toxicity due to the absence of acrylamide [22–26]. These modified polymer materials are non-toxic and have high photosensitivity. Their self-processing nature, together with their low production cost, make them an excellent candidate for different applications, such as holographic sensors, diffractive optics and security holograms.

The BA-based photopolymer is composed of a polymeric binder, one crosslinking monomer, an initiation system, glycerol and sensitising dyes. Polyvinyl alcohol (PVA) acts as a binder matrix for the other components. PVA is a non-toxic polymer with excellent adhesive and emulsifying properties. N, N'-methylene bisacrylamide (BA) is a cross-linking monomer. Triethylamine (TEA) is the free radical initiator, which generates free radicals upon exposure to laser light. Methylene Blue Dye is the photosensitive dye used in the photopolymer mixture. It is excited on exposure to light to produce oxidising radicals.

2.1.2. Preparation of Photopolymer Layers to Record Reflection Gratings

The aim of our experimental work was to prepare a holographic photopolymer without acrylamide and to analyse its ability to record reflection gratings. Three photopolymer compositions were prepared, as shown in Table 1. The photopolymer solutions were deposited on to glass slides and allowed to dry for 24 h in darkness under normal laboratory conditions (21–24 °C, 40%–60% relative humidity).

Table 1. N, N′-Methylene bisacrylamide photopolymer compositions.

Component	Function	Photopolymer BA1	Photopolymer BA2	Photopolymer BA3
N, N′-Methylene bisacrylamide (BA) (g)	Monomer/ Cross-linker	0.4	0.3	0.3
Polyvinyl alcohol (PVA) 10% w/v (mL)	Binder	20	20	20
Triethanolamine (TEA) (mL)	Free radical generator	2	1	1
Methylene Blue dye (MB) 0.11% w/v (mL)	Sensitising dye	4	4	2
Glycerol (mL)	Plasticizer/Free radical scavenger	2	2	1

2.1.3. Experimental Set-Up

Reflection gratings were recorded using a standard optical set-up, where the object is a mirror. The intensities of the two beams illuminating the holographic photopolymer layer from opposite sides were equal. The results from these measurements were used to calculate the refraction index modulation.

Denisyuk reflection holograms of two metal objects (coins) were recorded using the optical set-up shown schematically in Figure 2.

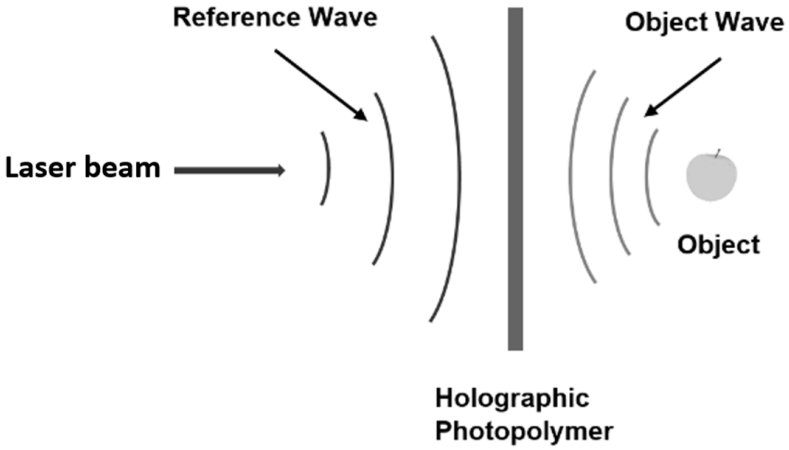


Figure 2. Recording a Denisyuk reflection hologram.

2.1.4. Results and Discussion

1. Refraction Index Modulation

The RIM (Equation (2)) for the three tested photopolymers was calculated to compare the qualities of different photopolymers in terms of their ability to record bright holograms. Figure 3 presents the results for the RIM of the new photopolymers.

Figure 4 presents the results for the RIM of the well-known acrylamide photopolymer, whose composition is reported in [4].

A refractive index modulation higher than that for the well-known acrylamide holographic photopolymer was achieved for the photopolymer composition BA1 in Table 1. This indicates that the BA-based photopolymer can record bright reflection holograms using relatively short exposure times. This may be due to glycerol's property as a plasticizer, which allows for the faster diffusion of unreacted cross-linking monomer within the

photopolymer during holographic recording. The photopolymer layers containing glycerol improved stability and optical quality.

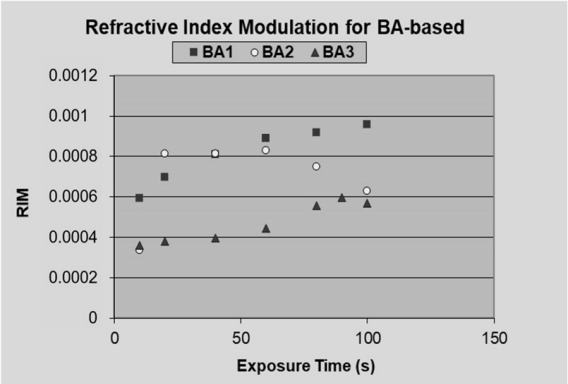


Figure 3. Refractive Index Modulation for the three BA-based photopolymers.

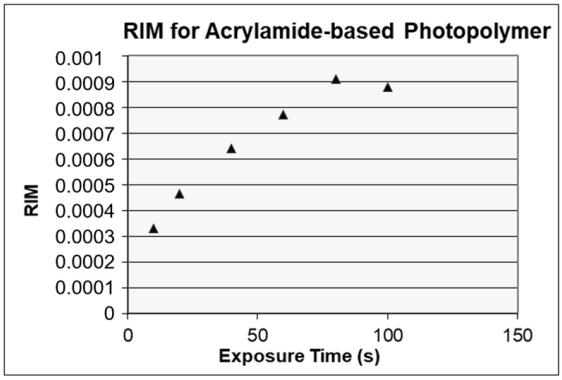


Figure 4. Refractive Index Modulation for an acrylamide-based photopolymer.

2. Recording of Reflection Holograms in a BA-Based Photopolymer

The BA1 photopolymer composition can record reflection and transmission holograms with high diffraction efficiency (DE). Some examples of Denisyuk reflection holograms, recorded in BA1 photopolymer, are shown in Figures 5 and 6.

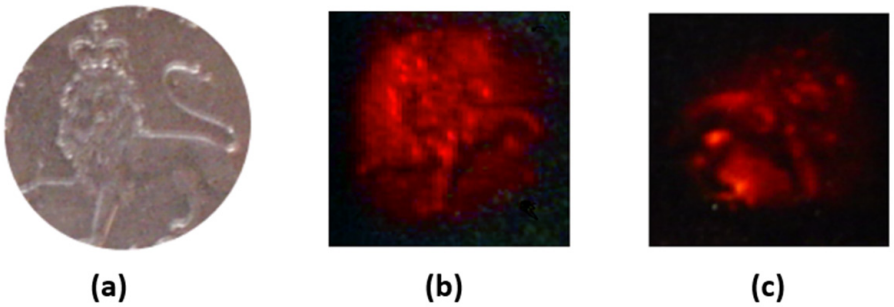


Figure 5. Reconstructed images from a Denisyuk type hologram, recorded using a He-Ne laser (λ = 633 nm): (**a**) the object; (**b**) reflection hologram 1 (recorded through interference from the reference beam and the object beam); (**c**) reflection hologram 2 (recorded through interference from the reflected reference beam and the reflected object beam).

2.1.5. Summary

The aim of this study was to prepare a holographic photopolymer without acrylamide and to analyse its ability to record reflection holograms. Three different compositions of the novel BA-based photopolymer (BA1, BA2 and BA3) were prepared and investigated.

The photopolymer BA1 has a refractive index modulation higher than that of well-known acrylamide-based photopolymers. The photopolymer BA2 reaches a refractive index modulation matching that of the standard acrylamide photopolymer material, but 40 seconds quicker. This indicates that this photopolymer can record bright-reflection holograms that are visible in diffuse light using relatively short exposure times. This result could be very useful for industrial applications, as shorter recording times would mean lower manufacturing costs.

A very attractive feature of the new family of BA-based photopolymers is their low-toxicity, making them very suitable for industrial applications.

The results indicate high potential for the further optimisation of the new photopolymer materials to achieve an even higher RIM.

2.2. Diacetone Acrylamide-Based Photopolymer

2.2.1. Composition of the DA-Based Holographic Photopolymer

A low-toxicity photopolymer was developed, with the main monomer being diacetone acrylamide (DA) [11,27,28].

Cody et al. optimised the composition of a DA-based holographic photopolymer, with a DE of 50% [16], to record reflection holograms. Figure 6 shows a photograph of a Denisyuk hologram of a 10-cent coin, recorded in a photopolymer with the chemical composition given in Table 2.

Table 2. Chemical composition of the DA-based holographic photopolymer.

Component	Function	Quantity
Diacetone-acrylamide (g)	Monomer	1
N, N′-Methylene bisacrylamide (BA) (g)	Monomer/cross-linker	0.2
Polyvinyl alcohol (PVA) 10% w/v (mL)	Binder	20
Triethanolamine (TEA) (mL)	Free radical generator	2
Methylene Blue dye (MB) 0.11% w/v (mL)	Sensitising dye	4
Glycerol (mL)	Plasticizer/free radical scavenger	2



Figure 6. Denisyuk reflection hologram of a 10-cent coin recorded in a DA-based photopolymer, illuminated in white light (reprinted from [28]).

2.2.2. Holographic Pressure Sensors in a DA-Based Photopolymer

Mihaylova et al. developed holographic pressure sensors [29]. It was found that Denisyuk holograms recorded in the DA-based photopolymer can reveal pressure spatial

distribution as colour maps. Pressure-sensitive holograms have potential for various medical and industrial applications, as listed in Figure 7 [29].



Figure 7. Applications requiring a thin film capable of tactile pressure measurements (reprinted from [29]).

The principle of operation (Figure 8) of the holographic pressure sensor is that the colour of the image reconstructed with the white light reflection hologram depends on the applied pressure, because the fringe spacing of the hologram changes under pressure. This change leads to a change in the colour of the reconstructed image seen by the observer (Figure 8).

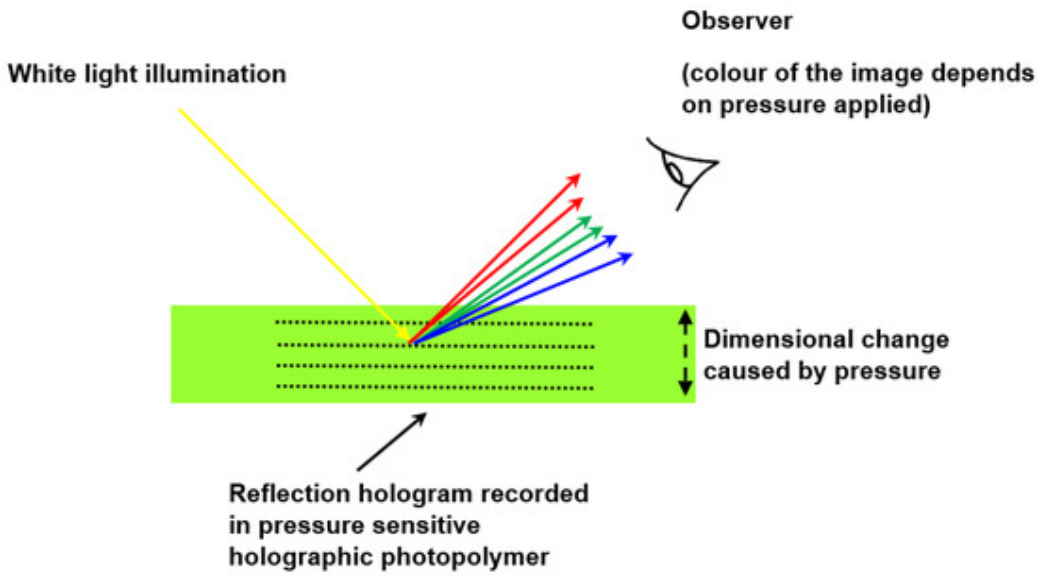


Figure 8. Principle of operation of a holographic pressure sensor (reprinted from [29]).

Figure 9 shows some examples of reflection holograms that were subjected to pressure. The holograms were recorded with a red He–Ne laser at a wavelength of 633 nm, so when reconstructed with white light, the image is red. It can be seen that when the applied pressure increases, the colour of the reconstructed image changes from red to yellow, through to green and then blue.

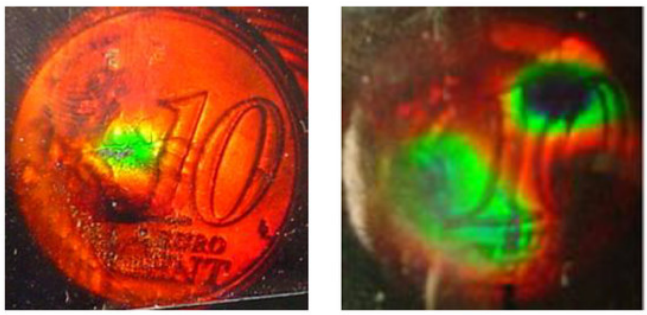


Figure 9. Denisyuk reflection holograms of a 10-cent coin subjected to pressure. The pressure applied was: (**a**) 50 N/cm^2 for the green spot; (**b**) 70 N/cm^2 for the dark blue spot (reprinted from [29]).

2.2.3. Summary

An optimised composition of a DA-based holographic photopolymer is reported with a significant improvement in holographic recording ability compared with other water-soluble, low-toxicity photopolymer materials [12–15,17]. Remarkably, the DA-based holographic layers can record reflection holograms, having a diffraction efficiency of 50% [16], the highest reported DE for a reflection hologram, recorded in photopolymer material. Interestingly, the sensitivity of this material to pressure can be controlled by changing the quantity of the plasticizer glycerol. This makes it possible for pressure sensors with different specifications to be fabricated. Another advantage of this holographic photopolymer is that it can be used for the development of new types of colour holograms for versatile displays.

2.3. N-Vinylpyrrolidone-Based Photopolymer

Huishi Pi et al. reported their work on the effect of glycerol on the holographic performance of an N-Vinylpyrrolidone-based Photopolymer [30]. NVP (N-vinylpyrrolidone) has a comparatively large molecular structure, making diffusion difficult during holographic recording. Glycerol was added to the original NVP-based photopolymer composition to overcome the slow-diffusion problem. The holographic performance of the photopolymer with glycerol was improved at low spatial frequencies. It was found that with the increase in glycerol concentration, the holographic performance of the NVP-based photopolymer first increases and then remains stable. Using the optimised holographic photopolymer, the authors recorded bright transmission holograms with a diffraction efficiency (DE) of 84% at spatial frequencies of 800 lp/mm.

2.3.1. Preparation of N-Vinylpyrrolidone-Based Photopolymer Layers with Different Concentrations of Glycerol

The optimised photopolymer has the following composition: polyvinyl alcohol, tetraiodofluorescein sodium, triethanolamine, N,N′-methylenebisacrylamide and NVP as a liquid [30]. The liquid photopolymer was divided into six equal parts, and a different amount of glycerol was added to each part. The liquid photopolymer was coated on glass slides and dried in a dark room. The final photopolymer layers were of good optical quality [30].

2.3.2. Effect of Glycerol on Holographic Parameters

The holographic parameters of the six photopolymer compositions, containing different amounts of glycerol, are shown in Figure 10. The effect of the glycerol addition is clearly seen: the maximum DE, maximum refractive index modulation and photosensitivity of the photopolymer all increased.

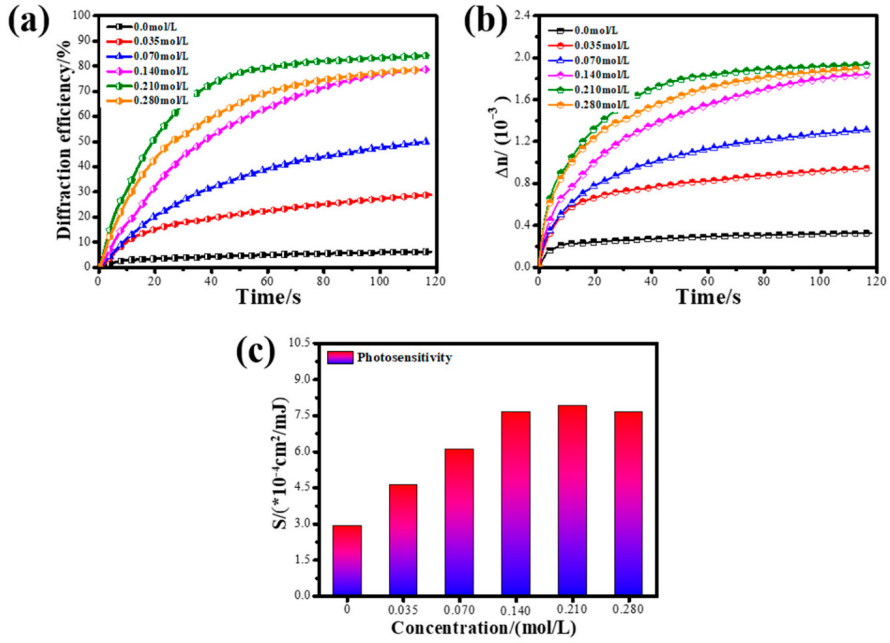


Figure 10. Effect of glycerol on the holographic performance of photopolymers: (**a**) curve of real-time diffraction efficiency; (**b**) curve of refractive index modulation; (**c**) photosensitivity (reprinted from [30]).

Figure 11 shows the schematic diagram of the internal refractive index distribution of the photopolymer. The authors assumed a uniform distribution of the components of the N-Vinylpyrrolidone-based photopolymer before exposure.

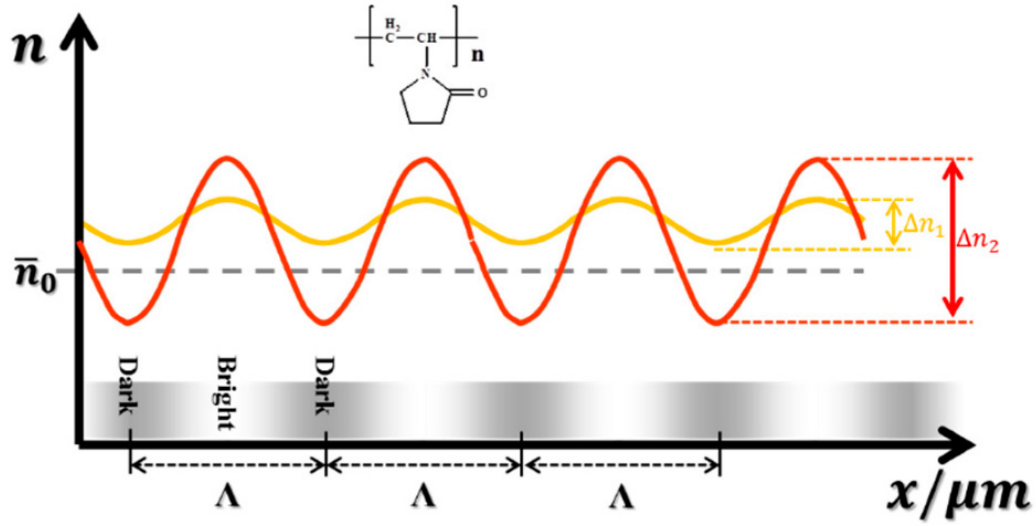


Figure 11. Schematic diagram of the internal refractive index distribution of photopolymer: n_0 is the average refractive index of the unexposed photopolymer material; Λ is the grating period; the yellow line represents the photopolymer without glycerol; the red line represents the photopolymer with glycerol, reprinted from [30]).

2.3.3. Summary

An N-Vinylpyrrolidone-based photopolymer containing glycerol was developed. It was shown experimentally that the NVP-based photopolymer, with the addition of 0.21 mol/L glycerol, has an excellent holographic performance. The maximum DE, maximum refractive index modulation and photosensitivity of the optimised photopolymer all increased by 11.19 times, 4.69 times and 1.71 times, respectively.

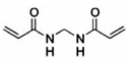
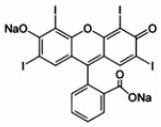
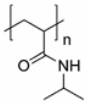
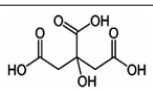
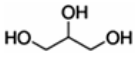
2.4. N-Isopropylacrylamide (NIPA) Photopolymers for Holographic Recording

2.4.1. Composition of Thermosensitive NIPA Photopolymer for Holographic Recording in Transmission and Reflection Modes

A NIPA-based photopolymer composition (Table 3) for holographic recording in transmission and reflection modes was developed [17]. Acrylamide was replaced with low-toxicity N−isopropylacrylamide. Holographic performance of the NIPA-based photopolymer in both transmission and reflection modes was characterised and the optimum recording conditions at different spatial frequencies of recording were identified. It is reported that the NIPA-based photopolymer can be used for fabrication of holographic temperature sensors. The holographic temperature sensors can operate in reflection or transmission mode [17].

In transmission gratings, the diffraction efficiency was unchanged within 14 days and a 30% decrease was noticed in 21 months. In reflection gratings, a few percent decay of the diffraction efficiency was observed within 24 h of the recording. After 41 days, a 20% decrease was noted. To prevent the decay in diffraction efficiency, reflection gratings were treated using a Dymax UV curing system (model ECE-200). After UV curing with a total exposure of 5.4 J/cm^2, a decrease of a few percent was observed within 41 days of the recording. Thus, the addition of glycerol followed by UV-curing enabled the holographic recordings in the NIPA-based photopolymer to be stabilized [17].

Table 3. Thermo-sensitive NIPA photopolymer compositions (reprinted from [17]).

Chemical Reagent	Chemical Compound	Transmission Mode	Reflection Mode
Polyvinyl alcohol		8.79% w/v	8.42% w/v
N-phenylglycine		0.0145 M	0.0139 M
N,N′-methylene bisacrylamide		0.053 M	0.051 M
Erythrosin B		1.37×10^{-4} M	1.32×10^{-4} M
N-isopropyl acrylamide		0.097 M	0.093 M
Citric acid		-	0.022 M
Glycerol		0.15 M	0.72 M

2.4.2. Temperature Response

1. Transmission Gratings

The thermal response of transmission gratings with spatial frequency 1000 lines/mm was investigated in the temperature range from 18 to 47 °C at a relative humidity of 60% [17]. With the increase in the temperature, the diffraction efficiency increases at temperatures below 30 °C and decreases above 30 °C. The transmission holograms produced in this photopolymer composition are suitable for the development of holographic temperature

sensors. This composition was subsequently optimised. Figure 12 shows the compositions (A, B, and C) of the three photopolymers that were used in [31] for the fabrication of temperature-sensitive transmission gratings—A and B—and for the reflection hologram recording—composition C (see the second part of Section 2.4.2). Composition A is a modification of an AA photopolymer with the inclusion of glycerol.

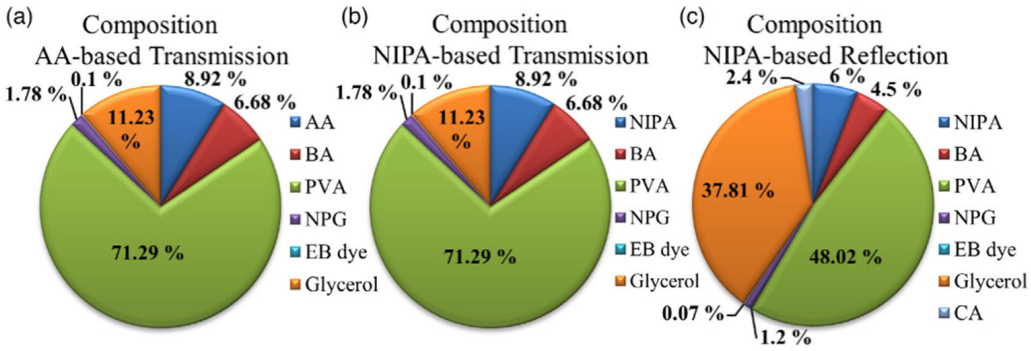


Figure 12. The chemical composition of dry layers of AA- and NIPA-based photopolymers for the fabrication of a holographic sensor (reprinted from [31]).

2. Reflection Gratings

The temperature response of reflection gratings is similar to that of the transmission gratings [31]. Composition C has a higher percentage of glycerol than A and B, which enhances the recording performance of the photopolymer in reflection mode.

The effect of exposure to elevated temperature on the colour change in the Denisyuk reflection holographic gratings recorded in the NIPA-based photopolymer (composition C) is discussed in this section. By exposing the holograms to the same temperature range studied in transmission holograms, the authors investigated the colour change and its reversibility. Colour changes in reflection holograms can indicate swelling and shrinkage of the layers. Denisyuk reflection gratings were selected for temperature study as their response can be observed after illumination with a white light source; therefore, they are comparatively easy to use. Without the need for an additional electronic read-out, one can observe, using the naked eye, the temperature-induced changes in the layer.

Photographs of the reconstructed images of the object [percentage sign (%)] and the results from the temperature study of the recorded Denisyuk reflection holographic gratings are shown in Figure 13. After recording with laser light of 532 nm wavelength, and then bleaching the remaining photoinitiator with white LED light, samples were exposed to a temperature cycle, as described in Figure 13**.** This study on reflection gratings was conducted to double-check the observed switchable memory effect in transmission gratings exposed to elevated temperature.

The temperature increase from 18 °C to 60 °C causes a colour change in the hologram. This is well-suited to the application of reflection holograms as visual temperature indicators that visibly change colour under temperature changes.

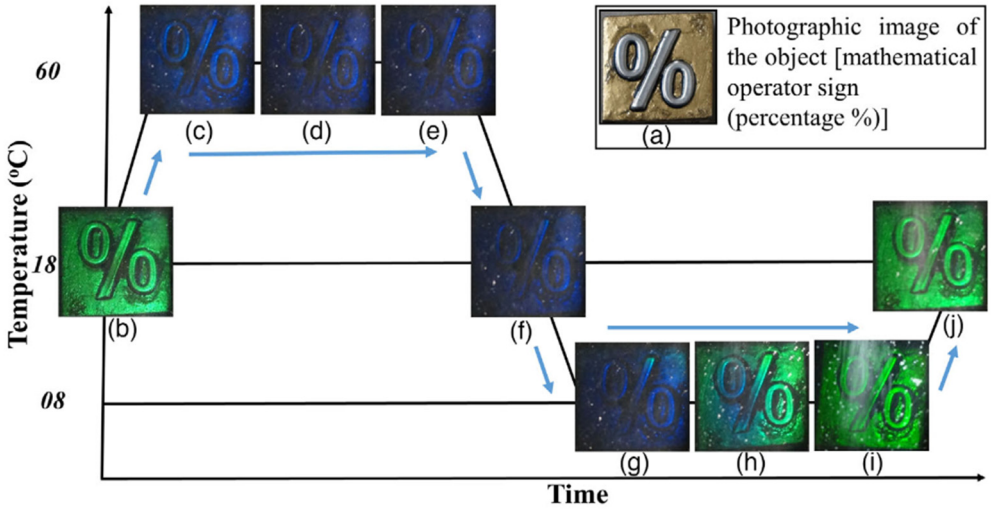


Figure 13. Temperature response of Denisyuk reflection holographic gratings recorded in NIPA-based photopolymer and the respective reflection peaks from their spectral response. (**a**) A photograph of the object; (**b**) hologram at 18 °C; (**c**) hologram as the temperature just reached 60 °C; (**d**) after 10 min at 60 °C; (**e**) after 20 min at 60 °C; (**f**) hologram at 18 °C during the temperature decrease; (**g**) hologram as the temperature just reached 8 °C; (**h**) after 10 min at 8 °C; (**i**) after 20 min at 8 °C; and (**j**) at 18 °C, at the end of the temperature cycle. The layer thickness is 60 μm (reprinted from [31]).

2.4.3. Summary

A NIPA-based photopolymer composition for holographic recording in transmission and reflection modes was investigated [17,31]. Glycerol was used in the photopolymer compositions to improve the exposure sensitivity and the stability of recorded photonic structures. It was found that the temperature-induced changes in the diffraction efficiency of transmission gratings and the wavelength shift in reflection gratings are reversible to within 2% and 5 nm, respectively.

2.5. A Magnetic Nanoparticle-Doped Photopolymer for Holographic Recording

2.5.1. Composition of the Photopolymer [32]

The composition of the magnetic photopolymer is given in Table 4. All three photopolymer compositions contain glycerol for recording reflection holograms with high diffraction efficiency, high refractive index modulation and high photosensitivity of the photopolymer.

Table 4. Composition of dry layer samples (C, D, E). The dry-layer samples, (C)-pure NIPA-based photopolymer and (D)-magnetic nanocomposite (NIPA-based photopolymer doped with MNPs), are used for recording volume transmission holographic gratings, while sample (E)-magnetic nanocomposite (NIPA-based photopolymer doped with MNPs) is used for recording Denisyuk reflection holographic gratings (reprinted from [32]).

Function/Role	Chemical Component *	Dry Layer Samples		
		(C)-Trans Pure NIPA	(D)-Trans Nanocomposite (MNPs in NIPA)	(E)-Reflect Nanocomposite (MNPs in NIPA)
Binder	PVA 10% wt./vol, mL	16	16	16
Monomer	NIPA, g	0.2	0.2	0.2
Cross-linker	BA, g	0.15	0.15	0.15
Free-radical generator	NPG, g	0.04	0.04	0.04

Table 4. *Cont.*

Function/Role	Chemical Component *	Dry Layer Samples								
		(C)-Trans Pure NIPA	(D)-Trans Nanocomposite (MNPs in NIPA)				(E)-Reflect Nanocomposite (MNPs in NIPA)			
Sensitising dye	Er B 0.11% wt./vol, mL	2	2				2			
Plasticiser/free radical scavenger	Glycerol, mL	0.2	0.2				1			
Chain transfer agent	CA, g	-	-				0.08			
magnetic nanoparticles (MNP)	Fe_2O_3 Alpha 20–30 nm		concentrations of MNPs in dry layer (% wt./wt.)							
		-	0.5	1	2	5	10	0	0.5	1
	Fe_3O_4 20 nm	-	0.5	1	2	5	10		-	

* Polyvinyl alcohol (PVA), N-isopropyl acrylamide (NIPA), N,N-Methylene bisacrylamide (BA), N-Phenylglycine (NPG), Erythrosine B (Er B), Glycerol, citric acid (CA), Maghemite (Fe_2O_3), Magnetite (Fe_3O_4).

2.5.2. Denisyuk Reflection Gratings

The authors record Denisyuk reflection holographic gratings in pure-NIPA and magnetic nanoparticle-doped material (composition E) with laser light from a frequency doubled Nd:YVO_4 laser (532 nm).

Figure 14 presents photographs of the object (mathematical operator percentage sign (%) and the Denisyuk reflection holograms recorded in the pure NIPA-based photopolymer layer and magnetic nanocomposite layers with two different concentrations of 0.5% wt./wt. and 1% wt./wt. Fe_2O_3 alpha MNPs. The Denisyuk reflection holograms can be observed during illumination with a white light source.

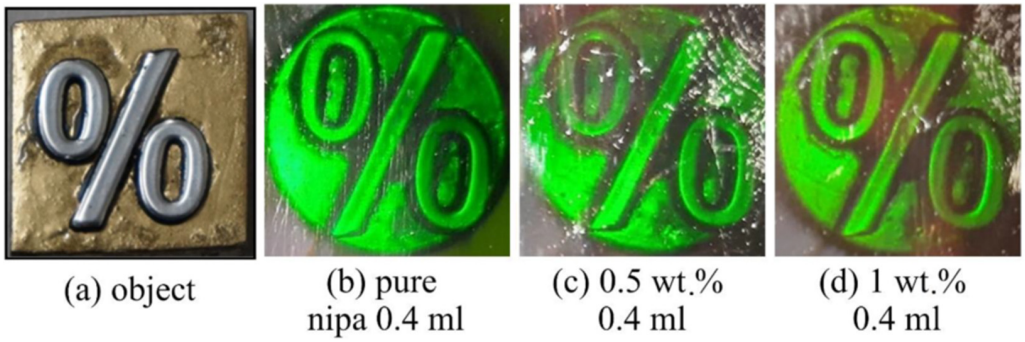


Figure 14. Results of Denisyuk reflection recording in NIPA-based photopolymer and magnetic nanocomposites doped with MNPs Fe_2O_3 Alpha (average size 20–30 nm). (**a**) A photograph of the object, (**b**) hologram recorded in pure NIPA layer (0% wt./wt. concentration of MNPs), (**c**) hologram recorded in magnetic nanocomposite layer having 0.5% wt./wt. concentration of MNPs, (**d**) hologram recorded in magnetic nanocomposite having 1% wt./wt. concentration of MNPs. The layer thickness was about 44 ± 5 μm (reprinted from [32]).

Figure 14b shows that a clearer and brighter hologram was recorded in the pure NIPA-based photopolymer layer. Figure 14c,d show that the holographic recording of reflection holograms is possible in a magnetic nanocomposite photopolymer. The lower quality of the Denisyuk holograms, recorded in the nanocomposite layers, is most probably due to the increased scattering in the material. In any case, the inclusion of glycerol in the photopolymer composition improved the quality of the recorded holograms.

2.5.3. Summary

The authors reported [32], for the first time, the recording of reflection holograms in NIPA-photopolymer doped with MNPs. A maximum diffraction efficiency of up to 30% was achieved in the NIPA-photopolymer without additives. The diffraction efficiency of the layers doped with MNPs decreased when the concentration of the nanoparticles increased. The results reported by the authors [32] confirm that holographic recording is possible in photopolymers containing MNPs, and show that such magnetic photopolymer nanocomposites can be heated locally through induction heating using an alternating magnetic field (AMF).

In general, the addition of glycerol to water-soluble holographic photopolymers makes it easier to incorporate different additives to modify the properties of the material for different applications.

3. Conclusions

The aim of this review was to explore the improvement in diffraction efficiency, photosensitivity and pressure sensitivity in holographic materials containing glycerol. It was found in 2009 that the addition of glycerol to the composition of water-soluble holographic photopolymers facilitates the faster formation of holograms with higher levels of photosensitivity. The presence of glycerol in holographic photopolymers makes the recorded holograms highly pressure-sensitive and allows for the development of new types of pressure sensors—namely, holographic pressure sensors. A new family of holographic photopolymer materials, containing glycerol and capable of recording bright-reflection holograms, was reported. No wet-processing of holograms is necessary. The holographic photopolymer film contains a bis-acrylamide (BA) monomer in polyvinyl alcohol matrix, triethanolamine and a dye solution: methylene blue, glycerol and water. It was shown that the new holographic material achieves a refractive index modulation matching that of the well-known acrylamide photopolymer material, but is faster. The new holographic photopolymer materials are cheap and environmentally friendly. The addition of glycerol to the standard acrylamide-based holographic photopolymer also makes the recorded hologram responsive to pressure. The use of glycerol for improving diffraction efficiency, photosensitivity and pressure sensitivity in holographic recording layers continues to expand. This review described the development and applications of the glycerol-containing holographic photopolymer materials known at present. Potential opportunities for future research developments in glycerol-containing photopolymers are outlined in the review.

4. Future Perspectives

Interest in holographic photopolymers for different technological applications is increasing. The most widely researched at present are photopolymerizable acrylamide (AA/PVA) compositions [33,34] with the inclusion of different additives, aiming to obtain materials with new properties. These additives include liquid crystals [35,36], inorganic nanoparticles [37–39], and materials with diffusion-based formation mechanisms [40,41]. This review highlights the published papers showing that the addition of glycerol to bisacrylamide-based photopolymers and to diacetone acrylamide-based photopolymers greatly improve the diffraction efficiency and photosensitivity. Another big advantage of the addition of glycerol to photopolymers is that the incorporation of different nanoparticles and other new possible additives is easier to achieve. It seems likely that all known acrylamide-based photopolymers will benefit from the addition of glycerol to their compositions. The same is true for bisacrylamide-based photopolymers and diacetone acrylamide-based photopolymers when developed for new industrial applications, such as data stor-

age [13,42,43], interactive holograms [18], brand protection and authentication [19], holographic solar concentrators [44–46] and holographic lenses [47].

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