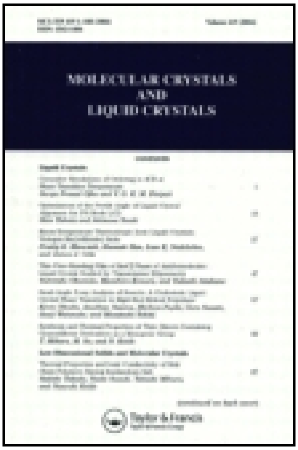

Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl20

Policryps Characterization in the Near Infrared

Giancarlo Abbate [a], Antigone Marino [a] & Francesco Vita [a]

[a] INFM UdR of Naples, DSF Università di Napoli Federico II, Naples, (Italy)

To cite this article: Giancarlo Abbate, Antigone Marino & Francesco Vita (2003): Policryps Characterization in the Near Infrared, Molecular Crystals and Liquid Crystals, 398:1, 269-280

To link to this article: http://dx.doi.org/10.1080/15421400390221817

Mol. Cryst. Liq. Cryst., Vol. 398, pp. 269–280, 2003
Copyright © Taylor & Francis Inc.
ISSN: 1542-1406 print/1563-5287 online
DOI: 10.1080/15421400390221817

POLICRYPS CHARACTERIZATION IN THE NEAR INFRARED

Giancarlo Abbate, Antigone Marino, and Francesco Vita*
INFM UdR of Naples and DSF Università di Napoli Federico II, Naples(Italy)

We have analysed the electro-optical behaviour at 1550 nm of a new polymer and liquid crystal composite, realized in form of a thick grating. This composite is known as Policryps, acronym that stands for "Polymer Liquid Crystals Polymer Slides", because it shows a sequence of polymeric sheets alternated by layers containing only liquid crystal. Thus, by its intrinsic nature a Policryps is realized in form of grating. It is known that Policryps gratings may have a good diffraction efficiency at 633 nm and they can be electrically switched between the diffracting and non-diffracting states, with time response in the millisecond range. Eventually, Policryps may form a key component in photonic devices for use in telecom applications. With this aim, we made an optical characterization in the telecom C-band of the very first samples produced by University of Calabria (Cosenza, Italy) with this material. We measured the diffraction efficiency versus temperature and applied voltage. We also made a dynamical characterization of the grating switching. By comparison among the results obtained in the red and in the Near Infra-Red (NIR) regions, respectively, we got important information for designing and realizing Policryps samples optimised for working in the C-band.

Keywords: polycryps; liquid crystals; near infrared; Bragg grating; diffraction efficiency

INTRODUCTION

A switchable diffraction grating is a key component for the realization of several devices used in the chain of optical communication networks, such as switches, Add-and-Drop systems, beam deflectors, routers. For instance, one of the easiest way to realize an optical multiplexing/de-multiplexing

---

This work has been performed under the Italian Ministry of Education and Research (MIUR) project PRIN2000 [7]. We gratefully acknowledge useful discussions with our partners of Rome and Cosenza groups in that project. In particular, Prof. C. Umeton group in Cosenza have provided all the samples investigated here. Moreover, Prof. Umeton and Dr. Roberto Caputo gave us a number of very important hints during many discussions.

*Corresponding author. E-mail: giancarlo.abbate@na.infn.it device is to integrate one or several switchable gratings in a guiding system such as a fiber or a waveguide [1]. A number of different technologies have been proposed for realizing such a component, including, quite recently, a technology based on Liquid Crystals (LC) materials. On the other hand, the possibility of high density information storage with low power consumption has been demonstrated in PDLC (Polymer Dispersed Liquid Crystals) systems [2]. That's why laser-induced formation of diffraction gratings in polymer-Liquid Crystals (LC) composites is an item of increasing scientific and technical interest.

At present, the most common process to get these gratings is realizing the so-called H-PDLC (Holographic PDLC) [3,4]. It consists in the alternate sequence of isotropic stripes of a polymer-LC mixture followed by stripes of anisotropic PDLC material; the optical contrast between the two series of stripes gives rise to the diffraction grating: it is electrically-switchable because the application of an electric voltage drives the PDLC stripes in an optical state, whose constants are almost matched with those of the isotropic stripes. Thus, under the application of the electric voltage the H-PDLC becomes a homogeneous layer and the grating disappears.

More recently, following the same scheme as above for the electrical switching, a new process has been proposed for the realization of diffraction gratings with polymer-LC composites. The resulting material has been named by the authors Policryps that is "POlymer LIquid CRYstals Polymer Slides" [5,6]. It is composed by a sequence of homogeneously aligned LC layers separated by isotropic polymer walls having approximately the same thickness of the LC ones, see Figure 1.

From a morphological point of view, the biggest difference amongst H-PDLC and Policryps is the following. In the former, in the UV-illuminated regions, a light-induced phase separation process creates LC droplets

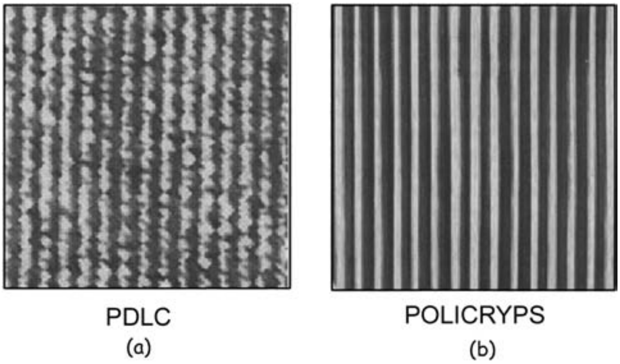

FIGURE 1 SEM microscope images of PDLC and Policryps gratings.

embedded in a polymer matrix, while the dark regions remain in the form of isotropic LC-polymer mixture, see Figure 1a. In the latter material, no phase separation happens within the illuminated regions, instead, under particular conditions, it happens between the illuminated and non-illuminated regions, so that one component (e.g. polymer) is almost totally confined within illuminated regions and the other component (e.g. LC) within non-illuminated regions, resulting in the geometry depicted in Figure 1b.

One essential feature these materials must exhibit in order to be attractive for telecom applications consists in their transparency in both the ON and the OFF states. This constraint imposes an upper limit to the size of the droplets in the PDLC; in fact droplet size of the order of the light wavelength gives rise to significant scattering losses. For this reason, nano-PDLC with droplet size of the order of 100 nm have been developed by different authors [3,4]. Nano-PDLC may show a very high diffraction efficiency and good transparency having however as a drawback a quite large driving voltage, increasing with decreasing droplet size.

Due to their morphology, being a stack of uniform and transparent layers, Policryps samples are inherently transparent. This material is very new and just some initial characterization has been performed by the same group that produces it [6]. However, this first characterization gives a clear indication that very high diffraction efficiency can be reached while the driving voltage can be kept at reasonable values: in few words, Policryps seems a very attractive material and it is well worth continuing the research studies on this composite in order to fully characterize it and eventually to get feedback for its optimisation at telecom wavelengths. The aim of the present work is to give an optical characterization of some Policryps samples both in the visible range, at $\lambda = 633\,\text{nm}$, and in the near IR, at $\lambda = 1550\,\text{nm}$ that is the C-band of the 3rd window for optical communications. It is worth noting that very few optical characterizations have been performed until now at telecom wavelengths on LC materials and in particular on polymer-LC composites. In our opinion, this lack of necessary data and the consequent uncertainty on working performances might be one reason for the limited present utilization of these materials in devices and systems for optical networks.

EXPERIMENTAL

The samples we have studied have been kindly provided by the group that has developed Policryps at the University of Calabria, in the frame of a running Italian National Research Project [7].

We got three different samples labelled as HCB-25, HCB-31, and HE-23, respectively. They are made by a composition of Norland Optical Adhesive

TABLE 1 Samples Characteristics

Acronym	HE-23	HCB-25	HCB-31
Liquid crystal	E7	5CB	5CB
Grating pitch (μm)	1.34	1.34	1.34
Grating depth (μm)	7.8	7.8	8.8

NOA-65, diluted in a 30% concentration of nematic LC. The nematic material was 5CB in the first two samples and E7 in the third one, both commercially available by Merck (Germany). The whole production process of Policryps samples is still patent pending and the final result are gratings as schematically shown in Figure 1a. Policryps samples are sandwiched between glass plates and sealed. The glass plates were ITO coated for applying an electric field to the cell and no further alignment treatment was made. The geometric characteristics of the three samples are summarized in Table 1, where the grating pitch is defined as usual and the grating depth is the distance between the two glass plates.

By application of a proper electric field, we can switch the grating amongst the ON and OFF states (Fig. 2). If there is no applied voltage, the incident light will see the phase grating formed by the two materials with different refractive index and it will be diffracted according with the Bragg law [8]. Applying an external voltage, the molecular director in the LC slides can be reoriented changing the refractive index seen by the impinging linearly polarized light. Eventually, we can reach the condition $n_{LC} = n_{polymer}$; if and when this condition is met, the beam is no more diffracted.

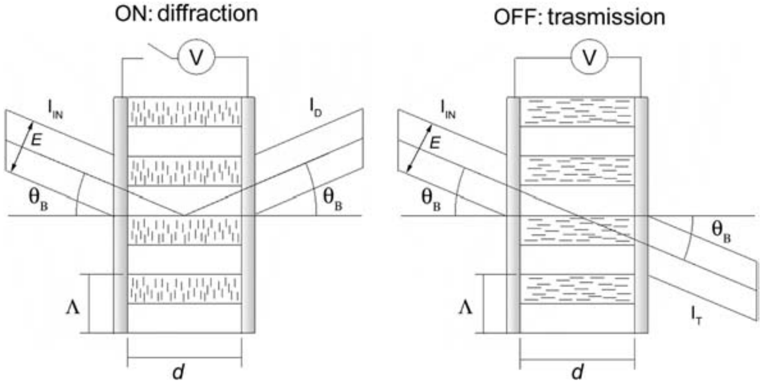

FIGURE 2 Policryps operating model.

We have performed measurements with both stationary and dynamic electric fields to fully characterize the samples: investigation with red light showed same characteristics as given by producer for the HCB samples; this was not the case for the sample HE-23 that appeared to be damaged, thus we shall avoid in the following any reference to that sample.

First, we have noticed that the behaviour of the grating is strictly dependent from the incident beam polarization: it is possible to reach the diffraction condition only with a p-polarized wave. The reason should be found in the molecular orientation inside the LC layers. First, let's recall that at $\lambda = 633\,\mathrm{nm}$ and 25°C the polymer NOA-65 refractive index is $n_p = 1.56$ and the values for 5CB indices are $n_o = 1.53$ and $n_e = 1.71$, respectively; in the isotropic phase (35°C) 5CB refractive index is $n_{iso} = 1.57$. Now, the polarization dependence clearly indicates that, without any applied field, the molecular director is not randomly distributed in Policryps, as is for instance in a PDLC sample. For light impinging at quasi-normal incidence, we observed no grating formation (OFF-state) for s-polarised wave, thus the LC regions must exhibit a refractive index value close to the polymeric one, as the ordinary index is. Instead, a p-polarized wave experiences a phase grating, thus the index seen in the LC layers must be quite different from the polymeric one, then closer to the LC extraordinary index value than in the previous case.

We can conclude that the most likely unperturbed molecular orientation within the LC layers is along the plane of incidence and orthogonal to the polymeric walls as shown in Figure 2. This is confirmed by further observations with applied external electric field.

DIFFRACTION EFFICIENCY VERSUS TEMPERATURE

In all measurements presented in the following and related discussions, the impinging light is always p-polarized, moreover we define the Diffraction Efficiency (DE) as the ratio between the diffracted light intensity and the total light intensity, diffracted plus transmitted, behind the sample. DE in a Bragg grating is ruled by Kogelnik formula [8], eventually in the implementation proposed by Montemezzani and coworkers [9] for anisotropic media. Not going into the details of all physical parameters entering those formulas, it is anyway evident that a major role is played by the index mismatch among the two media forming the grating stripes. In our samples, polymeric layers exhibit a refractive index not affected significantly by the applied electric field and with a slight temperature dependence in the explored range, which is far from its glass temperature transition. Opposite considerations apply for LC layers.

In particular, 5CB has a clearing point around 35°C, thus a strong temperature dependence of both n_e and n_o around room temperature. Further considering that 5CB refractive index in the isotropic phase is quite close to the polymeric index n_p, we should expect a drastic drop of the diffraction efficiency heating the sample from room temperature to over 35°C.

This is actually what we observed. Figures 3 and 4 show DE versus temperature using He-Ne light at 633 nm and a DFB laser diode at 1550 nm, respectively. It should be noted that DE drop off temperature is always less than 5CB clearing temperature indicating a mesophase modification, in particular a reduction of the nematic range, induced in this confined geometry. This loss of nematic behaviour could be ascribed to the effect of impurities certainly present in the LC regions owing to the production process of this composite: a low concentration of unreacted monomers as well as of photo-initiator molecules is still present within LC layers after the phase separation process. Eventually, impurities effect could have been enhanced by effect of confinement. The two samples give quite similar responses. The most significant difference is observed between the curves recorded with red and NIR light. DE, which at 25°C can arrive over 90% with red light, hardly reaches 15% with NIR light. We shall come back to this crucial point in the next section, but it is almost evident that the choice made by the samples producer, concerning the refractive indices values of the materials, was in the direction of optimising performances working with red light.

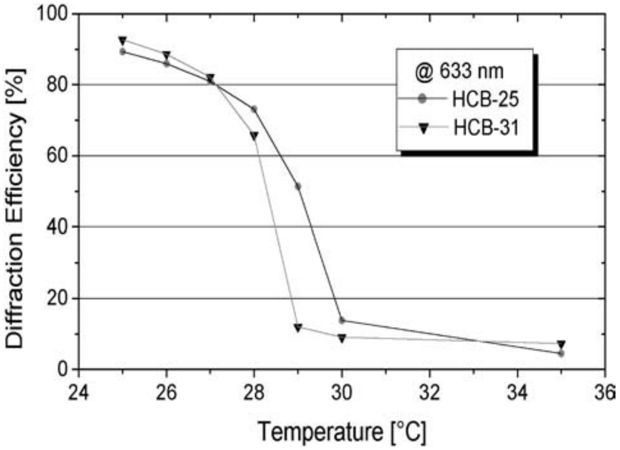

FIGURE 3 DE versus temperature at 633 nm.

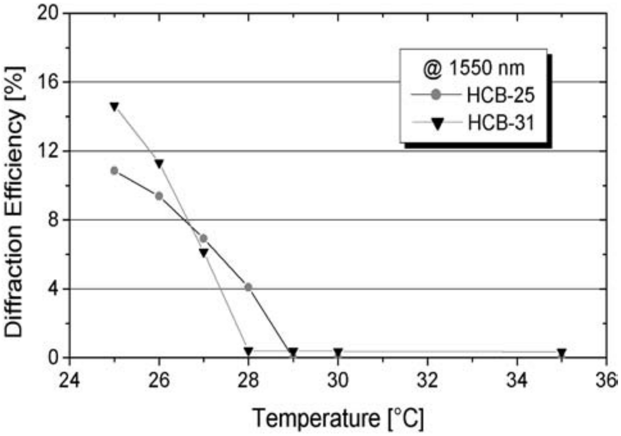

FIGURE 4 DE versus temperature at 1550 nm.

DE VERSUS APPLIED VOLTAGE

We made "static" measurements using actually an AC square voltage at a sufficiently high frequency, say 1 kHz, in order to avoid unwanted effects induced by the flow of free ions within the sample.

Results are shown in Figures 5 and 6 for red and NIR light, respectively. A very interesting features of Policryps gratings that can be inferred by

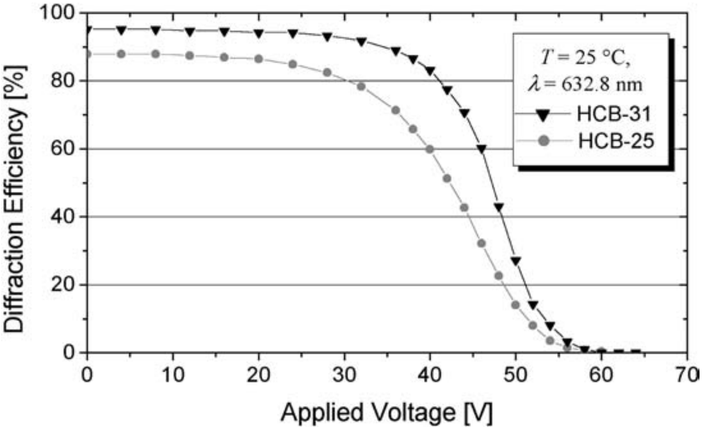

FIGURE 5 DE versus applied voltage at 633 nm.

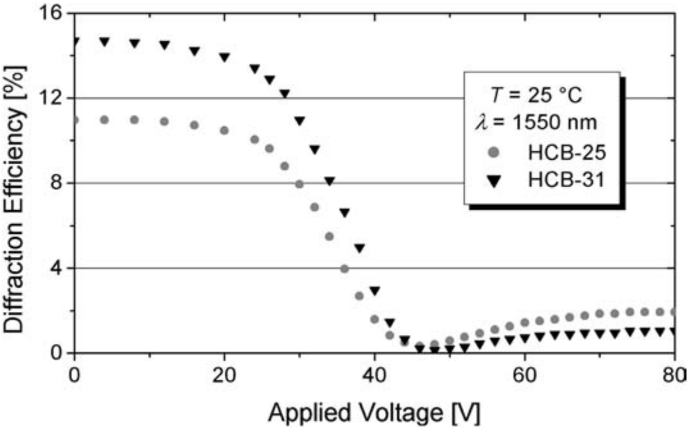

FIGURE 6 DE versus applied voltage at 1550 nm.

these results is the switching voltage. Actually, a threshold voltage of around 40 V was observed with red light in the sample HCB-31 and even less in the HCB-25 one, yielding a threshold field of about 4.5 V/μm in both cases. This is neither a very low nor a very high field for electric switching of LC-polymer composites, however if we compare it with competing nano-sized H-PDLC's, the Polycryps value is by far lower than those commonly observed in nano-PDLC's. Furthermore, it has been obtained in the very first Policryps samples, not optimised at all for electric performance.

In Figure 6, showing DE versus voltage obtained with NIR light, we notice again the large decrease in the maximum diffraction efficiency. Moreover, a slight decrease in the threshold voltage is observed. A complete switching off is reached for voltage around 45 V; however, further increasing the applied voltage above 50 V leads to a small "undesired" increase of the diffraction efficiency. We can deduce that, using NIR light, the OFF state of the grating is not reached when the molecular reorientation is completed in the LC regions, as is the case using red light, but instead for some intermediate configuration.

Observations in this and in the previous sub-section consistently confirm that the samples under investigations have fairly good performances at 633 nm, while they are not optimised at 1550 nm. This behaviour was expected and our purpose was exactly a quantitative evaluation of this performance degradation and possibly its correlation with material parameters' variation. In fact, if the three index values, n_e, n_o, n_p, are properly chosen at red wavelength, owing to the great difference between the dispersion curves of the polymeric material and the LC one, they cannot be optimised at the wavelength of 1550 nm.

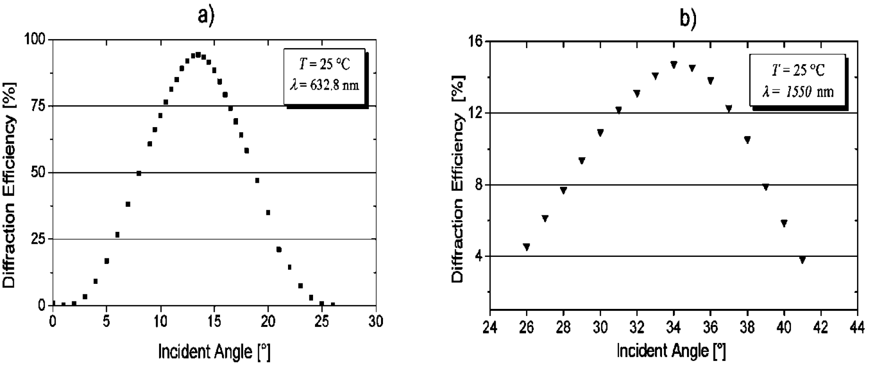

FIGURE 7 DE versus incident angle at 633 nm (a) and 1550 nm (b).

ANGULAR SELECTIVITY

In Figure 7, we report the grating response varying the angle of incidence of the light beam around the Bragg angle, because the angular selectivity is also an important parameter for photonic applications.

We observed a wide angular response at both wavelength in our samples but we know that we can affect and tune it by changing the geometry of the grating, especially the grating length, and/or by using the grating in reflection rather than in transmission.

DYNAMIC MEASUREMENTS

Response times of Polycryps gratings were measured by recording the diffracted beam signal detected by a photodiode. A driving voltage, 60 V in amplitude, was applied to the ITO electrodes at different frequencies in the range 1 Hz–100 Hz. In order to avoid static electric biasing of the sample, we always took care of applying a zero-average waveform. For impinging red light we show, in Figures 8a and 8b, the oscilloscope display with the applied waveform (lower part) and the optical response (upper part) at voltage frequency of 100 Hz and 1 Hz, respectively.

Figures 9a and 9b show the analogous oscilloscope display for impinging NIR light. In the optical response at 1 Hz of Figures 8b and 9b, it can be noticed a somehow irregular response and even a slight increase with time of the detected signal during the OFF state, that is with the voltage applied. This was unexpected and probably due to a molecular flow or turbulence induced by free charges motion during a quite long, 500 ms, constant voltage. To overcome this problem, we have further modulated at very high

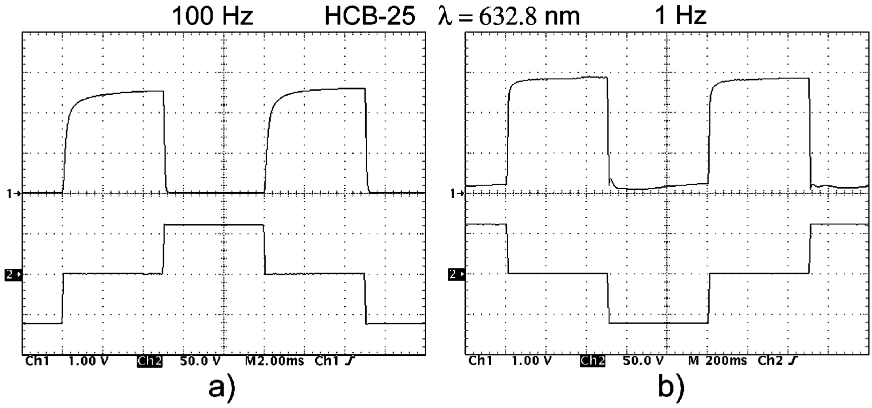

FIGURE 8 Optical response, upper curves, and driving waveform, lower curves, at 633 nm. a) voltage frequency 100 Hz; b) 1 Hz.

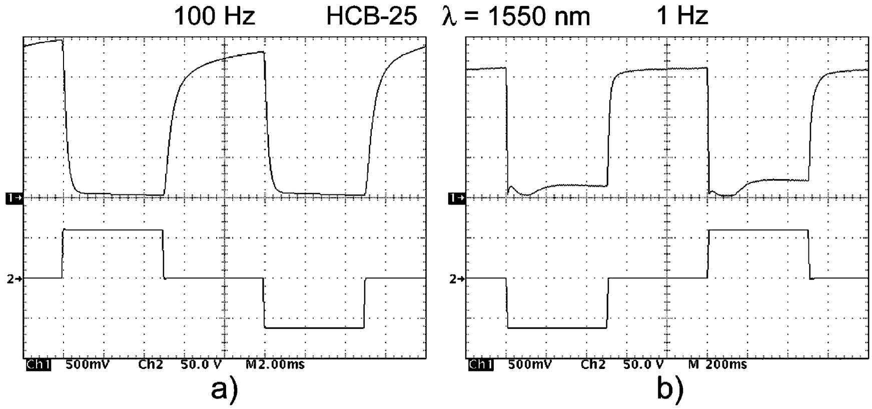

FIGURE 9 Optical response, upper curves, and driving waveform, lower curves, at 1550 nm. a) voltage frequency 100 Hz; b) 1 Hz.

frequency, say 10 KHz, the driving waveform. Furthermore, from Figure 9a it appears clearly that while the grating can be driven to a complete switch off within the first half-period of the applied voltage, restoring the grating on is not totally completed during the second half-period.

The most significant measured switching times are presented in Table 2, where the driving frequency is limited to 50 Hz for measurements at NIR wavelength, because of the previously mentioned observation. Differences among the achievements with red and infrared light are again evident,

TABLE 2 Response Times of Policryps Samples

Light wave length [nm]	Voltage frequency [Hz]	t_{ON} [μs]	t_{OFF} [μs]	Sample
633	1	1300	160	HCB-25
633	1	800	210	HCB-31
633	100	900	70	HCB-25
633	100	600	90	HCB-31
1550	1	2800	440	HCB-25
1550	1	2100	460	HCB-31
1550	50	2000	350	HCB-25
1550	50	1500	360	HCB-31

despite the fact that the molecular reorientation is obviously only driven by electric and elastic forces. However, the response of the diffraction grating is related to the molecular director reorientation in an indirect way, mainly mediated by the refractive indices values that once more prove to be the crucial parameters for Policryps performances. From Table 2, switching times to be dependent on the frequency of the driving voltage.

We notice also a big difference among switching ON and switching OFF times. Former is related to the molecular response to elastic forces and is in the millisecond range, while latter is related to the applied voltage thus electric forces and is in the microsecond range. This is not surprising, however we have indications that switching ON times could be significantly reduced in Policryps samples. Furthermore, we noticed that times t_{ON} are affected by large errors even greater than 25%, while t_{OFF} errors can be estimated in 5%. From the other hand, it seemed that the switching ON process is not ruled by a single time constant so that different competing physical mechanisms could be involved. Latter remark can also explain, at least qualitatively, why at lower driving frequencies we observed longer switching times: in fact, different competing physical processes may act with a different weight at different time scales, giving rise to the observed behaviour. Of course, we need further investigations to get a deeper knowledge about these mechanisms. Eventually, this knowledge may lead to improving the production process of Policryps, getting also t_{ON} in the microsecond range.

CONCLUSIONS

We performed an electro-optical characterization of the first produced Policryps samples using red light and NIR light in the telecom C-band. We wanted to make a comparison among Policryps performances at the two wavelengths, in particular observing how and how much performances were degrading at NIR wavelength. Obtained results are not only interesting but also very useful because they prove that this new LC-polymer composite can be conveniently exploited for photonic applications in the telecom field, once they have been optimised for use at telecom wavelengths. Our results together with a careful theoretical analysis, now in progress with Montemezzani theory, can give the needed feedback to the Policryps producer for its optimisation in view of the wanted applications. Very important in this regard will be also availability of accurate refractive index data, generally missing at present, in the C-band for LC materials [10]. Concluding, we are confident that Policryps grating with diffraction efficiency over 90%, good angular selectivity, and time response in the microsecond range, can be very soon obtained in the C-band, still maintaining a very good optical transparency with almost negligible scattering losses.

REFERENCES

[1] Domash, L., Crawford, G., Ashmead, A., Smith, R., Popovich, M., & Storey, J. (2000). *Proc. SPIE*, *4107*, 1.
[2] Lucchetti, L., Di Bella, S., & Simoni, F. (2002). *Liq. Cryst.*, *29*, 515.
[3] Lucchetta, D. E., Karapinar, R., Manni, A., & Simoni, F. (2002). *J. Appl. Phys.*, *91*, 6060.
[4] Bunning, T. J., Natarajan, L. V., Tondiglia, V. P., & Sutherland, R. L. (2000). *Annual Rev. Mat. Sci.*, *30*, 83.
[5] Caputo, R., Sukhov, A. V., Umeton, C., & Ushakov, R. F. (2000). *J. Exp. Theor. Phys.*, *91*, 1190.
[6] Caputo, R., Sukhov, A. V., Tabirian, N. V., Umeton, C., & Ushakov, R. F. (2001). *Chem. Phys.*, *271*, 323.
[7] Research Project of National Interest (PRIN 2000) by Italian Ministry of Education and Research, *Devices for routing in optical networks using new liquid crystals materials and composites*.
[8] Gaylord, T. K. & Moharam, M. G., (1985). *Proc. IEEE*, *73*, 894.
[9] Montemezzani, G. & Zgonik, M. (1997). *Phys. Rev. E*, *55*, 1035.
[10] Abbate, G., Del Gais, G., De Stefano, L., Marino, A., & Wagner, T. *Mol. Cryst. Liq. Cryst.*, appearing in the present issue.