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Silicon oxide nano-particles doped PQ-PMMA for volume holographic imaging filters

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Abstract

Holographic imaging filters are required to have high Bragg selectivity to obtain spatial-spectral information within a three-dimensional object. In this Letter, we present the design of holographic imaging filters formed using silicon oxide nano-particles (nano-SiO₂) in PQ-PMMA polymer recording material. This combination offers greater angular and spectral selectivity and increases the diffraction efficiency of holographic filters. The holographic filters with optimized ratio of nano-SiO₂ in PQ-PMMA can significantly improve the performance of Bragg selectivity and diffraction efficiency by 53% and 16%, respectively. We present experimental results and data analysis demonstrating this technique in use for holographic spatial-spectral imaging filters.

Volume holographic imaging (VHI) techniques have been successfully applied to microscopy and spectroscopic applications [1–6]. These systems require highly selective filters with high diffraction efficiency to obtain spatial-spectral information within objects. Nano-particles have recently been used in photopolymers to increase diffraction efficiency with enhanced index modulation for data storage applications [7–10]. Phenanthrenquinone- (PQ-) poly methyl methacrylate (PMMA) bulk materials [11–14] are attractive recording media in holographic imaging applications; they provide narrow filtering properties and minimal refractive index change to record holographic Bragg filters with a variety of forms and thicknesses. In this Letter, we present the design and performance of volume holographic transmission filters formed in nano-SiO₂ doped PQ-PMMA. Experimental results are also provided to demonstrate the ability of the new holographic imaging filters to reconstruct images of biological samples.

The holographic samples are prepared using a bulk polymerization process with optimized values [13,15–17] for concentration, exposure energy, and curing as well as fixing and preparation procedures. In this process MMA, 2,2 –Azobis(2-methylpropionitrile) (AIBN), and PQ are first mechanically mixed at an optimized weight ratio of 100:0.5:0.7 respectively, and then filtered with a 25μm pore filter. The nano-SiO₂ (BYK, LP-X-20740) is then added into the mixture. The size of the nano-SiO₂ is ~20nm. The nano-SiO₂ is incorporated into the mixture at different weight ratios, as shown in Table 1. The liquids are

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sonicated to mix them uniformly. The sonicated mixture is then poured into a mold and heated at 50°C for 120hrs to thermally activate polymerization and solidify the mixture. After the thermal polymerization process is complete, the resulting solid sample is ~1.6mm thick. The solid sample is then cut into 5cm×5cm squares and fit on a sample holder for exposure. The gratings are recorded using 488nm Argon ion (Ar⁺) laser light with an inter beam angle at ~60° and 1000mJ/cm² exposure energy. The grating vector is parallel to the sample surface.

The diffraction efficiency (η) of the transmission hologram shown in Table 2 was measured at 488nm for each group with different concentrations of nano-SiO₂. For sample Group 1, diffraction efficiencies up to 80% has been demonstrated in Ref [13,15–17]. The maximum η can be improved up to 93% in Group 2 with nano-SiO₂ at the weight ratio of 0.1. The samples with nano-SiO₂ at the weight ratio of 1.0 were highly non uniform [7,8] making grating recording barely possible for this mixture. In addition, Figure 1 shows the measured angular selectivity for high η samples of Groups 1 and 2. From the figure, it can be seen that Full-Width-Half-Maximum (FWHM) is reduced from 0.049 to 0.024 degrees with the inclusion of nano-SiO₂, and this indicates that grating effective thickness [15–17] has been significantly increased.

According to Kogelnik's coupled-mode theory [18,19], η and selectivity can be theoretically predicted for volume holograms with different parameters. For a lossy and un-slanted transmission phase grating, the η of the grating is given by

$$\eta = \exp\left(\frac{-2\alpha \cdot d}{\cos\theta_0}\right) \cdot \frac{\sin^2(\sqrt{\nu^2 + \xi^2})}{[1 + (\xi^2/\nu^2)]}, \quad \text{Eq. (1)}$$

$$\nu = \frac{\pi \cdot \Delta n \cdot d}{\cos(\theta_0 + \Delta\theta)}, \quad \xi = \frac{\vartheta \cdot d}{2\cos(\theta_0 + \Delta\theta)}, \quad \text{Eq. (2)}$$

where Δn and d represent index modulation and grating thickness of the hologram, respectively. $\Delta\theta$ is the angular deviation from Bragg-matched angle θ_0 , and α is the loss coefficient including absorption and scattering losses. The dephasing parameter ϑ can be written as

$$\vartheta = \Delta\theta \cdot K \cdot \sin\left(\frac{\pi}{2} - \theta_0\right) - \Delta\lambda \frac{K^2}{4\pi n}, \quad \text{Eq. (3)}$$

$$\frac{d\lambda}{d\theta} = 4\pi n \cdot \sin\left(\frac{\pi}{2} - \theta_0\right)/K, \quad \text{Eq. (4)}$$

where the grating vector $K = 2(2\pi n/\lambda)\sin\theta_0$ and n is the index value of the hologram. With Bragg-matched wavelength $\lambda = 488$ nm, the dephasing parameter $\vartheta = \Delta\theta \cdot K \cdot \sin(\frac{\pi}{2} - \theta_0)$.

Coupled wave theory is used to determine the index modulation and grating thickness by comparison with the experimental results. Figure 2 shows the comparison for the Group 2

formulation which gave the highest diffraction efficiency. The solid line represents the theoretical values and the star markers represent the experimental values. The index modulation is determined by matching the amplitude of the diffraction efficiency curve, while the grating thickness is determined by matching the FWHM bandwidth. The simulation gives an index modulation of 2.3×10^{-4} and grating thickness of 0.96 mm with parameters $n = 1.49$ and $\alpha = 0.028/\text{mm}$ and are listed in Table 3. Using $n = 1.49$ and $\alpha = 0.009/\text{mm}$ for Group 1 without nano-SiO₂ in PQ-PMMA [15,16], an index modulation of 2.0×10^{-4} and an grating thickness of 0.62 mm were determined and are also listed in Table 3.

In addition, for the Group 2 mixture the angular FWHM is reduced from 0.049 to 0.024 degrees with the inclusion of nano-SiO₂. According to Eq. 4, the spectral FWHM will also be reduced from 0.72 nm to 0.34 nm. This indicates that the index modulation and diffraction

efficiency are improved significantly up to and $\frac{2.3 \times 10^{-4} - 2.0 \times 10^{-4}}{2.0 \times 10^{-4}} \times 100\% = 15\%$ and $\frac{93 - 80}{80} \times 100\% = 16\%$, respectively. In addition, the spectral selectivity and grating thickness can be improved by $\frac{0.72 - 0.34}{0.72} \times 100\% = 53\%$ and $\frac{0.96 - 0.62}{0.62} \times 100\% = 55\%$, respectively.

Multiplexed gratings were also made using different nano-SiO₂ concentration sample groups as shown in Table 1. The bulk polymerization process is the same as the procedure described earlier. A pre-exposure of 360 mJ/cm² and a dark delay time of 10 s were used as described in Ref. [14]. The exposure energies for the two multiplexed gratings were 1000 mJ/cm² and 825 mJ/cm², respectively. The nominal angle between signal and reference arms was $\sim 60^\circ$ and was changed by 2° between exposures to record a hologram with a different reference beam angle and point source location. The displacement between point source locations along the optical axis was 50 μm . The η values of the multiplexed transmission hologram were measured at 488 nm for each group with different concentrations of nano-SiO₂ shown in Table 4. The maximum η values for two multiplexed holographic gratings were 59% and 42% and were obtained with Group 2. The angular FWHM was also reduced from $\sim 0.05^\circ$ to $\sim 0.02^\circ$. This indicates that sample Group 2 with the inclusion of nano-SiO₂ enhances both η and Bragg selectivity for multiplexed holographic gratings.

Figure 3 shows the multiplexed hologram using sample Group 2 of nano-SiO₂ doped PQ-PMMA in the experimental imaging system. Each multiplexed grating acts as a spatial-spectral filter to simultaneously project images from different depths on a CCD camera. An Olympus objective lens (ULWDMSP50X), a Mitutoyo imaging lens (MPlanAPO20X), and an Andor iXon CCD array (Andor X-2647) were used. The resultant images from two depths within a grapefruit skin are shown in Figures 4(a) and (b) and were reconstructed by the same multiplexed hologram. Very small features of $\sim 8 \mu\text{m}$ size can be resolved. The separation between the two depths within the grapefruit tissue was $\sim 65 \mu\text{m}$. One reconstructed layer is close to the grapefruit surface, and the other is at the depth of $\sim 65 \mu\text{m}$. The grapefruit skin was illuminated using a red LED with a central wavelength at $\sim 630 \text{ nm}$ and a spectral bandwidth of $\sim 25 \text{ nm}$. The field of view of both images is $\sim 1 \text{ mm} \times 0.8 \text{ mm}$.

In summary, nano-SiO₂ doped PQ-PMMA material has been demonstrated to offer high angular and spectral selectivity for holographic imaging filters. Analysis of the data shows that the holographic gratings formed in the optimized nano-particle doped PQ-PMMA material can significantly improve Bragg selectivity and increase the diffraction efficiency

by 53% and 16%, respectively. It is also possible to multiplex gratings in this material with high diffraction efficiency for sampling information from multiple object depths.

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Figure 1.

Angular spectrum of the 0.0 and 0.1 weight ratio nano-SiO₂ in PQ-PMMA. Dashed line data obtained from Ref. [17].

Figure 2. Comparison of theoretical and experimental diffraction efficiency and Bragg selectivity values obtained from single grating nano-SiO₂ sample Group 2.

Figure 3.
Experimental imaging setup.

Figure 4. Two depth-resolved images of a grapefruit skin obtained using multiplexed sample Group 2 with two holographic gratings. Grapefruit tissue features ($\sim 8\mu\text{m}$) can be visualized.

Table 1

Weight ratios of mixture components for different nano-SiO₂ concentration sample groups with the same thickness of ~1.6mm.

Main Components	Sample Group (weight ratio)			
	Group 1	Group 2	Group 3	Group 4
Nano-SiO ₂	0.0	0.1	0.5	1.0
MMA	100.0	100.0	100.0	100.0
AIBN	0.5	0.5	0.5	0.5
PQ	0.7	0.7	0.7	0.7

Table 2Measured diffraction efficiency for different nano-SiO₂ concentration sample groups

Sample Group	Diffraction efficiency η (%)
1	80 %
2	93 %
3	38 %
4	< 1 %

Table 3

Summary of the parameters and performance for high η sample groups.

Parameters and Performance	Sample Group 1	Sample Group 2
Index value n	1.49	1.49
Loss coefficient α	0.009/mm	0.028/mm
Index modulation Δn	2.0×10^{-4}	2.3×10^{-4}
Grating thickness d	0.62 mm	0.96 mm
Angular selectivity $\Delta\theta$	0.049°	0.024°
Spectral selectivity $\Delta\lambda$	0.72 nm	0.34 nm
Diffraction efficiency η (%)	80 %	93 %

Table 4

Measured η of multiplexed gratings for different nano-SiO₂ concentration sample groups

Sample Group	η (%)	
	Grating 1	Grating 2
1	47 %	39 %
2	59 %	42 %
3	22 %	17 %
4	< 1 %	< 1 %