Direct Inscription of on-surface waveguides in polymers using a mid-ir fiber laser

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Abstract

A detailed study of photo-inscribed optical waveguides in PMMA and polycarbonate using a mid-IR laser is presented. The wavelength of the laser is tuned near the absorption peaks of stretching C-H molecular bonds and the focused beam is scanned onto the surface of planar polymer samples. For the first time, we report the formation of optical waveguides in both polymers through resonant absorption of the laser beam. The optical properties of the waveguides were thoroughly assessed. An elliptic Gaussian mode is guided at the surface of both polymers. Insertion losses of 3.1 dB for a 30 mm long on-surface waveguide inscribed in PMMA were recorded. Such waveguides can interact with the external medium through evanescent coupling. As a proof of concept, the surface waveguides are used as highly sensitive refractometric sensors. An attenuation dynamical range of 35 dB was obtained for a liquid that matches the index of the PMMA substrate. Our results pave the way for large scale manufacturing of low cost biocompatible photonic devices.

1. Introduction

Over the last decade polymer-based photonic devices have been deployed in a wide range of scientific and industrial fields [1–4]. Polymers are rugged, lightweight and low-cost. Most importantly, their physical properties can be specifically customized to add functionalities or fulfill the requirements of a given application. The optical waveguides that form the basis of photonic devices in polymers are usually fabricated through photolithographic processes [5], hot-embossing and molding [6], 3-D printing [7] and the so-called mosquito technique [8].

Up to now, only few demonstrations of the photo-inscription of waveguides in polymers were reported [9,10]. More recently, Pätzold et al. presented the formation of tridimensional waveguides in poly-(methyl methacrylate) (PMMA) based on stress induced structures inscribed using a femtosecond laser [11]. Laser inscription is advantageous since it is flexible, can be used to form complex geometries and carried out in a single-step process.

Lately, we have demonstrated that the laser ablation of polymers can be enhanced by tuning a mid-infrared fiber laser at wavelengths resonant with the C-H bond fundamental stretching frequency [12]. In this communication, we present a novel method based on resonant absorption to form on-surface waveguides in polymers. Mid-IR lasers were tuned close to the absorption peaks of PMMA and polycarbonate and focused on the surface of bulk samples to induce a localized modification of the refractive index of the materials. The laser beam was scanned at high speed ($\sim m \cdot s^{-1}$) across the surface of the samples to process on-surface waveguides. A detailed study of the photo-induced waveguide is presented. The optical properties of the waveguides are also assessed. We report insertion losses of 3.1 dB for a 30 mm long waveguide inscribed in poly-(methyl methacrylate), inferring propagation losses of 0.8 dB cm^{-1}.

Remarkably, the on-surface waveguides can readily be used as refractometric sensors. As a proof of concept, small drops of index matching oils were deposited on the surface of the waveguide sensors. Attenuations of 23.8 and 35.1 dB were recorded for liquids that match the indices of polycarbonate and poly-(methyl methacrylate) respectively. This represents an improvement of the sensitivity spanning orders of magnitudes over previously reported refractive index sensors based on optical waveguides [13,14] or fibers [15]. Our results open a new pathway toward the fast processing of photonic sensing devices based on polymers.

2. Experimental method

Polymer samples (PMMA and polycarbonate) were obtained from an industrial supplier (Plastiver Inc.). The dimensions of the samples are 20 x 30 mm^2 with a thickness of 1.5 and 6 mm for PMMA and polycarbonate respectively. Two mid-infrared fiber laser sources were used to irradiate the surface of the polymer samples. The main source is a high repetition femtosecond laser system with a tunable central wavelength [16]. The laser emits 250 fs pulses at a repetition rate of 60 MHz with a spectrum width of ~50 nm (FHWM). To perform our experiments, the central wavelength was tuned between λ = 3.33 and 3.42 µm. The other source is a watt-level continuous wave fiber laser centered (spectral width $\Delta\lambda$ = 0.6 nm) at a wavelength of 3.425 µm [17]. The output beam of both fiber lasers is collimated and directed toward a galvanometric scanner equipped with a 75mm F-theta lens in order to focus and scan the beam on the surface of the polymer samples. A more detailed description of the inscription set-up is given in [12]. The beam diameters at the focus were 50 µm for the fs laser (set at λ = 3.33 µm) quand 56 µm for the CW laser, both measured with a beam analyzer (NanoScan NS-Pyro/9/5, Ophir).

After the inscription process, the end faces of the samples were cut and polished. Images of the waveguides were taken using a bright-field microscope (Olympus IX71). To measure the photo-induced refractive index modifications, structures were examined using a camera equipped with a bi-dimensional Hartmann grating (Phasics SID4Bio). The camera is a wave-front analyzer that uses lateral shearing interferometry (QWLSI) to yield a quantitative phase image of transparent objects [18]. A methodology similar to the one described in a previous report [19] was implemented to recover the index contrast profiles of the waveguides from the phase map. In order to obtain quantitative phase images of the cross section of the waveguides [20], transverse slices were cut from the 30 mm long sample and polished down to a thickness of 80 µm or less.

Light emitted from a λ = 633 nm laser diode (QPhotonics, QFBGLD-633-20) was injected in the waveguides in order to investigate their guiding properties. The laser diode was connected to a single mode fiber (Thorlabs SM600) and was butt-coupled to the input face of the waveguide. Light transmitted through the waveguide was imaged using a 20X microscope objective (Olympus Lwd-UPLAN) and directed onto either a CCD camera to measure the near-field intensity profile.

Finally, the throughput of the waveguides was evaluated by replacing the pigtailed laser diode with a λ = 633nm He-Ne laser and free space optics. The beam passed through a linear polarizer, a half-wave plate and was focused on the waveguides facet using a 10X (0.25 NA, f = 18 mm) focusing objective (Olympus Lwd-UPLAN). The guided light exiting the waveguide was collimated using the 20X microscope objective, filtered from scattered and stray light using an iris and directed to a power meter (Gentec XLP12-3S).

3. Results

3.1. Resonant photo-inscription of on-surface waveguides

The absorption spectra in the mid-IR of PMMA and polycarbonate were first determined experimentally using a Fourier transform spectrometer as described in ref. 12. The absorption curves of both polymers are plotted in Fig. 1.

These spectra were then used as a reference guide for the irradiation of PMMA and polycarbonate samples with both the cw and fs mid-IR fiber lasers. The central wavelength (λ_c) of the femtosecond laser was tuned at 3.33, 3.365, 3.395 and 3.42 µm and the output power was ranging between 710 and 880 mW. As for the wavelength of the CW laser it was fixed to 3.425 µm ($\Delta\lambda$ = 0.6 nm) with an output power set to 1 W. The beam was scanned linearly across the surface of the samples at speeds ranging between 0.5 and 3 $m \cdot s^{-1}$ and for 1, 3, 5 and 10 successive passes.

First, it was revealed that irradiation with both lasers set at the same central wavelength (λc = 3.42 µm), yield very similar results. At this wavelength resonant absorption dominates the inscription process and the energy is linearly absorbed at the surface of the polymer sample which produces a sustained heat source. Hence the interaction mechanism is essentially photo-thermal and irradiation of the polymers with both lasers results in a localized melt of the material.

As expected, it proved impractical to form optical waveguides in polycarbonate when tuning the laser at λ_c = 3.33 µm which falls outside the strong absorption windows of the polymer. In such case, the optical penetration length (L_α) increases [12] and the energy deposition process becomes inefficient resulting in the formation of large shallow structures spreading over diameters in the range of a hundred microns. Consequently, the formation of a smooth refractive index change is not possible without inducing damage or ablation of the surface. In contrast, when the fs laser is tuned at wavelengths that overlap the absorption spectrum of the polymers, smooth modifications of the material are induced near the surface of the samples using similar irradiation conditions. Images of the longitudinal and cross sections of modifications inscribed using different laser fluences are shown in Fig. 2. The laser fluence is calculated at the center of the irradiated track by integrating the Gaussian irradiance profile over the laser dwell time [8].

Important modifications of the material, appearing as half-circles, are formed just below the surface of the polymer samples. Traces were formed even at the lowest fluence of 0.35 and 0.55 $J \cdot cm^{-2}$ in PMMA and polycarbonate respectively. For traces induced with a single pass of the laser, the depth L of the structure is roughly equal to the sum of the optical (L_α) and thermal (L_{th}) penetration lengths as expected [12]. At low fluence, small shallow traces confined to the vicinity of the surface are formed. The induced traces increase in size with increasing laser fluence or number of successive passes. When the fluence reaches 1 $J \cdot cm^{-2}$ in PMMA and 6.3 $J \cdot cm^{-2}$ in polycarbonate (at λ_c = 3.425 µm), ablation of the surface of the material is observed. Inspection using crossed polarizers also reveals the presence of strong residual mechanical stress along the outer diameter of the photo-induced modifications (not shown).

Since the spectrum of the tunable fs laser is relatively broad, there is resonant absorption from λ_c = 3.36 up to 3.42 µm in both polymers and accordingly, similar results are observed over this wavelength range. It is observed that when the laser is tuned directly over a resonant peak, for

example at λ = 3.39 in PMMA, the optical penetration length (L_α) of the beam is shortened and the laser affected zone confined to a smaller volume [12]. Accordingly, smaller waveguides are then formed. However, the fluence window for the formation of smooth waveguides gets narrower as the threshold for ablation is lessened.

3.2. Optical properties of the on-surface waveguides

The optical properties of the photo-induced index modification traces were thoroughly examined. First, the refractive index profiles of traces inscribed in polycarbonate and PMMA were obtained. In order to generate quantitative phase image of the cross section of the waveguides, transverse slices were cut from cm-long samples and polished down to a thickness of 30 and 80 μm for PMMA and polycarbonate, respectively. The index profiles of the traces are depicted in Fig. 3.

It is revealed that under low laser fluence and for a single pass, the refractive index of both polymers decreases. A small gradient of the index becomes apparent, i.e. the index is lower at the bottom of the structure than near the surface. This index gradient is observed in both polymers but is steeper in polycarbonate. An increase of the index is observed right outside the depressed modification, which is almost undistinguishable from the background in PMMA but readily apparent in polycarbonate. This feature is related to the expansion of the material in the heat affected zone. For higher laser fluence and/or multiple passes of the laser, more complex index profiles are observed. The gradient observed at low fluence grows significantly and becomes apparent in PMMA. The refractive index becomes higher than that of the unaffected material near the surface of both polymers, thus creating a region where light can be guided. This feature of resonant photo-inscription is particularly interesting since it provides a unique scheme to confine light up to the air-material interface.

It also appears that under the tested irradiation conditions, the photo-induced refractive index change is generally more pronounced in polycarbonate than PMMA. The laser induced index change is of $\pm$ 1 x 10^{-3} or less in PMMA while a maximum index contrast close to $\pm$ 4 x 10^{-3} is reached in polycarbonate.

Next, intensity distribution of transmitted light through the waveguides was assessed. Depending on the position of the incident beam on the cross section of the traces, different guided modes are observed at the output face. The near-field intensity profiles measured at the output face of the waveguides are presented in Fig. 4.

Two distinct guiding regions can be defined in both polymers which are directly correlated to the index profiles. Most importantly, an elliptically-shaped guided mode (M1) propagates in a region extending from the very surface of the polymers to a few microns below. M1 can be linked with the index gradient observed near the surface of the photo-induced modification traces as depicted in Fig. 4.

Also, light can be guided (defined as mode M2) underneath the photo-induced structure in the annular region of the compressed material depicted. The circular M2 mode is large, presenting diameters of 20 μm or more. Seldom coupling was observed between the two guided modes M1 and M2; weak power transfer does occur but only in the case of large waveguides inscribed using high fluence. The rippled and noisy intensity profiles of M1 modes observed in polycarbonate are caused by the damaged edges of the sample at the vicinity of the surface.

The propagation losses of the two modes, presented in Table 1, were estimated using the cutback method on waveguides inscribed in the 3 cm long PMMA sample. The total losses of two waveguides were measured at four lengths. Polymers may exhibit intrinsic birefringence and, as such, some waveguides exhibit a preferential transmission axis depending on the orientation of the incident linear polarization. Prior to each transmission measurement the half-wave plate was rotated to optimize the throughput of the waveguides. In the case of polycarbonate, an upperbound for the propagation loss was inferred as the sample was too short to apply the cutback method (L = 1.5 cm). The other column of Table 1 presents the insertion losses, which encompass material attenuation, modal mismatch between the incident Gaussian beam and the guided mode and misalignment. The Fresnel losses at the air-polymer interface were accounted for. It is shown that the insertion losses range between 3 and 4 dB for both modes in PMMA but are much larger in polycarbonate. Especially for the near-surface mode M1, which exhibits insertion losses of 10 dB caused primarily by the damaged edge at both facets of the sample. In PMMA, the insertion losses are caused mainly by a significant modal mismatch.

Propagation losses also follow a similar trend, being estimated to be ranging between 0.6 and 0.8 $dB \cdot cm^{-1}$ in PMMA, and between 2.2 and 3.4 $dB \cdot cm^{-1}$ in polycarbonate. Further studies would be needed to explain the difference in overall transmission between the two materials since the bulk attenuation is comparable, being of 0.22 $dB \cdot cm^{-1}$ for PMMA and 0.35 $dB \cdot cm^{-1}$ for polycarbonate. Nevertheless, the transmission of the waveguides is good enough for most targeted sensing and probing applications as shown below.

3.3. Highly sensitive refractometric sensors

As a proof of concept, the surface waveguides were used as refractometric sensors. For the experiment, the best waveguides in terms of the transmission of the M1 mode were selected for each material. Index matching oils (Cargille) were put in contact with the surface of the waveguide sensor. The transmitted light was butt-coupled to a multimode optical fiber (Thorlabs MM30, core diameter of 30 μm) and directed to an optical spectrum analyzer (Yokogawa AQ6373). To ensure repeatability and quantify the amount of liquids that affect light propagation in the waveguide, small 4 mm wide bands of optics cleaning tissue were soaked in the liquid. The bands were then carefully placed upon the surface of the sample across the longitudinal axis of the waveguides, so as to limit the interaction length between the guided light and the liquid to 4 mm. The transmitted power through the waveguide was recorded prior (P0) to the measurement and approximately one minute after the liquid was placed on the samples and the power (P) had stabilized. The relative output (P/P_0) of the sensor plotted against the refractive index of the liquids is presented in Fig. 5.

The transmitted light experiences a very sharp attenuation when the index of the oil exactly matches the index of the polymer and reaches a plateau for higher indices. This is in agreement with results and theoretical predictions reported for sensors based on the interaction of guided waves in close proximity with external media [21]. It is revealed that the waveguides can act as highly sensitive refractometric sensors. Indeed, an interaction length of only 4 mm with liquids exhibiting an index close to the index of the polymer suffices to severely attenuate the transmitted light. Extinction ratio of 23.8 and 35.1 dB were recorded for oils that match the index of polycarbonate and PMMA respectively. To our knowledge, this represents the most sensitive measurement of the refractive index using an optical waveguide or fiber optics by orders of magnitude [13,15]. Furthermore, a significant and readily measurable attenuation is also observed for an index higher than the index of the substrate. Our results highlight the great potential of on-surface waveguides inscribed in polymers for sensing applications and pave the way for the development of a wide range of novel sensors and photonic devices based on evanescent coupling.

Such sensitivity represents a highly desirable feature that transcends the context of refractometry. Given the nature of the inscription process, light is guided directly under the surface and thus, evanescent coupling with any object on the surface is maximized de facto. This attribute could allow for, among numerous benefits, enhanced plasmonic coupling with metallic nanoparticles, facilitated interaction with molecules and detection of very small concentrations of a chemical compound.

4. Conclusion

We presented a novel fabrication technique for the fabrication of on-surface photonic devices in polymers. For the first time, mid-IR lasers were used to form optical waveguides on the surface of poly-(methyl methacrylate) and polycarbonate by taking advantage of resonant absorption. It was revealed that a smooth refractive index change can be induced when the wavelength of the laser is tuned near the absorption peaks associated to the stretching vibrations of the C-H bonds. The single-step inscription method is carried out at speeds of the order of the $m \cdot s^{-1}$ enabling fast, large scale processing of on-surface waveguides in polymers.

Complex refractive profiles composed of regions that exhibit positive and negative index change are observed. The magnitude of the index change was evaluated to be of $\pm 1 \times 10^{-3}$ and $\pm 4 \times 10^{-3}$ in PMMA and polycarbonate respectively. Using sufficient laser fluence, an index gradient is formed extending from the surface to a few microns below the surface, creating a region where light can be guided by total internal reflection. Accordingly, we showed that an elliptical guided mode propagates directly beneath the surface of the polymer samples. Insertion losses of 3.1 and 6.3 dB were recorded for visible light transmitted through the near-surface waveguides in PMMA and polycarbonate respectively.

The waveguide's morphology allows light confinement up to the very interface of the polymer with the external medium, and thus, opens novel avenues for enhanced sensing and probing applications. As a demonstration, the waveguides were used as refractive index sensors. Liquids with different indices were put in contact to cover the surface above the photo-induced waveguides. Attenuations of 23.8 and 35.1 dB were recorded for liquids that match PMMA and polycarbonate indices, highlighting unparalleled sensitivity and precision for a refractometric sensor based on directly laser written optical waveguides. Our results pave the way for the fast processing of novel photonic devices in polymers. Most specifically, it enables the development of low-cost highly sensitive sensors to be deployed in a wide range of scientific and industrial domains.

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References

1. J. Clark and G. Lanzani, "Organic photonics for communications," Nat. Photonics 4(7), 438–446 (2010).
2. B. Ibarlucea, C. Fernandez-Sanchez, S. Demming, S. Buttgenbach, and A. Llobera, "Selective functionalization of PDMS-based photonics lab on a chip for biosensing," Analyst 136(17), 3496 (2011).
3. M. Moirangthem, R. Arts, M. Merkx, and A. P. H. J. Schenning, "An optical sensor based on a photonic polymer film to detect calcium in serum," Adv. Funct. Mater. 26, 1154–1160 (2016).
4. H. Ma, A. K.-Y. Jen, and L. R. Dalton, "Polymer-based optical waveguides: Materials, Processing and Devices," Adv. Mater. 14(19), 1339–1365 (2002).
5. Z. Nie and E. Kumecheva, "Patterning surfaces with functional polymers," Nat. Mater. 7(4), 277–290 (2008).

6. O. Rötting, W. Röpke, H. Becker, and C. Gärtner, "Polymer microfabrication technologies," Microsyst. Technol. 8(1), 32–36 (2002).
7. T. Merkle, R. Götzen, J.-Y. Choi, and S. Koch, "Polymer multichip module process using 3-D printing technologies for D-band applications," IEEE Trans. Microwave Theory Tech. 63(2), 481–493 (2015).
8. K. Yasuhara, F. Yu, and T. Ishigure, "Circular core single-mode polymer optical waveguide fabricated using the Mosquito method with low loss at 1310/1550 nm," Opt. Express 25(8), 8524–8533 (2017).
9. A. Baum, P. Scully, M. Basanta, C. Paul Thomas, P. Fielden, N. Goddard, W. Perrie, and P. Chalker, "Photochemistry of refractive index structures in poly(methyl methacrylate) by femtosecond laser irradiation," Opt. Lett. 32(2), 190–192 (2007).
10. S. Sowa, W. Watanabe, T. Tamaki, J. Nishii, and K. Itoh, "Symmetric waveguides in poly(methyl methacrylate) fabricated by femtosecond laser pulses," Opt. Express 14(1), 291–297 (2006).
11. W. Patzold, C. Reinhardt, A. Demircan, and U. Mörgner, "Cascaded focus laser writing of low-loss waveguides in polymers," Opt. Lett. 41(6), 1269–1272 (2016).
12. C. Frayssinous, V. Fortin, J. P. Bérubé, A. Fraser, and R. Vallée, "Resonant polymer ablation using a compact 3.44 μm fiber laser," J. Mater. Process. Technol. 252, 813–820 (2018).
13. A. A. Khalil, J. P. Bérubé, S. Danto, T. Cardinal, Y. Petit, R. Vallée, and L. Canioni, "Direct laser writing of near-surface waveguides in silver containing glasses with no additional processing," Proc. SPIE 10908, 27 (2019).
14. J. Lapointe, F. Parent, E.S. de Lima Filho, S. Loranger, and R. Kashyap, "Toward the integration of optical sensors in smartphone screens using femtosecond laser writing," Opt. Lett. 40(23), 5654 (2015).
15. N. Jing, J. Zheng, X. Zhao, and C. Teng, "Refractive index sensing based on a side-polished macrobending plastic optical fiber," IEEE Sens. J. 15, 1 (2014).
16. S. Duval, J. Gauthier, L.-R. Robichaud, P. Paradis, M. Olivier, V. Fortin, M. Bernier, M. Piché, and R. Vallée, "Watt-level fiber-based femtosecond laser source tunable from 2.8 to 3.6 μm," Opt. Lett. 41(22), 5294 (2016).
17. V. Fortin, F. Maes, M. Bernier, S. Bah, M. D'Auteuil, and R. Vallée, "Watt-level erbium-doped all-fiber laser at 3.44 μm," Opt. Lett. 41(3), 559–562 (2016).
18. P. Bon, G. Maucort, B. Wattelier, and S. Monneret, "Quadriwave lateral shearing interferometry for quantitative phase microscopy of living cells," Opt. Express 17(15), 13080 (2009).
19. E. Bélanger, J. Bérubé, B. De Dorlodot, P. Marquet, and R. Vallée, "Comparative study of quantitative imaging techniques for refractometry of optical waveguides," Opt. Express 26(13), 17498–17510 (2018).
20. E. Ampem-Lassen, T. Huntington, N. Dragomir, K. Nugent, and A. Roberts, "Refractive index profiling of axially symmetric optical fibers: a new technique," Opt. Express 13(9), 3277 (2005).
21. S.-M. Tseng and C.-L. Chen, "Side-polished fibers," Appl. Opt. 31(18), 3438 (1992).

Table 1. Propagation loss of the guided modes in both polymers

Material	Mode	Insertion loss (dB)	Propagation loss (dB·cm^{-1})
PMMA	M1	3.1 ± 0.5	0.6 ± 0.2
PMMA	M2	3.8 ± 0.5	0.8 ± 0.2
polycarbonate	M1	10.2 ± 0.8	6.8 ± 0.5[a]
polycarbonate	M2	3.7 ± 0.5	2.2 ± 0.4

[a]Upperbound, the cut-back technique could not be applied to M1.

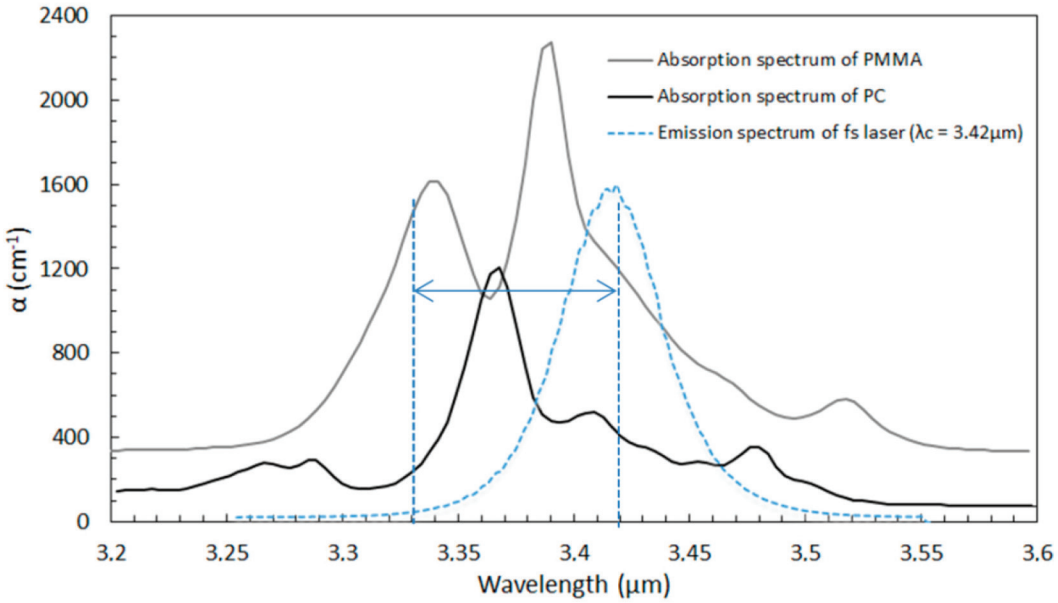

Fig. 1. Absorption spectra of PMMA and polycarbonate along with the emission spectrum of the tunable mid-IR laser. The region delimited by the blue arrow depicts the tunable range of the central wavelength of the femtosecond laser.

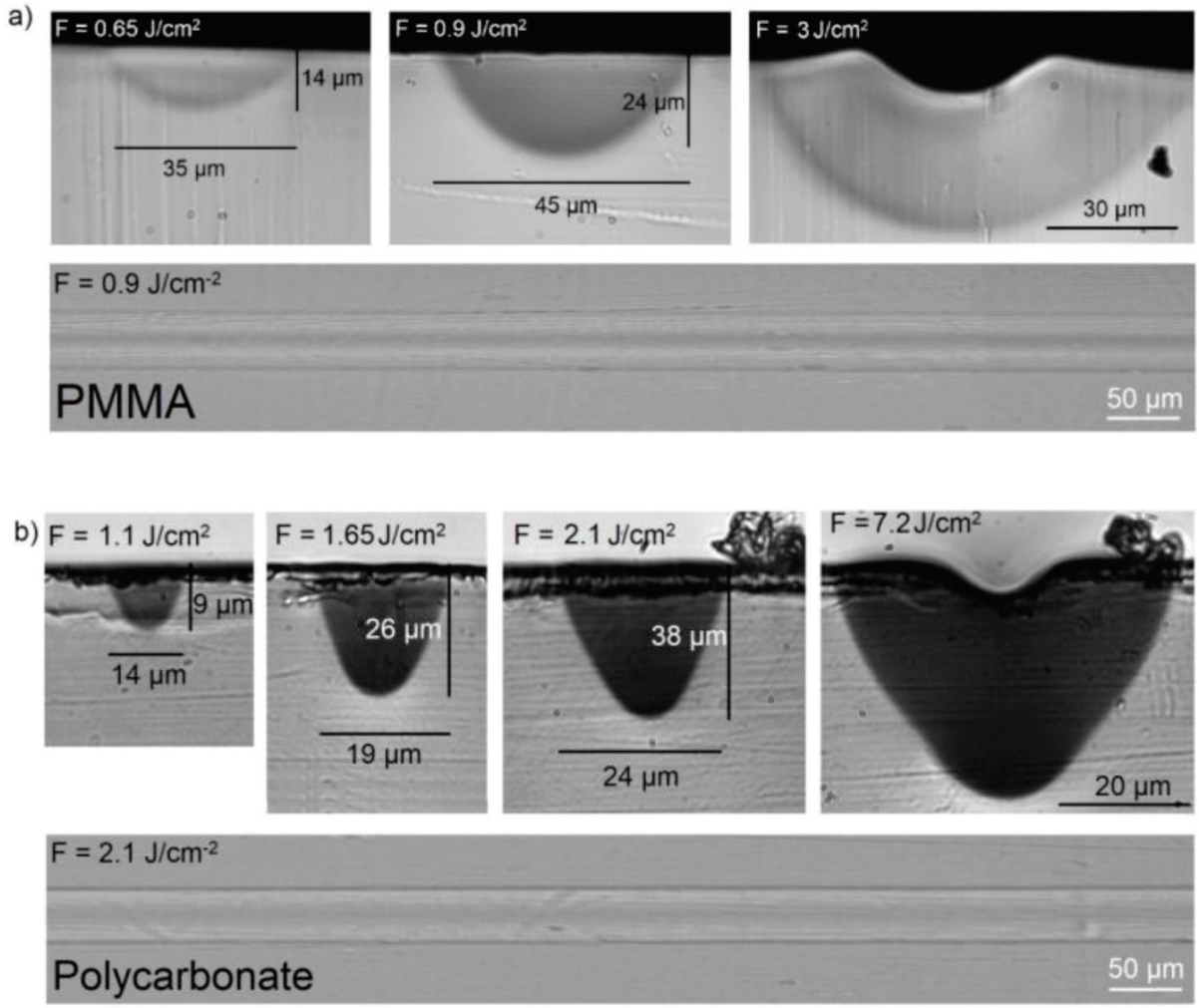

Fig. 2. a) Brightfield images of the cross section and longitudinal section of photo-induced modifications inscribed using a single pass of the laser in a) PMMA and b) polycarbonate. The cw laser was used to inscribed the modifications ($\lambda_c = 3.425$ µm).

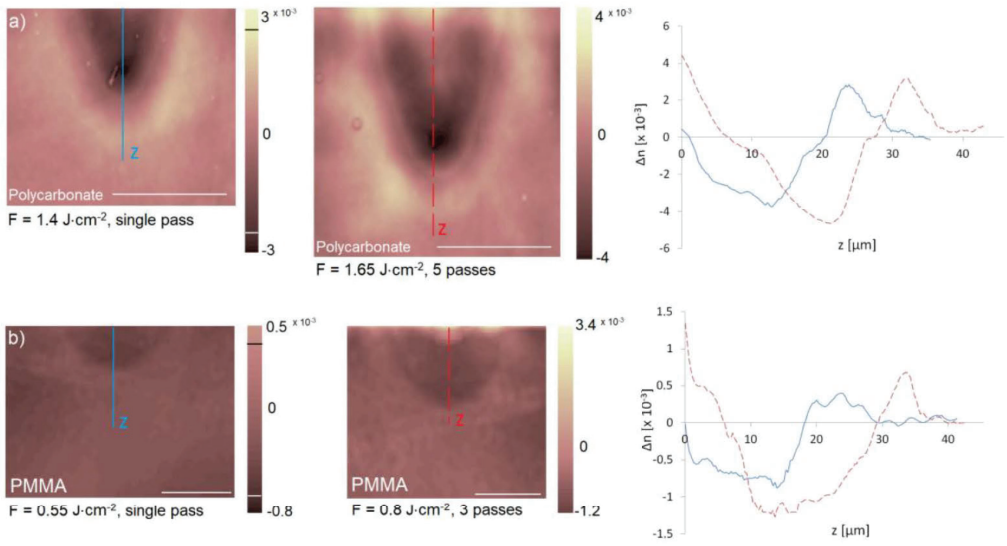

Fig. 3. Refractive index profiles (~2 µm below the surface) in a) Polycarbonate and b) PMMA using different laser fluences and number of successive passes. On the right: plot of the data points taken along the blue (solid) and red (dashed) lines. The white scale bars in the phase images equal 20 µm. For feasibility reasons, the phase image had to be taken about two microns below the surface. The cw laser was used to inscribed the modifications ($\lambda_c = 3.425\,\mu m$).

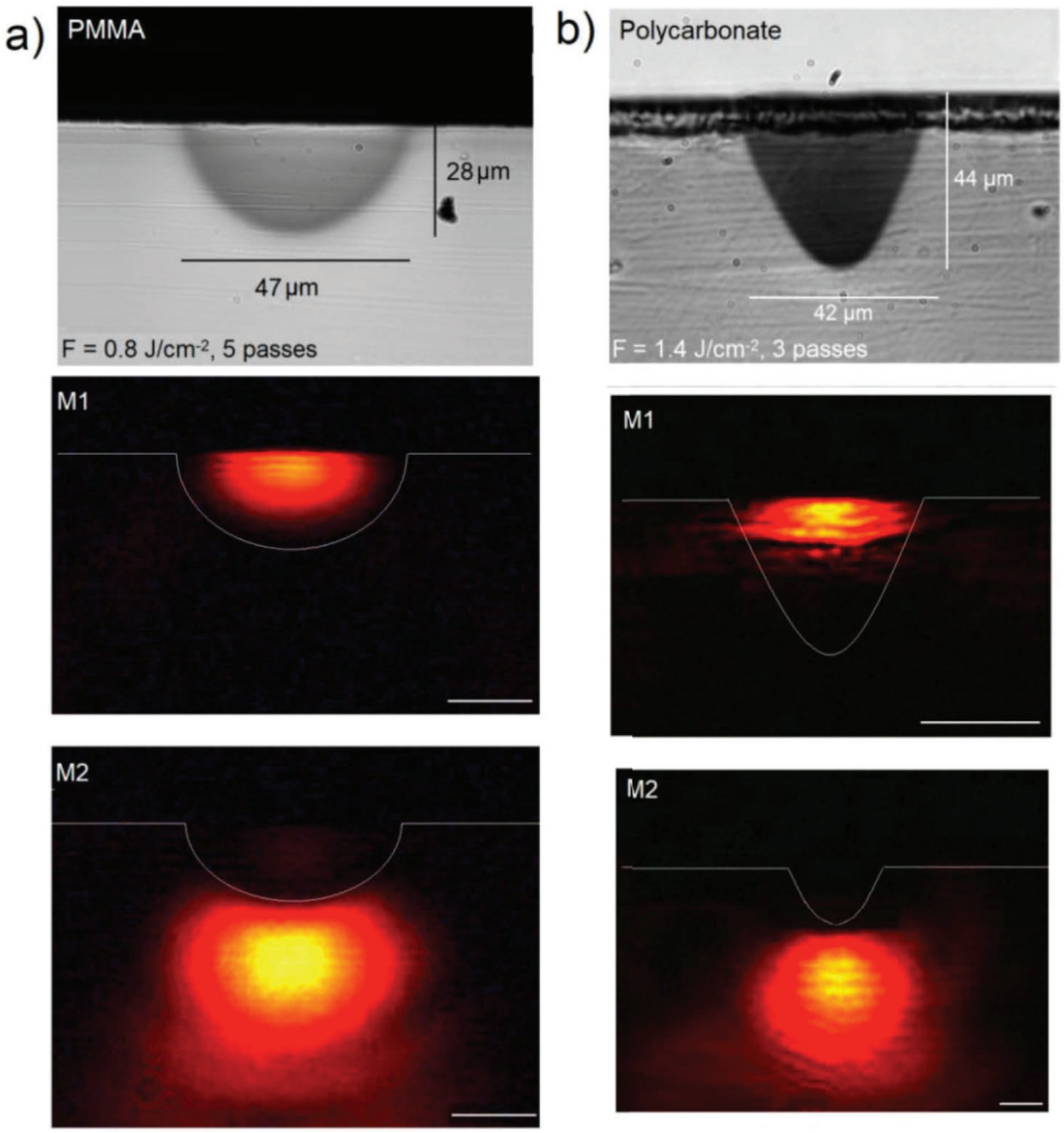

Fig. 4. Near-field intensity profile of $\lambda = 633\,\text{nm}$ light transmitted through waveguides inscribed in a) PMMA and b) polycarbonate. The white scale bar equals 20 µm. The boundary of the photo-induced modification area along with the surface of the sample were added to the near-field images for illustrative purposes. The cw laser was used to inscribed the waveguides ($\lambda_c = 3.425\,\mu\text{m}$).

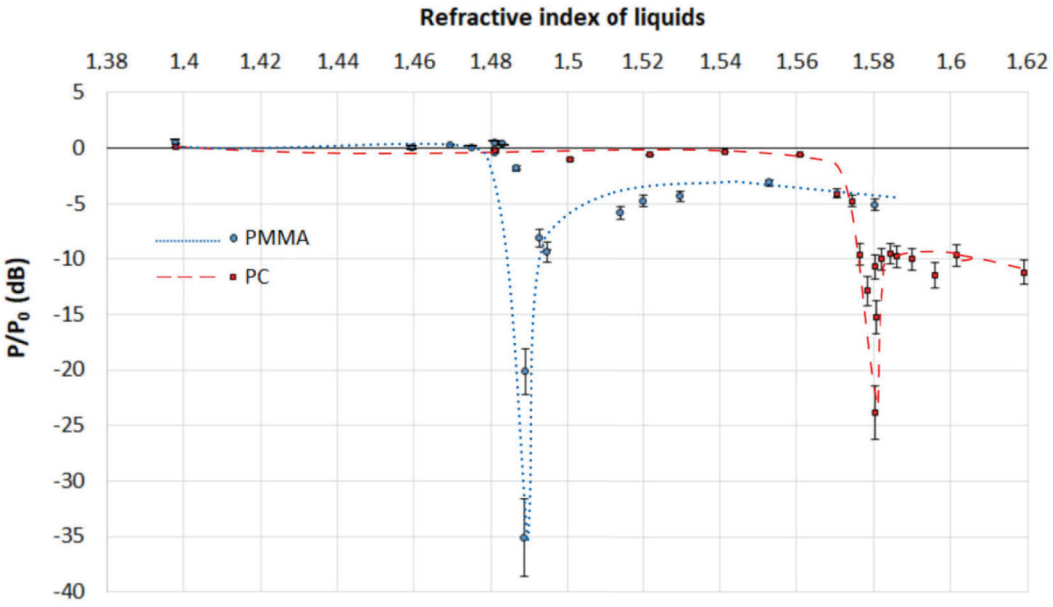

Fig. 5. Ratio of the transmitted power with (P) and without (P_0) the soaked tissues deposited on the surface of the sensor waveguides.