



Publication Year	2017
Acceptance in OA	2020-11-18T14:20:46Z
Title	Process optimization for the manufacturing of holographic elements using Darol photopolymer
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Publisher's version (DOI)	10.1016/j.optmat.2017.08.050
Handle	http://hdl.handle.net/20.500.12386/28416
Journal	OPTICAL MATERIALS
Volume	73

Process optimization for the manufacturing of holographic elements using Darol photopolymer

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Abstract. Design and manufacturing of high efficiency and reliable volume phase holographic optical elements require photosensitive materials where the performances of the device are straightforwardly related to the recording and post process parameters. In this context, Darol[®] photopolymers are promising materials since the modulation of the refractive index is set during the thermal process that follows the holographic exposure.

In the present study, dry Darol[®] films are thermally processed to determine the optimized conditions to achieve a large modulation of the refractive index together with good optical properties of the hologram (i.e. transparency, flatness, and homogeneity). For each condition, volume phase gratings are written and characterized in terms of diffraction efficiency. A threshold temperature of 150°C is found, and only latent holograms are obtained at lower temperature, as confirmed by thermal characterizations. Moreover, a dependence of the refractive index modulation on the line density and laser intensity is highlighted.

Keywords. Refractive index modulation, holography, photopolymers, diffraction gratings.

1. Introduction

Holographic Optical Elements (HOEs) [1] cover nowadays a relevant position in optics as they modify an optical wavefront depending on the recorded holographic pattern. In this field, Volume Phase Holographic Gratings (VPHGs) work as dispersing elements, and they are currently preferred for spectroscopic instrumentation thanks to the high diffraction efficiency that can theoretically reach 100% [2,3]. Moreover, the grating can be *ad-hoc* customized according to the scientific requests. In these elements, a periodic modulation of the refractive index Δn in a film of constant thickness is responsible for light diffraction according to the refractive index pattern. The design and the manufacturing of high efficiency and reliable dispersive elements require photosensitive materials (as recording substrate) where the set parameters allow for a control over the final performances.

There are different classes of holographic materials suitable for VPHGs [4] and some of them are on the market for a long time. Dichromated gelatin (DCG) is considered the reference material, with the largest modulation of the refractive index so far reported [5,6]. However, it requires a complex post-exposure chemical developing to fix and adjust the final Δn , which may prevent an actual large scale production of holograms. Thus, new holographic materials with self-developing properties is highly desirable.

At present, photopolymers [5,7–9] are becoming a promising alternative, since they can be produced on a large scale and large size. Moreover, they are self-developing, i.e. the volume hologram is readily produced just after the holographic exposure without any chemical post processing as in the case of dichromated gelatins and silver halides. Unfortunately, the different working mechanism [10–12] gives a lower modulation of the refractive index that limits their use in some applications.

Recently, Darol[®] by Polygrama-Lynx [13] has been proposed as highly efficient photopolymer mimicking the DCG material. Materials are available as dry films for holography where a latent image is generated through the holographic exposure and the efficient hologram is obtained only after a thermal treatment that can be assimilated to the developing process in DCGs [14]. Optimization of the thermal treatment is crucial not only to reach high diffraction efficiencies, but also to preserve good optical qualities of the hologram, without inducing distortions and deformations. This issue becomes definitely relevant if the holographic film is applied in spectroscopy as dispersing element.

In this paper, we investigate the light-induced and the thermal physical-chemical processes the refractive index modulation in Darol[®] VPHGs is based on by means of thermal and spectroscopic analyses. We demonstrate that a threshold temperature of 150°C exists to obtain the efficient hologram, and we show the experimental thermal conditions to provide a reliable developing process. Moreover, a dependence of the Darol[®] performances on the writing parameters is found.

2. Darol[®] substrate

Darol[®] is a dry photopolymer, which means that the coating onto the substrate forms a solid layer film for holography, i.e. a thin layer of holographic material coupled with a polyester substrate. The photosensitive layer is sensitive to the presence of oxygen during the writing procedure, hence protection is required to prevent contact with air. This effect is

common for most photopolymers [15] and, for this reason, commercially available photopolymers for holography share the same layer-protected structure [16]. The material here studied was green sensitive with a variable thickness of the photopolymeric layer between 9 and 13 μm .

According to the material datasheet [14] this photopolymer can achieve a maximum Δn value of ca. 0.08 in the visible, and the suggested method to fabricate the high-performances hologram can be summarized, as follow:

- removal of the cover and lamination onto glass;
- exposure to green light (532 nm) with a total energy from 2 to 20 mJ/cm^2 ;
- bleaching with incoherent white light;
- thermal treatment by means of a film-heat drum or a jet of hot air for a few seconds in a range from 100 to 130°C. As an alternative, use of a Microfilm development machine for 0.75-1.5 seconds at 125°C or for 0.33-0.75 seconds at 130°C.

Since different heat treatments, which differ both for temperature and techniques, allow for the hologram formation, we have explored different post-process techniques to find a controlled and reliable process to address specific Δn values, along with material characterization which supports the understanding of the overall fabrication method.

3. Materials and methods

3.1 Sample preparation, VPHG writing procedure and characterization

The Darol® photopolymeric sheets are made of three layers: a polyester substrate, the photopolymer and a protective layer on top. For the recording step the protective layer must be removed, and the sample was laminated on a clean glass slide. The pristine photopolymer layer was sticky, and it remained attached to the glass surface; hence, the sensitive material was protected from the environment at both sides. A red glass filter was put onto the back layer of the sample with a fluid in between, in order to prevent the formation of parasitic patterns during the holographic exposure. Holographic recording was performed using both a classical two-plane-mirror configuration and a Lloyd's mirror configuration. A DPSS laser (Torus, Laser Quantum, 500 mW single mode TEM00) at 532 nm, integrated onto an isolated optical table, was used and the beam was delivered to the different holographic setups through different folding mirrors. After the holographic recording, samples were post-exposed to a halogen lamp (20 W of power) to complete the polymerization also in the unexposed regions of the grating and **to bleach the residual unreacted dye, as it is suggested in the material datasheet [14]. In this work, bleaching irradiation took 10 minutes, and the distance between light source and sample was 10 cm (equivalent to about 28 mW/cm^2).**

The VPHGs were characterized in terms of diffraction efficiency at fixed and monochromatic wavelengths for various incidence angles and different polarizations. **The setup diagram is displayed in Figure 1.** The diffracted light (according to the grating's equation) was measured by a Si photodiode, mounted on a motorized rotating stage. The retrieved data were further processed by adopting the Kogelnik's model [17] that allows to find the unknown grating parameters (i.e. index modulation, thickness) **through the fit of the measured data.** To verify the applicability of this model we had to discern between the two different polarizations and take into account the Fresnel reflection losses included in the diffraction efficiency measurements (no AR coating was present onto the surfaces). The approach was to apply the classical Fresnel equations as function of the incidence and diffraction angles to the substrate/air interface. Hence, we neglected the reflection losses between the holographic material and the substrate, since they were much smaller than the losses between the substrate and air.

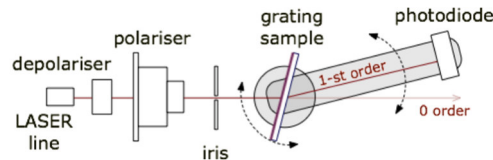


Figure 1: Scheme of the optical setup used to characterize the holographic samples.

3.2 Thermal and optical analysis

To perform the TGA, the photopolymer layer was removed from its polyester substrate. Unfortunately, this delamination process was not easy due to the adhesion between the two layers. Nevertheless, a scalpel was successfully used, which removed the layer to be placed in the aluminum sample holders. The instrument used was a Seiko Instruments EXSTAR 6000 TG/DTA 6300.

The UV-Vis spectra were collected using a Jasco V570 spectrometer. The photopolymer layers were analyzed onto their polyester substrate and only the protective cover was removed. Samples were prepared according to the description reported in the main text.

4. Darol® characterization and results

4.1 Transmission spectra

We recorded the UV-Vis-NIR spectra of the unexposed material (pristine), of the bleached sample (only exposed to the incoherent light) and of a volume phase grating written and developed (Figure 2). The VPHG is characterized by 3000 l/mm, it was written using a power density of 0.5 mW/cm² for 40 s and bleached by means of white light (28 mW/cm²) according to the manufacturer's specifications. A such large line density, along with the recording of the spectrum at normal incidence, was selected in order to have a negligible diffraction efficiency in the 300 – 1600 nm region. As a consequence, only the scattering and absorption by the film were detected.

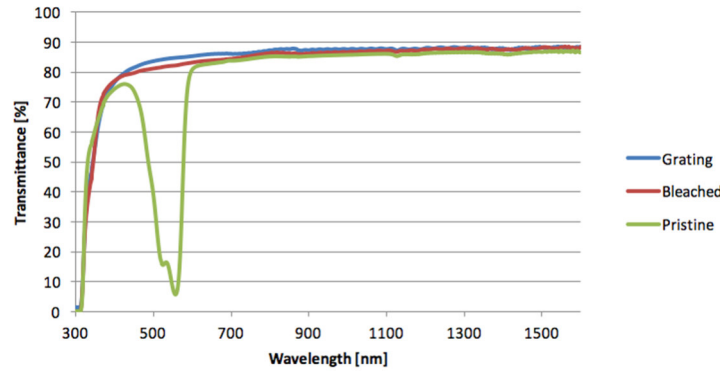


Figure 2: Transmission spectra of Darol® photopolymer samples: unexposed (pristine, green line), film only exposed to incoherent light (28 mW/cm²), namely bleached (red line) and grating written and developed (blue line).

The pristine sample shows an intense absorption band at 554 nm with FWHM of ca. 90 nm, confirming the green sensitivity of this photopolymers. The transmittance in the near infrared is about 87%, this value is slightly lower than the one ascribed simply due to the Fresnel reflection losses (about 8%), thus, a small amount of scattered light should be present and has to be confirmed with further tests. At short wavelengths, the material starts to absorb at 310 nm and it becomes completely opaque below 330 nm. Moreover, there is not any evidence of residual absorption of the dye in the visible (400-600 nm). This is true both for the bleached sample and the grating sample. The spectra of the bleached sample and of the grating are very similar, they do not show the dye absorption band or any other evident decrease in transmittance between 400 and 1600 nm. All the gratings written with conditions that do not diffract the light used in the transmission characterization behaved identically.

4.2 Thermal treatment

As previously mentioned, different thermal development techniques were initially investigated to find the best procedure to fabricate high performance diffraction devices.

With the same holographic recording conditions, VPHGs with 2000 l/mm were prepared and their diffraction efficiency was measured prior and after the post-processing step. This particular line density was chosen in order to be in the Bragg diffraction regime, making possible the application of the Kogelnik model to fit the measured data. With this line density and at 532 nm, a ρ value > 27 is calculated and this guarantees the reliability of using the Kogelnik model to fit the diffraction efficiency [2].

We tested the heating process by means of heating-gun (air temp. output 350°C, ca. 27 cm distance), IR lamps (100 W, ca. 3 cm distance) and oven (110°C for 1.5 s). To perform the post-process, the film was detached from the glass substrate and heated face up, in order to expose the photopolymeric surface to the air and let any volatile products to leave. Although these methods are reported in the material datasheet, we did not see any improvement in the diffraction efficiency in none of the cases. As also suggested, samples were also placed onto a pre-heated (125°C) aluminum plate for 25 seconds. Even with this method, the measured efficiencies remained similar to the untreated samples; worth to be mentioned, an important drawback of this method is the worsening of the transparency and the large mechanical deformation of the layer.

According to all these preliminary results, we selected the thermal heating with oven as the most controllable method to be optimized. To avoid any possible deformation of the substrates caused by the thermal scans, the heated gratings were promptly sandwiched between two glass covers, coupled with paraffin oil, and allowed to cool down to room temperature. This coupling fluid was suggested by the material manufacturer as it should not make any effect on the photopolymer. Different runs of thermal post-process were carried out varying time and temperature of the treatment. Results are reported in Table 1.

Table 1: Unpolarized diffraction efficiency [%] of 2000 l/mm VPHGs (**0.5 mW/cm² for 40 s**), tested at 532 nm, as function of the temperature and duration of the thermal treatment. **The uncertainty of the efficiency measurements is $\pm 0.5\%$ (standard deviation) for all the samples.**

t [s] \ T[°C]	110	130	150
2	9.15	5.25	16.95
5	4.40	10.80	21.75
10	6.55	8.50	23.20
20	7.70	9.85	32.70
60	8.55	9.55	48.05
120	7.45	8.65	67.20

Heating temperatures of 110 and 130°C do not induce any effective increase in the diffraction efficiency and no relevant differences appear as function of the heating time, meaning that only the latent hologram remains impressed. At 150°C, there is an increase in the diffraction efficiency even for a few seconds of treatment. Moreover, the longer the thermal heating, the higher the efficiency.

The trend suggested that heating at higher temperatures could lead to larger efficiency, but the processability of the samples at this temperature is critical and only for few samples were possible to obtain a trustworthy efficiency value. The film deterioration (substrate heavily bended and deformed) prevented any further optical test on these samples.

From these preliminary tests we concluded that 150°C is the threshold temperature below which only the latent hologram remains and no enhancement of the hologram takes place.

4.3 Duration of the thermal treatment

In order to determine the heating time to achieve the best diffraction efficiency, more prolonged tests **on different gratings written with 0.5 mW/cm² for 40 s and heated at 150°C were performed (line density of 2000 l/mm).**

Figure 3 reports the measured values for the diffraction efficiency at 532 nm in the two polarizations and the **mean** refractive index modulation determined by applying the Kogelnik model through a fitting procedure (**examples in Figure 4**).

It is evident time longer than 500 s is required to reach high performances. Concerning the s-polarization, efficiency increases up to **1200 s** treatment and then **starts** decreasing, whereas the refractive index modulation continues to rise. This behavior occurs due to the over-modulation effect, which is typical in gratings with high Δn and specific values of the product $\Delta n \times d$ [18].

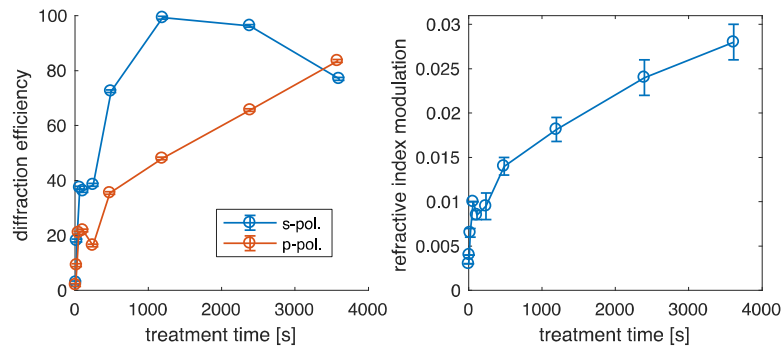


Figure 3: on the left, **measured** diffraction efficiencies at 532 nm for the two polarizations and the corresponding **mean** refractive index modulation on the right. The samples were heated to 150°C as function of the duration of the thermal treatment. The diffraction efficiency of the two polarizations (blue and orange curves) show a different trend with the treatment time because of the overmodulation effect. **Error bars due to the fitting procedures are reported.**

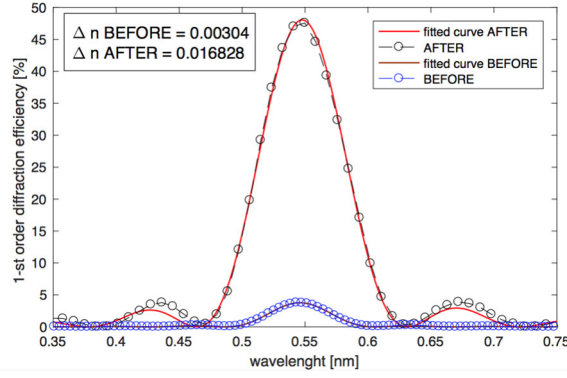


Figure 4: Examples of p-polarization diffraction efficiency measurements with the corresponding Kogelnik fit of a VPHG without any thermal treatment (BEFORE: written at 0.5 mW/cm² for 40 s and bleached) and a VPHG post-processed with the thermal treatment (AFTER: 1200 s @ 150°C). In the box are displayed the DN values retrieved by applying a fit with the Kogelnik model.

4.4 Writing power density

In photopolymer-based VPHGs, diffraction efficiency often depends on the writing power density [19–21]: a high power can prevent a complete diffusion of the monomers from the dark areas to the illuminated areas. To evaluate this possible effect, gratings were exposed to different power densities (from 0.25 mW/cm² to 1 mW/cm²) while maintaining the total energy dose, during the holographic exposure, within the values reported in the datasheet (20 mJ/cm²).

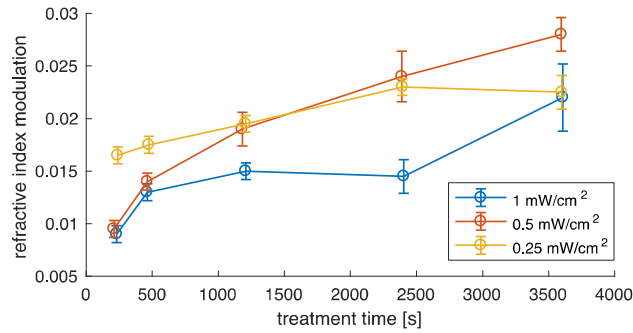


Figure 5: Refractive index modulations achieved in samples heated at 150°C as function of the duration of the thermal treatment for different power densities. Error bars due to the fitting procedures are reported.

For each writing power, we have a set of samples with different post-processing duration: the results, reported in Figure 5, show again that the increasing Δn trend with thermal treatment time is common for every writing power set. On the other end, we cannot identify a clear trend of the Δn with the power density, at constant thermal treatment.

4.5 Grating period

Considering the reaction-diffusion mechanism that generates the hologram in photopolymers [22], it is apparent that the grating period Λ (i.e. the inverse of the line density G) strongly affects the refractive index modulation of the recorded VPHG. In order to evaluate this effect, samples were recorded with G values of 1000, 2000 and 3000 l/mm using a power density of 0.5 mW/cm² for 40 s. After the recording step, the samples were thermally treated at 150°C for different times. The retrieved refractive index modulations are reported in Figure 6.

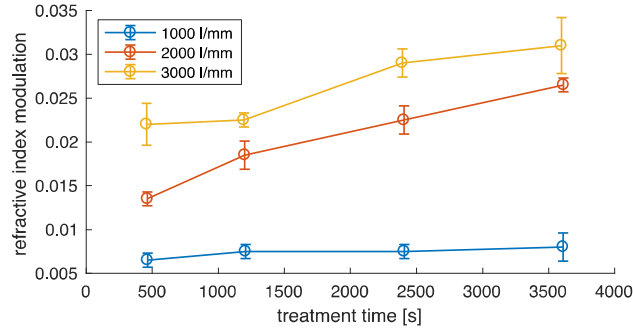


Figure 6: Refractive index modulations in gratings exposed to 0.5 mW/cm^2 for 40 s, and heated to 150°C as function of the duration of the thermal treatment; Grating periods: 1000 l/mm (blue line), 2000 l/mm (red line) and 3000 l/mm (yellow line). **Error bars due to the fitting procedures are reported.**

An increase of the refractive index modulation occurs with the duration of the thermal treatment for all the studied line densities. Interestingly, we identify a clear enhancement of the Δn with the increase of the line density. This trend is consistent with other results reported in the literature [19], and it is related to the fact that the diffusion mechanism is more effective when the grating period is small. Indeed, the distance between two consecutive fringes is smaller for large G , for this reason monomers have to diffuse for a shorter distance to reach the unexposed regions. The overall trend, i.e. higher modulation for longer durations of the thermal treatment, confirms the results of the previous tests; however, the effect is more evident in samples with high G , which are characterized by a more efficient diffusion of monomers upon exposure giving large differences in the different pattern areas. This strong material modulation in the latent hologram leads to a stronger enhancement through thermal treatment. In conclusion, high values of the refractive index modulation (up to 0.03) and large diffraction efficiency can be obtained in Darol[®] photopolymer with suitable thermal treatment. Moreover, the target Δn can be actually tuned, by varying the duration of the thermal treatment over a threshold temperature.

5. Morphological and thermal characterization

The discover of a threshold temperature to generate efficient Darol[®] gratings has open to further investigation aimed at understanding the mechanism behind the thermal post-process.

5.1 AFM measurements

In a general photopolymeric material, the holographic process involves the diffusion of the different chemical species inside the volume of the active film, in order to spatially resemble the interference pattern that is projected during the exposure [20]. This will usually generate almost purely volume phase holographic gratings [4].

In order to understand if the periodic structure of the produced devices has to be attributed to a volume phase grating or also to a surface relief grating, further tests were performed.

A morphological analysis was carried out to study the possible surface modification of the sample upon the optimized thermal treatment, which can originate either from a dynamical material rearrangement or the loss of volatile compounds from the exposed area. The latter can be likely related to the enhancement of the refractive index modulation, hence of the diffraction efficiency. Therefore, Atomic Force Microscope (AFM) was used to retrieve the surface maps of recorded grating. **Three different samples were studied: a uniformly bleached film (without any grating), a VPHG with 2000 l/mm without any thermal treatment and a VPHG treated for 3600 s at 150°C .** The AFM maps are reported in Figure 7.

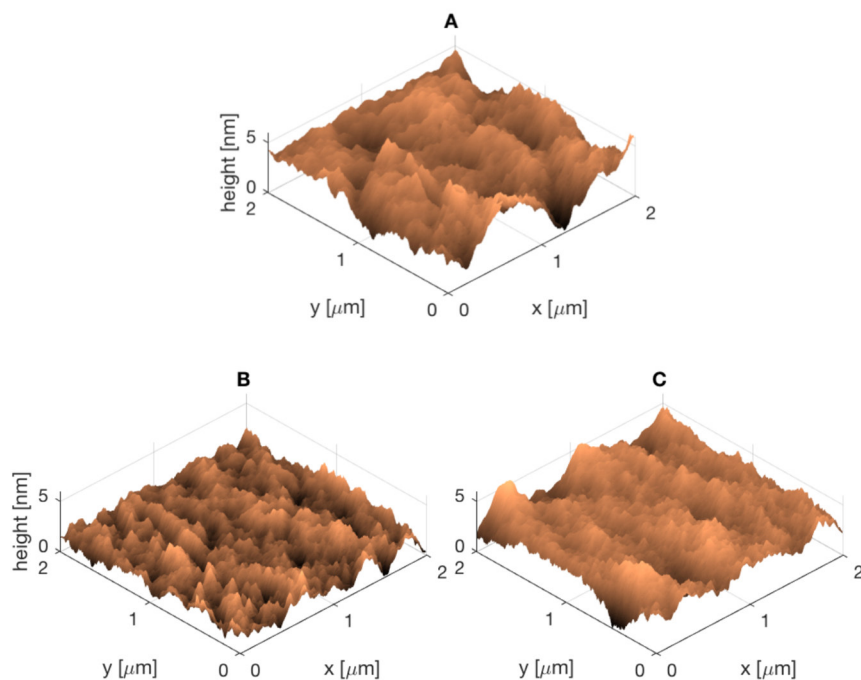


Figure 7: AFM images of the A) bleached film without any grating, B) untreated grating (power density of 0.5 mW/cm² for 40 s) and C) thermal treated grating, 3600 s at 150°C (right).

The roughness RMS is 3.73 nm for the bleached film, 1.63 nm for the untreated grating and 3.49 nm for the 3600 s thermal treated grating. According to the AFM images, no periodic structure related to the impressed grating is visible, meaning that, the diffraction of the light is due only to the presence of a periodicity (of the refractive index) stored within the volume and not to a surface grating induced by the production of volatile compounds.

5.2 Thermo Gravimetric Analysis (TGA): material is lost after 150°C

To obtain a more complete understanding of the thermal mechanism in the hologram formation, a set of thermal characterization was carried out. We studied different samples, namely the *pristine*, the *bleached* (only halogen exposure), the *thermally treated* photopolymer (halogen exposure and 1h post process at 150°C), and the *grating* (recorded at 0.5 mW/cm² for 40 s and with a spatial frequency of 2000 l/mm) with and without the thermal treatment at 150°C.

According to the datasheet, it is not definitely clear if the thermal treatment promotes the migration of the reacted species or causes volatilization of by-products thus leaving micro-voids which resemble the mechanism in DCGs. Thermo-Gravimetric Analysis (TGA) can provide important information on weight loss as function of the temperature. Measurements were performed up to 300°C with a heat ramp of 10°C/min (see Figure 8).

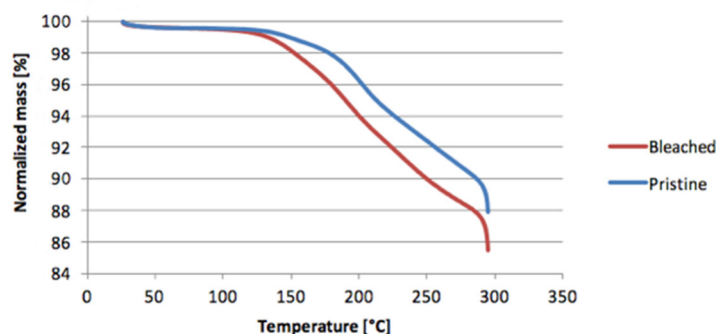


Figure 8: TGA plots for the pristine (blue line) and the bleached (red line) samples.

It is noticeable that the loss of mass starts approximately at 150°C. This means that above this temperature, volatile compounds evaporate or chemical reactions take place providing volatile products. Interestingly, both samples show the same trend of mass loss, meaning that this process occurs both in light-developed sample and in the untreated material.

These results are consistent with the threshold temperature found in the fabrication of gratings previously discussed, which promotes the significant enhancement of the diffraction efficiency. The thermogravimetric curves have two slopes of weight loss, which may lead to the hypothesis that two different mechanisms take place. We also notice that approaching 290°C a steep mass loss occurs for all the samples, which is ascribed to the overall degradation of the photopolymer.

Isothermal treatment at 150°C was also performed and monitored by further TGA, to better reproduce the thermal post-process we performed to develop the holograms. Specifically, samples were heated up to 150°C with a heat ramp of 10°C/min and then kept at 150°C for one hour. The normalized-mass vs. time curves are reported in Figure 9.

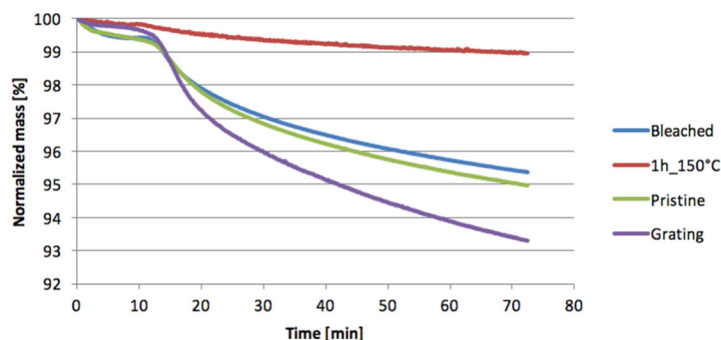


Figure 9: TGA plots for a 10°C/min heat ramp from 0 to 840 s and an isothermal heating at 150°C from 900 to 10500 s; pristine sample: green line; bleached sample: blue line; sample bleached and thermal treated at 150° for 3600 s: red line; grating: purple line.

Looking at the curves, no mass loss occurs before reaching 150°C (0 - 840 s), whereas mass decreases for pristine, bleached and grating samples. The sample which was thermally treated for 3600 s at 150°C before TGA was performed does not show significant weight loss. This is not surprising since this sample has already lost most of the volatile compounds. The decrease in figure shows an average speed of about 0.04%/min for the pristine and bleached samples, while for the grating sample is about 0.1%/min. This finding suggests that the reaction induced by the holographic exposure (grating sample) generates a large amount of volatile co-products in respect to the bleaching treatment only. Indeed, if the holographic exposure did not provide volatile co-products during the photopolymerization but only the insertion of the monomer in the growing polymer chains, a larger mass loss would be expected both for the pristine sample and the bleached samples. The opposite behavior observed from TGA leads to the hypothesis that the photoreaction provides volatile products, thus mimicking the mechanism in DCGs.

6. Conclusions

Volume holograms based on Darol® photopolymer have been written and the thermal treatment to develop the material has been optimized. We demonstrated that a treatment at 150°C in an oven with a suitable support for the sample is the optimal condition to enhance the diffraction efficiency of the hologram preserving the opto-mechanical properties **and to avoid the deformations that are preventing post-processing at higher temperatures**. The modulation of the refractive index was proportional to the duration of the thermal treatment and values up to 0.03 have been achieved. The effect of the line density of the grating and the writing laser power has been highlighted. **It seems that a reduction of the Δn with the writing power occurs; unfortunately, the trend is not so clear as for the case of the line density.** AFM measurements confirmed that pure volume holograms have been obtained, without any appreciable periodic surface relief. In addition, TGA measurements evidenced that the threshold temperature of 150°C to produce efficient gratings corresponds to a strong release of volatile compounds from the material, which enhances the refractive index modulation.

Acknowledgments. This work was partly supported by the European Community through the OPTICON project (Optical Infrared Co-ordination Network for astronomy, Horizon 2020, grant agreement no. 730890) and by the INAF through the TECNO-INAF 2014 “Innovative tools for high resolution and infrared spectroscopy based on non-standard volume phase holographic gratings”.

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