# Planar polymer waveguides with a graded-index profile resulting from intermixing of methacrylates in closed microchannels

Jeroen Missinne[‡,*,1], Lara Misseeuw[‡,2], Xiang Liu[3], Patrick S. Salter[3], Geert Van Steenberge[1], Kehinde Adesanya[4], Sandra Van Vlierberghe[2,4], Martin J. Booth[3], Peter Dubruel[4]

[1]Centre for Microsystems Technology, Ghent University & imec, Technologiepark 914A, Ghent B-9052, Belgium.
*E-mail: jeroen.missinne@elis.ugent.be

[2]Dept. of Applied Physics and Photonics (IR-TONA), Brussels Photonics Team (B-PHOT), Vrije Universiteit Brussel, Pleinlaan 2, B-1050 Brussels, Belgium

[3]Dept. of Engineering Science, University of Oxford, Parks Road, Oxford OX1 3PJ, United Kingdom

[4]Polymer Chemistry & Biomaterials Research Group, Ghent University, Krijgslaan 281 (S4 Bis), Ghent B-9000, Belgium.

‡ J. Missinne and L. Misseeuw contributed equally to this work as co-first authors.

*E-mail: jeroen.missinne@elis.ugent.be

## Abstract

Graded-index waveguides are known to exhibit lower losses and considerably larger bandwidths compared to step-index waveguides. The present work reports on a new concept for realizing such waveguides on a planar substrate by capillary filling microchannels (cladding) with monomer solution (core). A graded-index profile is obtained by intermixing between the core and cladding material at the microchannel interface. To this end, various ratios of methyl methacrylate (MMA) and octafluoropentyl methacrylate (OFPMA) were evaluated as starting monomers and the results showed that the polymers P50:50 (50:50 MMA:OFPMA) and P0:100 (100% OFPMA) were suitable to be applied as waveguide core and cladding material respectively. Light guiding in the resulting P50:50/P0:100 waveguides was demonstrated and the refractive-index profile was quantified and

compared with that of conventional step-index waveguides. The results for both cases were clearly different and a gradual refractive index transition between the core and cladding was found for the newly developed waveguides. Although the concept has been demonstrated in a research environment, it also has potential for upscaling by employing drop-on-demand dispensing of polymer waveguide material in pre-patterned microchannels, for example in a roll-to-roll environment.

**Keywords:** Capillary filling, graded-index, intermixing, microchannel, polymer waveguide.

## 1. Introduction

Optical waveguides are mostly known in the form of fibers but can also be implemented on a planar substrate. For long-distance communication, only silica based fibers are currently used because of their unprecedentedly low propagation loss. On the other hand, for shorter optical links (i.e. several centimeters to meters) or for sensor applications [1], fibers or planar waveguides constituted from polymers are considered attractive alternatives [2].

Although the transparency of polymers is lower than that of silica, polymer-based optical materials allow rapid prototyping, cost-effective fabrication and implementation of high-density interconnections and complex circuits on planar substrates. The use of these planar substrates, which can be rigid, flexible or stretchable [3], for implementing the waveguides increases the level of integration, enables implementation of complex circuits and may also lead to clear cost advantages compared to using individual fibers for every single connection.

Compared to the fabrication of fibers, which are in fact extruded versions of a macroscopic preform, the fabrication of waveguides on a planar substrate is clearly different and is generally performed in a sequential manner. First, the lower cladding layer is applied using a suitable coating technique. In a second step, the waveguide core is patterned (e.g. using standard photoresist-based lithography and etching, direct-write technologies, or replication-based technologies [4, 5]) and finally, another cladding layer is applied to cover the core. These traditional approaches for realizing planar optical waveguides, although well-established and therefore reliable, show some inherent disadvantages. First, expensive equipment is required to fabricate the fine core structures (which can exhibit dimensions down to several microns) with the required optical quality (down to several nanometers in terms of surface roughness) since any roughness at the core-cladding interface has a significant impact on the waveguide losses. Secondly, there is limited flexibility in the waveguide design. Although the size of the core diameter can be selected, its refractive index is constant and determined by the applied material leading to a step-index waveguide profile. However, especially in the case of multimode waveguides, it is known that a graded-index waveguide (with a gradual refractive index transition between core and cladding) can have lower crossing and bending losses and can greatly reduce the dispersion and therefore increase the bandwidth by a few orders of magnitude [6-9].

Although common in case of fibers [9], achieving such a graded-index profile in waveguides on a planar substrate is not straightforward. A recently developed method, i.e. the so-called ‘mosquito technique’[10], dispenses the waveguide core monomer directly into a layer of cladding monomer material and a graded-index profile results from subsequent inter-diffusion between both components. In a second approach, waveguides are obtained by coating a core material on top of microchannels consisting of not-completely cured cladding monomer. After coating, the excess core material is wiped out, and the graded-index profile results from diffusion of a dopant out of the core material [11].

Herein, we present an alternative concept, see Figure 1 (a): the graded-index profile is obtained by intermixing after capillary filling of closed microchannels (constituted by the cladding polymer formulation) with another polymer formulation (characterized by a higher refractive index) which forms the core. An advantage of this single-step methodology is that uniform and completely closed microchannels are filled resulting in the same graded-index profile along all radial directions. Additionally, since the filling is controlled on the level of the individual microchannels, the methodology is compatible with the recently reported concept of bend-insensitive waveguides, where low refractive index trenches are applied on both sides of the waveguide to reduce bend losses even further [12].

The current paper demonstrates this concept using methacrylate-based polymer chemistry, synthesized upon mass polymerization and initiated with UV-irradiation, but it can be anticipated that the concept will also be applicable starting from other types of polymers (e.g. commercially available optical polymers), as long as they are optimized for waveguide fabrication and a certain degree of intermixing is allowed at the core-cladding interface upon capillary filling. Furthermore, when more substrates or longer waveguides are required, we propose a slightly different concept allowing process upscaling. Although the basic principle remains the same, channel filling should then ideally be achieved using a drop-on-demand technology such as inkjet printing of the liquid (monomer) core material (see Figure 1 (b)). After dispensing the controlled amount of waveguide material, the cladding layer is laminated on top for closing the channels and subsequent UV-curing of the core can be initiated. The methacrylate-based materials described in the present paper have been developed in-house for future inkjet printing purposes, but as an alternative, commercially available materials could also be used [13].

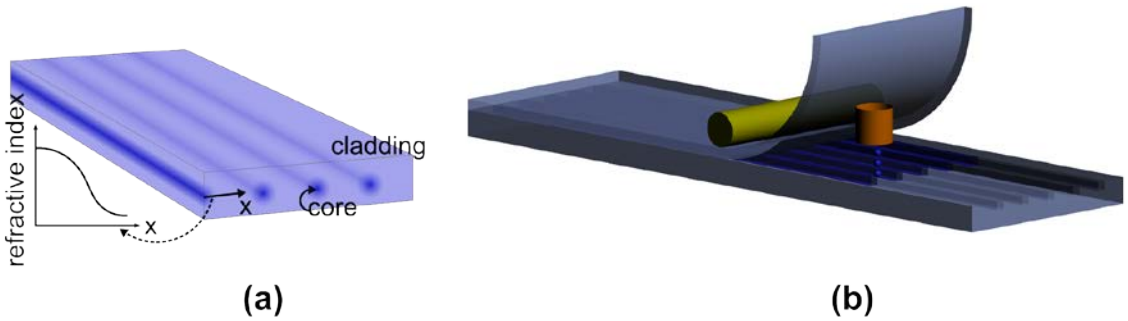


**Figure 1.** (a) The principle of graded-index waveguides formed in a planar layer, (b) a concept allowing upscaling of the fabrication process.

## 2. Materials

Inspired by the composition of PMMA-based graded-index plastic optical fibers [8], two monomers were selected as the building blocks for the optical waveguide production, i.e. methyl methacrylate (MMA) and octafluoropentyl methacrylate (OFPMA). By combining these monomers in different ratios, (co)polymers with different refractive index were synthesized and were labeled according to their MMA:OFPMA (x:y) ratio in mol %, i.e. $M_{x:y}$ (for the monomer solutions) and $P_{x:y}$ (for the corresponding polymers). A higher ratio of OFPMA leads to lowering of the refractive index (because of the increased fluorine amount) [2, 9]. Both materials were obtained from Sigma Aldrich and distilled prior to use. For polymerizing the material, mass polymerization (pure monomer in the liquid phase and adding a soluble initiator) was applied, initiated upon exposure to UV-light. The monomers

were mixed in the appropriate ratios and 4 mol % of the photo-initiator Irgacure 651 (BASF) was added. The conversion of the polymerization was fully achieved (~100%) when illuminated for two hours using UV-light (UV-A, 365 nm, 13 mW/cm$^2$).

The $P_{50:50}$ and $P_{0:100}$ were selected as waveguide core and waveguide cladding material respectively based on 2 main criteria: their refractive index (RI) and their compatibility with the fabrication process. The RI of polymers $P_{50:50}$ and $P_{0:100}$ were found to be 1.41 and 1.39 respectively yielding a waveguide with a numerical aperture (NA) equal to $(RI_{P50:50}\text{-}RI_{P0:100})^{0.5} \approx 0.24$, which is similar to and therefore compatible with standard silica graded-index multimode optical fibers. Furthermore, $P_{0:100}$ was found to be the most compatible material for structuring using a polydimethylsiloxane (PDMS) mold which was required for the fabrication process.

More details regarding the structural and thermal analysis of the materials and the applied method for determining their refractive indexes can be found in the Supplementary Data file.

## 3. Methods

### *3.1. Waveguide fabrication*

To obtain graded-index waveguides starting from these methacrylates, an elegant replication process was established based on soft-lithography and capillary filling of microchannels [3, 5], as illustrated in Figure 2. First, a polydimethylsiloxane (PDMS, Sylard®184, Dow Corning) substrate having 50 µm wide, 50 µm high ridges was used as a mold to pattern microchannels in $P_{0:100}$ (POFPMA) (step i). To this end, an 80 w% $P_{0:100}$ solution in acetone was prepared and 150 µL of this solution was spin-coated (1000 rpm during 60 s, acceleration 300 rpm/s) on a glass substrate (24 mm x 40 mm and a thickness of 140 µm). The latter yielded a uniform and transparent $P_{0:100}$ coating on glass. Subsequently, a drop of 0.5 ml of $M_{0:100}$ monomer solution was dispensed (100 mol% OFPMA and 4 mol% Irgacure 651 as initiator) on the spin-coated layer and the PDMS mold was placed on top. Although the $M_{0:100}$ monomer drop slightly dissolved the earlier spin-coated layer, the spin-coated layer was needed to make the cladding thicker. After UV-induced polymerization of the $M_{0:100}$ monomer for 2 hours (UV-A, 365 nm, 13 mW/cm$^2$), the PDMS mold could be removed and the PDMS structures were replicated into the polymerized $P_{0:100}$ material. Next, this structured layer of $P_{0:100}$ was bonded (vacuum, room temperature, 0.5 atm) to a uniform $P_{0:100}$ coating (spin-coated on a second glass substrate) to realize closed microchannels in the $P_{0:100}$ material which were sandwiched between two glass substrates (step ii). Subsequently, these channels were capillary filled by placing a droplet of $M_{50:50}$ monomer solution at one side of the channels (step iii). Owing to the low viscosity of the $M_{50:50}$ solution, the channels were filled within a few seconds and afterwards, the sample was UV-irradiated (UV-A, 365 nm, 13 mW/cm$^2$) for 1 hour to cure the core material resulting in the final waveguides (step iv).

The two glass substrates were only added for ease of processing and subsequent microscopic analysis; in a production process, they could for example be replaced by a (temporary) carrier or foil [14].

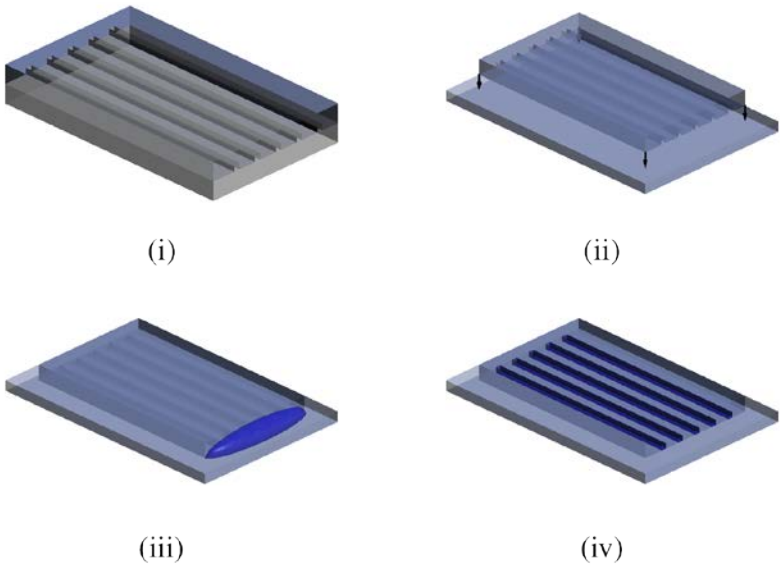


**Figure 2.** The evaluated fabrication process based on capillary filling of microchannels.

The resulting refractive index profile is compared with that of know step-index waveguides fabricated using a traditional, sequential fabrication approach. Those waveguides were prepared as follows. First, a uniform, ~50 µm thick cladding layer (EpoClad, MicroResist Technology) was applied onto a Borosilicate glass substrate using spin-coating, followed by a baking step to remove the solvent. Afterwards, the cladding layer was polymerized using UV-exposure and further curing was achieved by a series of baking steps. Then, a uniform 50 µm thick core layer (EpoCore, MicroResist Technology) was applied onto a Borosilicate glass substrate using spin-coating, followed by a baking step to remove the solvent. The waveguide structures (width 50 µm) are defined by locally UV-illuminating the core layer using a Direct-Write Laser Lithography system (Heidelberg DWL66+). After local UV-exposure, the unexposed (unpolymerized) material was removed in a dedicated developer solvent (mrDev600, MicroResist Technology), and finally, another ~50 µm thick cladding layer (EpoClad, MicroResist Technology) was applied using spin-coating, and undergoes similar processing as the first cladding layer.

### *3.2. Waveguide refractive index profiling*

A third harmonic generation (THG) laser scanning microscope was used to reveal interfaces between media of different refractive index (RI) or third-order nonlinear susceptibility ($\chi(3)$) [15]. The microscope was equipped with adaptive optic capabilities for compensating sample induced aberrations thus maintaining optimum resolution at all points in the sample [16].

Quantitative phase microscopy (QPM) [17] was used to determine the magnitude of the refractive index change between core and cladding in the waveguide. Two slightly defocused intensity images from a widefield transmission microscope with partially coherent illumination reveal the intensity gradient along the optical axis. This data can be used to solve the transfer of intensity equation (TIE) and hence infer the cumulative optical path length of light passing through the waveguide.

## 4. Results and discussion

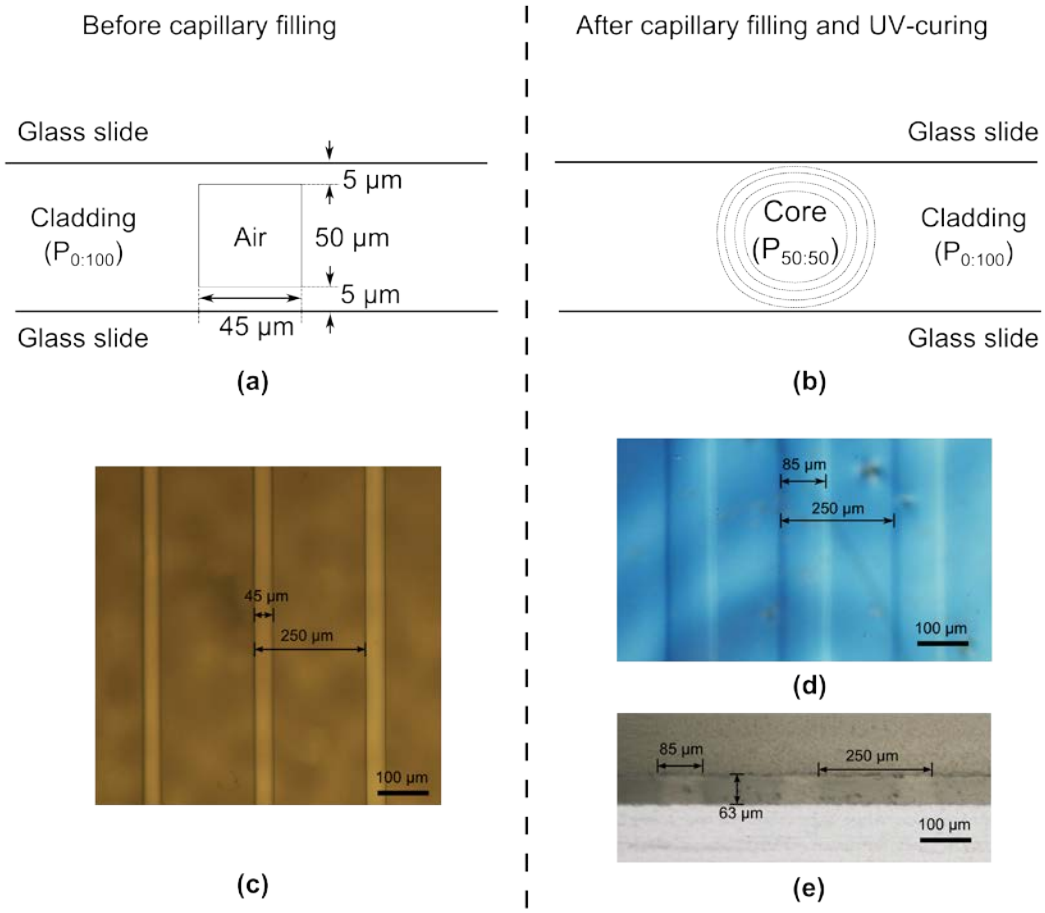


**Figure 3.** Comparison of the microchannels before and after capillary filling and UV-curing: (a-b) cross-sectional schematic configuration, (c-d) comparison of top view microscope images, (e) cross-sectional microscope image after filling.

Figure 3 illustrates the principle of the graded-index waveguide formation comparing the configuration just before (Figure 3 (a)) the capillary filling step and after capillary filling and UV-curing (Figure 3 (b)). This is confirmed by observing the microchannel: after capillary filling and subsequent UV-curing, both the width as well as the height of the channels increases (Figure 3 (c-e)). This is a first indication of intermixing between $P_{50:50}$ and $P_{0:100}$ materials at the channel sidewalls resulting in slightly broader waveguides but with a transition region between core and cladding. This was particularly noticeable from the blurred waveguide boundaries as seen in the top view microscopic images.

In addition to the favorable graded-index profile, this intermixing process also results in waveguides with excellent adhesion between the core and the cladding, which is a common problem when polymer waveguides are fabricated in a sequential manner. Secondly, this effect eliminates the roughness at the core-cladding interface, which is often the cause of excessive waveguide losses. Finally, because of the capillary filling of closed channels, there is no residual layer between the waveguides which could

cause cross-talk and the graded-index profile is generated in a single step, making it uniform in all radial directions.


**Figure 4**. Light confined in a fabricated planar graded-index waveguide

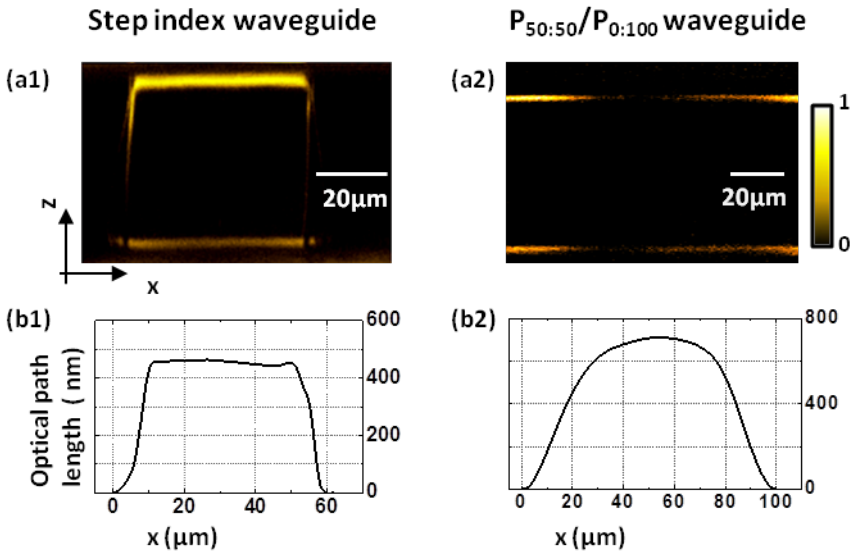


**Figure 5.** (a) Waveguide cross-sectional images measured via THG microscopy. (b) The cumulative optical path length through the same waveguides, measured by QPM.

Light guidance in the fabricated waveguide structures was demonstrated by coupling light (635 nm) in the waveguides using an optical fiber (50 µm core diameter graded-index multimode silica fiber, NA 0.2) on one side and imaging the other side using a CCD camera with a microscope objective (Figure 4). This simple experiment clearly shows that the light is confined inside the waveguide, but does not yet reveal information about its RI profile. The transverse RI profile of the waveguides was investigated using a variety of microscopy techniques, which evaluated the sample from the top. This non-destructive technique is advantageous over RI profiling strategies in which cross-sections of the waveguide need to be prepared for profiling. Firstly, these techniques are destructive, and secondly they require preparation of precise cross-sections which is challenging and introduce to errors.

Figure 5 (a) shows 2D images of the waveguide morphology in the plane cross-sectional to the waveguide axis obtained using third harmonic generation (THG) microscopy revealing interfaces between media of different refractive index (RI) or third-order nonlinear susceptibility ($\chi(3)$) [15, 16]. Data is shown comparing standard waveguides with a known step-index profile and nearly square 50 x 50 $\mu m^2$ core, and the new $P_{50:50}/P_{0:100}$ waveguides. For the waveguide with known step-index profile in

the first image, RI boundaries are uncovered with fine detail. It can be noticed that the thickness of the upper and lower boundaries appears greater than any of the near vertical interfaces, which is caused by the slightly reduced resolution of the microscope in the axial direction of the probe laser beam. In this set-up, the axial resolution (z-axis) was approximately 1.3 μm, while the lateral resolution (x-axis) was around 500 nm.

The strong signal in the THG image for the $P_{50:50}/P_{0:100}$ waveguide (Figure 5 (a)) is mostly due to the large $RI/\chi^{(3)}$ change between the substrate glasses and the cladding material. There is no sign of a sharp interface bounding the $P_{50:50}/P_{0:100}$ waveguide in the x direction, as seen for the step-index waveguide in Figure 5 (a1). This indicates that the $P_{50:50}/P_{0:100}$ waveguide has a gradual $RI/\chi^{(3)}$ transition, since the THG signal is negligible when the RI change is small over a distance comparable to the focal spot of the microscope (~500nm). It is also interesting to note that the THG signal arising at the glass substrate drops markedly in the presence of the waveguide, denoting a smaller change in $RI/\chi^{(3)}$ between the polymer and the glass. This suggests that there is significant diffusion of the $P_{50:50}$ monomer in the z direction through the $P_{0:100}$ layer, modifying the polymer composition at the glass substrate.

In order to further probe the RI profile, quantitative phase microscopy (QPM) was applied and the results are presented in Figure 5 (b) [17]. The results show the accumulated optical path length (OPL) through the waveguide in the z direction: $OPL(x) = \int \delta n(x,z)\, dz$, where $\delta n(x,z)$ is the difference in refractive index between point (x) and the cladding. The OPL profile for the step-index waveguide in Figure 5 (b1) is consistent in width with the measurements of waveguide morphology from the THG microscope. An estimation of the waveguide dimension in *z* from the THG image (51 μm) combined with knowledge of the expected RI difference between core and cladding ($\delta n$ = 0.009) gives an expected OPL of ~460 nm. There is a good correspondence between this number and the magnitude of the OPL from the QPM measurements, confirming the applicability of our RI profiling strategy.

Consulting Figure 5 (b2) showing the OPL for the $P_{50:50}/P_{0:100}$ waveguide, we initially note that the OPL width is much greater than the initial 50 μm width of the channel in the $P_{0:100}$ material prior to capillary filling. Furthermore, the OPL profile exhibits a much lower gradient along the x-axis relative to that of the step-index waveguide. The initial height of the channel before capillary filling was 50 μm and the RI difference between $P_{50:50}$ and $P_{0:100}$ is 0.02 indicating an OPL of ~1000 nm before the intermixing. The measured peak OPL magnitude of just 711nm at the center of the waveguide structure shows the effect of the intermixing and all this provides further compelling evidence of the graded-index nature of the waveguide.

## 5. Conclusions

A new concept for obtaining graded-index polymer waveguides on a planar substrate was presented and evaluated. The desired gradual refractive index transition was obtained by intermixing between methacrylate-based core and cladding materials at a microchannel interface after capillary filling.

A first indication of the intermixing effect was supported by the increased final waveguide width compared to the initial channel dimensions and the rather indistinct waveguide boundaries observed when inspected by optical microscopy. Secondly, several quantitative techniques (THG microscopy and QPM) were applied for mapping the refractive index distribution inside a material and revealed the graded-index nature of these new waveguides.

This new concept was demonstrated using methacrylate-based chemistry and lab-scale processes. However, it could also be applied starting from commercially available materials and has potential for

scaling up by employing drop-on-demand dispensing of waveguide materials in pre-patterned microchannels.

The development of such graded-index waveguides with a gradual transition between core and cladding is important because they enable considerably larger communication bandwidths compared to step-index waveguides and lower losses compared to planar waveguides fabricated in a sequential way by avoiding roughness at the core-cladding interface.

## Acknowledgements

J. Missinne and L. Misseeuw contributed equally to this work as co-first authors.

This work was partially supported by ERC (ERC-FP7/2007-2013 grant 336940); and by the Flemish Government (through the SBO project “Self-Sensing Composites (SSC)”, IWT-nr 120024); and by Ghent University (GAO, “Flexible electro-photonic integration platform”); and by UK EPSRC grant EP/K034480/1.

## References

[1] M.L. Calvo, V. Lakshminarayanan, Optical waveguides: from theory to applied technologies, CRC Press, USA, 2007.
[2] H. Ma, A.K.Y. Jen, L.R. Dalton, Adv. Mater., 14 (2002), pp. 1339-1365.
[3] J. Missinne, S. Kalathimekkad, B. Van Hoe, E. Bosman, J. Vanfleteren, G. Van Steenberge, Opt. Express, 22 (2014), pp. 4168-4179.
[4] A. Elmogi, E. Bosman, J. Missinne, G. Van Steenberge, Opt. Mater., 52 (2016), pp. 26-31.
[5] Y. Xia, G.M. Whitesides, Annu. Rev. Mater. Sci., 28 (1998), pp. 153-184.
[6] T. Ishigure, K. Shitanda, Y. Oizmi, Opt. Express, 23 (2015), pp. 22262-22273.
[7] K. Makino, T. Ishigure, Y. Koike, J. Lightwave Technol., 24 (2006), p. 2108.
[8] O. Ziemann, J. Krauser, P.E. Zamzow, W. Daum, POF handbook: optical short range transmission systems, Springer Science & Business Media, USA, 2008.
[9] Y. Koike, K. Koike, J. Polym. Sci. Pol. Phys., 49 (2011), pp. 2-17.
[10] K. Soma, T. Ishigure, IEEE J. Sel. Top. Quant, 19 (2013), pp. 3600310-3600310.
[11] T. Ishigure, Y. Nitta, Opt. Express, 18 (2010), pp. 14191-14201.
[12] O. Kogan, S.R. Bickham, M.-J. Li, P. Tandon, J.S. Abbott, S.A. Garner, Design and characterization of bend-insensitive multimode fiber, Proc. 60th Internat. Wire & Cable Symp2011, pp. 154-159.
[13] A. Voigt, U. Ostrzinski, K. Pfeiffer, J.Y. Kim, V. Fakhfouri, J. Brugger, G. Gruetzner, Microelectron. Eng., 88 (2011), pp. 2174-2179.
[14] E. Bosman, J. Missinne, B. Van Hoe, G. Van Steenberge, S. Kalathimekkad, J. Van Erps, I. Milenkov, K. Panajotov, T. Van Gijseghem, P. Dubruel, H. Thienpont, P. Daele, IEEE J. Sel. Top. Quant., 17 (2011), pp. 617-628.
[15] G.D. Marshall, A. Jesacher, A. Thayil, M.J. Withford, M. Booth, Opt. Lett., 36 (2011), pp. 695-697.
[16] A. Jesacher, A. Thayil, K. Grieve, D. Débarre, T. Watanabe, T. Wilson, S. Srinivas, M. Booth, Opt. Lett., 34 (2009), pp. 3154-3156.
[17] A. Jesacher, P.S. Salter, M.J. Booth, Opt. Mater. Express, 3 (2013), pp. 1223-1232.