# Enhanced sensitivity in polymer slot waveguides by atomic layer deposited bilayer coatings

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The refractive index sensitivity of a polymer slot waveguide coated with a bilayer of $Al_2O_3/TiO_2$ was investigated theoretically and optimized for biosensor applications. The influence of atomic-layer-deposition-coated thin high-refractive-index layers on the slot confinement factor and the homogeneous sensitivity of polymer slot waveguides with different geometries were simulated. The results were compared with those of an optimized noncoated polymer slot waveguide, both operating at visible wavelengths. The simulations reveal that the proposed structure offers a significant improvement in the confinement factor and the sensitivity. These calculations present guidelines for the design and fabrication of relatively sensitive polymer slot waveguide devices for low-cost biochemical sensor applications. 

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## 1. Introduction

Growing demand for detection tools in various fields of sensing requires development of compact, low-cost, highly sensitive, and integrated devices. For biochemical sensor applications, integrated optical waveguide-based sensors are promising, and they typically rely on interrogation of the evanescent wave with an analyte, which usually functions as a cladding layer [1]. Slot waveguides [2] are structures that can strongly confine light in a low-refractive-index analyte material, with a high percentage of the field in the analyte, which can considerably increase the sensitivity. In general, high-refractive-index waveguide materials, such as silicon and silicon nitride, offer high index contrast between the waveguide and the low-refractive-index slot region, which enhances the light confinement in the slot region. For this reason, most of the demonstrated slot-waveguide-based biosensor devices are based on these materials. However, exploiting silicon slot-waveguide devices for sensing applications with water cladding is not an optimal option, since the water absorption is high at the wavelength range of silicon transparency [3]. Also, the fabrication costs of Si-based devices are low only in the case of very large device volumes. Recent studies of slot waveguides based on low-refractive-index materials have enabled the possibility of design and fabrication of low-cost slot waveguide sensors operating at the visible range of wavelengths, where the water absorption is significantly lower than at infrared wavelengths [4–6].

Typically, interaction of light with the analyte in low-refractive-index-based structures, such as polymer waveguides, is weak and a higher portion of light is confined inside the polymeric structure. The reported confinement factor in the slot region at visible wavelengths in a polymer slot waveguide in water is around 7%, which leads to a relatively low device sensitivity [5]. The development of polymer slot waveguides for biochemical sensor applications has encouraged further studies to improve the slot

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confinement factor and sensitivity with these structures. So far, utilizing high-refractive-index materials, such as $TiO_2$ and $Ta_2O_5$, on inverted strip polymer waveguides in order to increase the evanescent wave in the upper cladding has been demonstrated for sensing applications [7,8]. However, with this approach, the overlap of the optical mode with the cladding is increased to about 10%, which is slightly higher than the confinement factor in a purely polymeric slot waveguide.

Atomic layer deposition (ALD) is a technique that offers growth of thin films in such a way that the film thickness can be controlled at an atomic level. Additionally, ALD growth is conformal and the thin films are formed uniformly, even on the vertical sides of nanostructures [9]. Therefore, ALD can provide low-temperature-grown conformal thin layers that are compatible with polymer structures and the method is ideally suited for film deposition in narrow slots [10]. It is indeed a proven deposition method (so far the only one) that has been used to uniformly fill nanoscale slot waveguides with oxide materials [11]. Furthermore, the ALD-grown films are virtually pinhole free, and they are often used as barrier layers to prevent diffusion of different materials. Measurements of water vapor diffusion in polymers have shown that, with film thicknesses greater than only 10 nm, these films have barrier properties similar to those of glass [12].

Based on the above considerations, in this paper we theoretically investigate and propose a polymer slot waveguide configuration with a bilayer of ALD-grown $Al_2O_3/TiO_2$ for biochemical sensor applications. Note that we chose a bilayer instead of just a single high-index layer. Based on our initial calculations, the bilayer of $Al_2O_3/TiO_2$ provides better confinement in the slot and it can easily be fabricated with ALD. In addition, ALD-grown bilayers are virtually pinhole free, minimizing the problem of water diffusion in polymers. The structure is designed for operation at a wavelength of 633 nm. The slot waveguide structure is optimized for homogeneous sensing by taking into account the fabrication limitations. The homogeneous sensitivity of the proposed structure is compared with that of an optimized noncoated polymer slot waveguide.

## 2. Polymer Slot Waveguide with a Bilayer of $Al_2O_3/TiO_2$ ALD-Grown Films

A cross section of the polymer slot waveguide with a bilayer of $Al_2O_3/TiO_2$ thin films is illustrated schematically in Fig. 1. We assume Ormocore polymeric rails on a Si wafer separated by a 3 μm $SiO_2$ layer. Taking into account the fabrication of the structure by UV nanoimprinting and the features of this method, a fixed residual layer of $R = 150$ nm is assumed in the slot region between the polymer rails. This is typical in polymeric structures fabricated by the nanoimprinting technique. For adopting $Al_2O_3/TiO_2$ layers in both sides of the polymer rails, it is assumed that the conformal layers of $Al_2O_3/TiO_2$ are coated by the ALD technique, and, subsequently, layers on top of the rails and in the slot region are removed by reactive ion etching. The reason for selecting these materials is their high suitability for deposition by the ALD technique. In addition, these materials have an excellent biocompatibility and studies have shown that a bilayer of $Al_2O_3/TiO_2$ enables a virtually pinhole-free layer with high impermeability against diffusion of water molecules [12]. The dimensions of the polymer rails and the thickness of the $Al_2O_3$ and $TiO_2$ layers on both sides of the polymer rails are indicated in Fig. 1. The width of the slot $g$ is defined after introducing the bilayer of the inorganic materials.

The key optical properties of the structure, i.e., the field distribution and the effective refractive index of the guided mode, were calculated using the Fourier modal method [13,14]. The refractive indices used in the simulations were 1.553 for polymer (Ormocore) and 1.457 for $SiO_2$. With the purpose of having amorphous layers of inorganic materials, and based on our own experiments, we assume ALD at low temperatures of 200°C and 120°C for $Al_2O_3$ and $TiO_2$, respectively. The corresponding refractive indices are 1.64 and 2.1. An aqueous buffer solution with refractive index of 1.33 is adopted as the slot filling and the top cladding material, which is indicated as a cover medium in these calculations.

First, the confinement factor only in the slot (S) and in the whole cover (C) medium was calculated as a function of $g$ and $w$ with the fixed parameters as $h = 400$ nm, $ha = 25$ nm, and $hb = 90$ nm and by defining it as a fraction of the optical mode in the area of calculation $i$:

$$\Gamma_i = \frac{\iint_i |\mathrm{Re}\{(E \times H).u_z\}| \mathrm{d}x\mathrm{d}y}{\iint_{\mathrm{total}} |\mathrm{Re}\{(E \times H).u_z\}| \mathrm{d}x\mathrm{d}y}, \qquad i = \mathrm{S}, \mathrm{C}, \quad (1)$$

where $E$ and $H$ are the electric and magnetic fields, respectively [15].

In a vertical slot waveguide, the field enhancement in the slot region occurs for the quasi-TE mode, and the lateral component of the electric field is the major component in the slot region. Figure 2 illustrates the

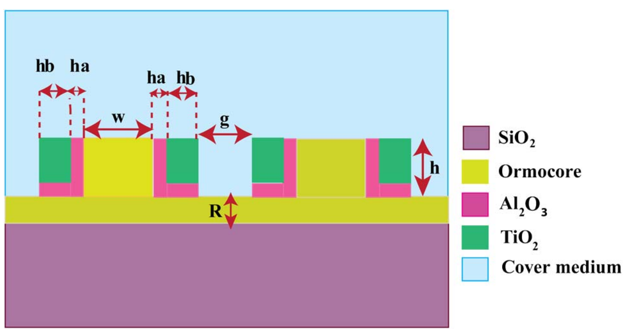


Fig. 1. Schematic view of the cross section of a polymer slot waveguide with a bilayer of $Al_2O_3/TiO_2$ thin films. The polymer rails have a height $h$ and a width $w$. The thickness of the $Al_2O_3$ and $TiO_2$ layers are $ha$ and $hb$, respectively.

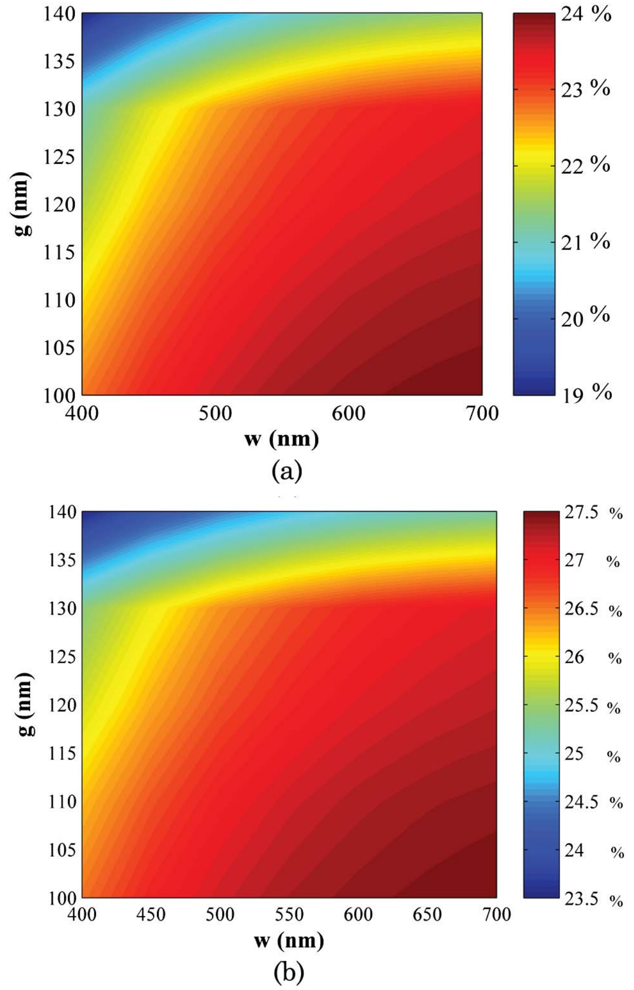


Fig. 2. Variation, with $g$ and $w$, of the confinement factor in (a) the slot and (b) the whole cover medium of the polymer slot waveguide with a bilayer of $Al_2O_3/TiO_2$ with $h = 400$ nm, $ha = 25$ nm, and $hb = 90$ nm.

effect of varying $w$ and $g$ on the confinement factor in the slot, $\Gamma_s$, and in the whole cover medium,$\Gamma_c$;. According to previous studies on modeling and fabrication of polymer slot waveguides, the height of the polymer rails needs to be adequate to achieve a large confinement factor in the slot region [4,5]. However, fabrication of structures with a high aspect ratio is always challenging and easily brings about slanted rails and other fabrication errors. Here we considered the slot width $g \geq 100$ nm in our calculations. In wider gaps the capillary effect is smaller and surface tension is lower [1]. This is an advantage when the slots need to be fully filled with liquid solutions, which is preferred in aqueous sensing and optofluidic applications. The confinement factor calculations reveal that adopting the thin layers of $Al_2O_3/TiO_2$ results in a feasible fabrication process and a much increased confinement factor, in comparison with a noncoated single polymer slot waveguide. As seen from Fig. 2, the confinement factors in the slot and the whole cover region increase with increasing $w$, and changing the slot width $g$ from 100 nm to larger values decreases monotonically both confinement factors.

In order to optimize the waveguide dimension, the slot and rail widths are selected according to the final application and fabrication limits. Our simulation results display that increasing the rail width more than $w = 680$ nm has only a slight influence on the confinement factor, yet it increases the possibility of the appearance of the second quasi-TE mode and extends the final device footprint. Presence of higher order modes due to corresponding low effective indices increases the scattering losses, which is undesirable for sensor applications. With values of $w = 600$ nm and $g = 120$ nm the confinement factor in the slot is 23% and in the whole cover medium 26.5%. These dimensions offer a reasonable aspect ratio to facilitate the fabrication of polymer-based waveguide devices with low-cost replication processes and maintain the advantage of a wider slot width for aqueous sensing. With these values, the waveguide supports only the fundamental quasi-TE mode in the slot, which is tightly confined. The presence of the high-index thin-film layers decreases the portion of light in the polymer rails and "pushes" light to the slot region.

Figures 3(a) and 3(b) depict the mode profile in the polymer slot waveguide with a bilayer of $Al_2O_3/TiO_2$ and show that the optical field strongly lies within

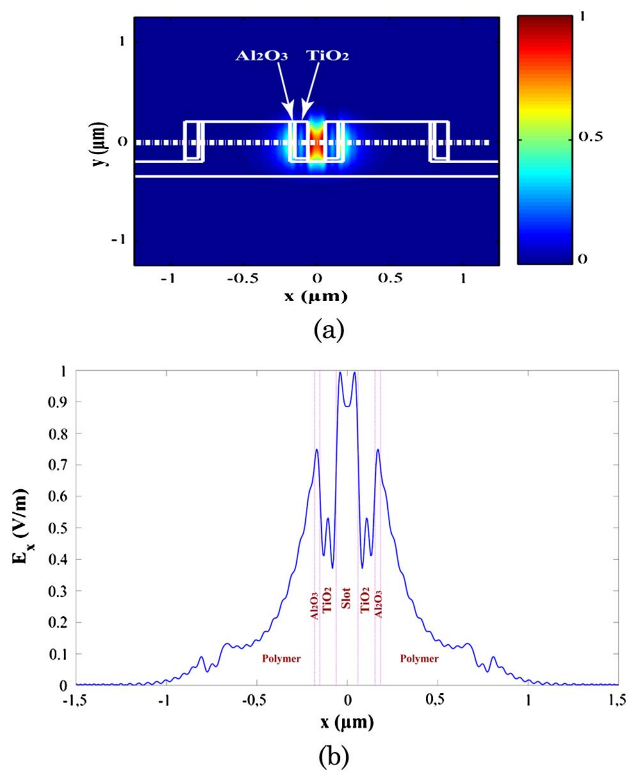


Fig. 3. (a) Lateral component of the electric field of the quasi-TE mode in the slot waveguide and (b) distribution of the field along the dashed line indicated in (a); the vertical lines indicate the positions of the interfaces. The slot waveguide parameters are $h = 400$ nm, $w = 600$ nm, $g = 120$ nm, $ha = 25$ nm, and $hb = 90$ nm.

the slot. However, as can be seen from Fig. 3(a), a portion of the field is in the polymer rails. Thus they are a part of the slot waveguide structure and do not provide only mechanical support to ALD-grown films.

We also investigated the confinement factors for a noncoated polymer slot waveguide with optimized dimensions of $h = 750$ nm and $w = 450$ nm [5] and with different values of the slot gap and compared the values with the proposed structure. The results shown in Fig. 4 clearly illustrate that the variation of the confinement factor with an increasing slot width in a noncoated polymer slot waveguide is comparatively flat. Increasing the gap to 130 nm results in the largest confinement in the slot.

In the polymer slot waveguide with thin layers of $Al_2O_3/TiO_2$, increasing the gap width slightly decreases the slot confinement. Nevertheless, with a fixed value of $g = 120$ nm, the confinement factor in the slot region for the noncoated polymer slot waveguide is near 7%, and it is more than 23% in the proposed structure, demonstrating an improvement of over 3 times in the confinement factor in both the slot and the whole cover medium. These dimensions imply a much easier nanoimprinting process in comparison with the corresponding dimensions of $h = 750$ nm and $w = 450$ nm for the optimized noncoated polymer slot waveguide. The results confirm that the presence of these layers "pushes" light to the slot gap and enhances the light confinement in the slot region. It is clear that the increase of the portion of light in the slot gap results in higher sensitivity in slot-waveguide-based biochemical sensors.

## 3. Sensitivity Investigations

Most waveguide-based sensors can provide label-free sensing based on the change in the effective refractive index of the guided mode. This change can be due to the change in the refractive index of the cover medium or the change in either the refractive index or the thickness of a thin layer of biomolecules distributed on the surface of the waveguide, which is due to the binding of the target molecules. In homogeneous sensing the device is usually immersed in a buffer solution that acts as the cladding of the waveguide. The change of its refractive index due to changes in concentration or the amount of the solution is then measured. The sensitivity is defined as $S_h = (\partial n_{\mathrm{eff}}/\partial_{n_c})$, where $n_{\mathrm{eff}}$ is the waveguide effective index and $n_c$ is the refractive index of the cover medium [16]. Following immersion of the device in an aqueous solution, the effective index changes slightly due to the change of the cladding from air to aqueous solution (i.e., water), and this affects the modal behavior of the structure. In polymeric structures, due to the molecular properties of polymers, the diffusion of water molecules causes a significant delay in reaching the saturation state and extends the response time of the device. Introducing an inorganic layer on the polymer structure can block the diffusion of water molecules and shorten the device response time [17]. It is expected that in the proposed device the presence of thin layers of $Al_2O_3/TiO_2$ will result in the advantage of a fast response time because of the impermeability against water molecules.

Here we investigate only the homogeneous sensitivity of the proposed waveguide structure. As defined in the homogeneous sensitivity, $S_h$, calculation equation, the numerical sensitivity can be calculated by a small perturbation of the cover medium and finding the relevant effective refractive index shift. Therefore, in order to calculate the dependence of the homogeneous sensitivity on the dimensions of the waveguide, we first change the refractive index of the cover medium slightly from 1.33 to 1.335 and investigate the homogenous sensitivity $S_h$ as a function of both the rail width $w$ and the slot width $g$. As expected and seen in Fig. 5, the homogeneous sensitivity of the proposed waveguide structure has behavior similar to that of the confinement factors.

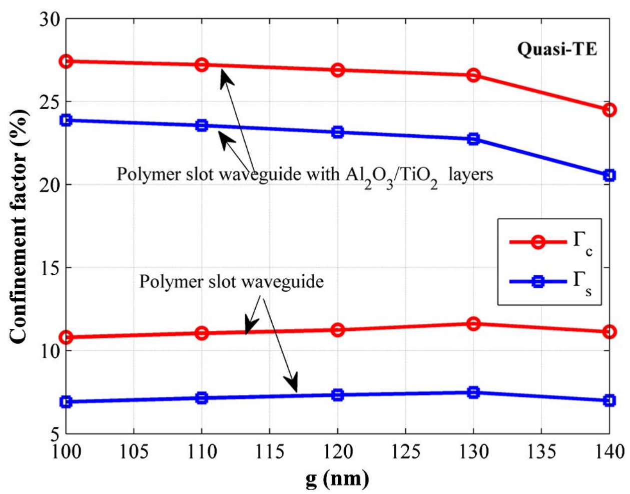


Fig. 4. Comparison of the confinement factors versus $g$ in a noncoated polymer slot waveguide with $h = 750$ nm and $w = 450$ nm and a polymer slot waveguide with inorganic layers $h = 400$ nm, $w = 600$ nm, $ha = 25$ nm, and $hb = 90$ nm.

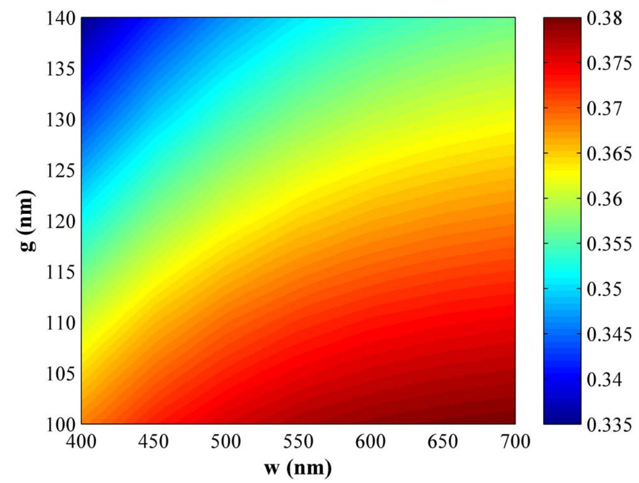


Fig. 5. Dependence of the homogeneous sensitivity on $w$ and $g$ in a polymer slot waveguide with bilayers of $Al_2O_3/TiO_2$ with $h = 400$ nm, $ha = 25$ nm, and $hb = 90$ nm.

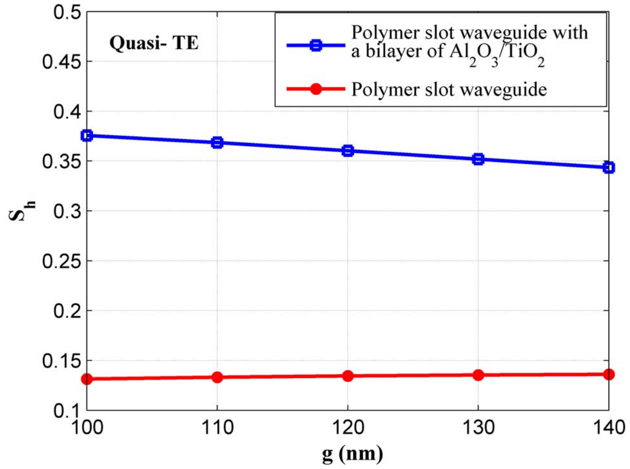


Fig. 6. Homogeneous sensitivity $S_h$ versus $g$ in an optimized noncoated polymer slot waveguide with $h = 750$ nm and $w = 450$ nm and in a polymer slot waveguide with layers of $Al_2O_3/TiO_2$ with $h = 400$ nm, $w = 600$ nm, $ha = 25$ nm, and $hb = 90$ nm.

With $g = 120$ nm, increasing $w$ from 600 to 700 nm increases the sensitivity by 30%, but it adds another quasi-TE mode in the slot.

In Fig. 6, we plot the variation of the homogeneous sensitivity in this structure and in a noncoated polymer slot waveguide together.

This comparison reveals that the homogeneous sensitivity of the proposed structure increases by a factor of 3 in comparison with the noncoated polymer slot waveguides. A waveguide structure with dimensions of $h = 400$ nm, $w = 600$ nm, and $g = 120$ nm supports only the fundamental quasi-TE mode in the slot and gives a homogeneous sensitivity of $S_h = 0.36$, which is, according to our calculations, comparable with that of high-refractive-index slot waveguides, such as a $TiO_2$ slot waveguide. We also studied the influence of the changes in the thickness of the dielectric layers on the confinement factor in the slot region and on the homogeneous sensitivity of the structure with these optimized values. Figure 7 depicts the homogeneous sensitivity of the structure versus layer thickness. It can be seen that increasing $hb$ improves the sensitivity. However, our calculations showed that increasing $hb$ to 100 nm adds another quasi-TE mode to the structure. In order to keep the single-mode behavior of the waveguide structure, we fixed the layer thicknesses as $ha = 25$ nm and $hb = 90$ nm. With these parameters the calculated homogeneous sensitivity of the proposed slot waveguide structure is 0.36. We studied the influence of $h$ on waveguide sensitivity. Our calculations showed that, in the single-mode slot waveguide structure with $w = 600$ nm, $g = 120$ nm, $ha = 25$ nm, and $hb = 90$ nm, the maximum sensitivity, which is 0.37, can be achieved with $h = 420$ nm.

The proposed structure that employs thin layers of $Al_2O_3/TiO_2$ increases the sensitivity of a polymer slot waveguide. However, in practice, there is a need to use dry etching to remove the ALD-grown inorganic layers from the top of the polymer rails, which increases surface roughness and scattering loss in the structure and decreases the detection limit. Previous studies on application of the ALD technique on Si-based slot and rib waveguides have shown that introducing a thin layer of ALD-grown

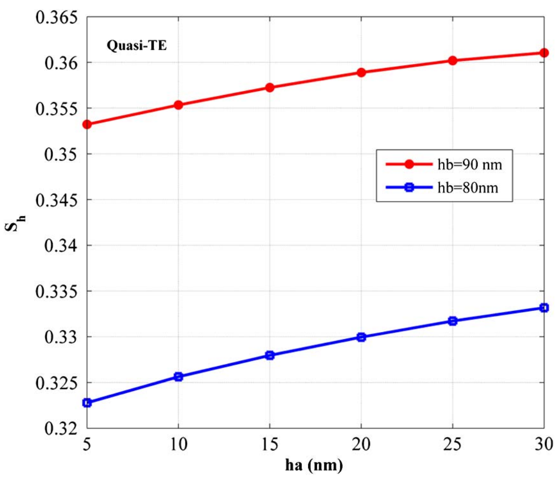


Fig. 7. Homogeneous sensitivity versus $ha$ and $hb$ with $g = 120$ nm, $h = 400$ nm, and $w = 600$ nm.

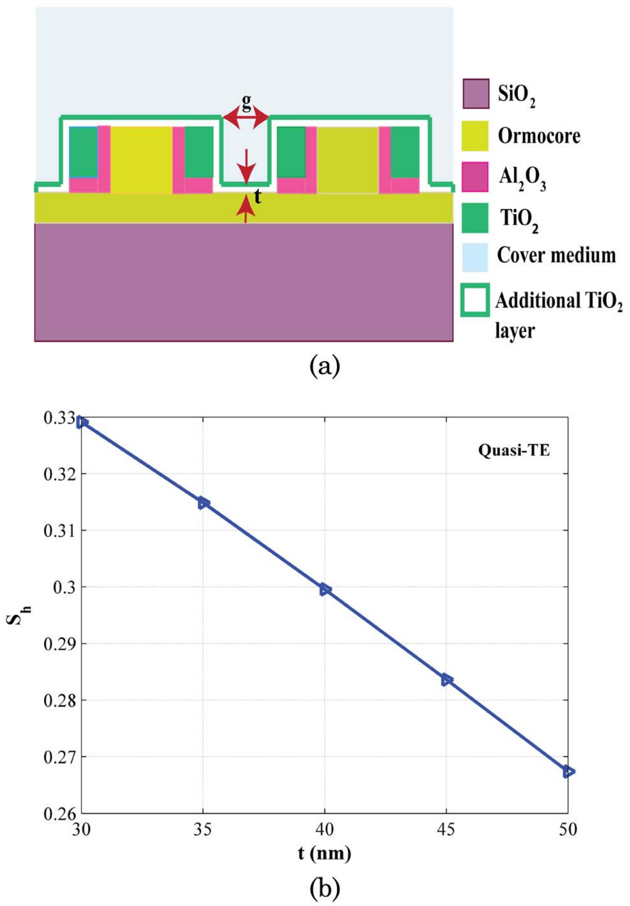


Fig. 8. (a) Schematic view of the cross section of the slot waveguide with nonuniform layers and an additional layer of $TiO_2$ with a thickness of $t$ and (b) its homogeneous sensitivity as a function of $t$.

films reduces scattering loss [11]. We believe that adding an additional thin layer of $TiO_2$ can reduce the effect of surface roughness to some extent in the proposed structure. For this purpose, we investigated the effect of applying a thin layer of $TiO_2$ on the homogeneous sensitivity of the proposed structure [see Fig. 8(a)]. We varied the thickness $t$ of this layer from 30 to 50 nm and calculated its impact on waveguide sensitivity.

The sensitivity of the structure shown in Fig. 8(a) decreases slightly from 0.36 to 0.3 by introducing a thin layer of 40 nm due to "pulling" a small portion of the modal light into $TiO_2$ layers. Nevertheless, this additional layer can result in a meaningful improvement in the surface roughness and improve the detection limit. There is a trade-off between the sensitivity and the detection limit. Furthermore, this additional hydrophobic layer is able to block the water diffusion also in the uncovered portion of the polymeric structure and reduce the device response time.

## 4. Conclusion

We theoretically investigated in detail the homogeneous sensitivity and the confinement factor in a polymer slot waveguide coated with a bilayer of $Al_2O_3/TiO_2$ operating at 633 nm. Calculations showed that employing a thin layer of these inorganic materials can strongly confine light in the slot region and thus improve the confinement factor. Consequently, the homogeneous sensitivity of the proposed structure is more than 3 times larger than that of a noncoated polymer slot waveguide. In addition, the low aspect ratio of the polymer waveguide structure relaxes the fabrication requirements of the structure, which facilitates its fabrication with low-cost nanoimprinting lithography. The nearly pinhole-free layers of $Al_2O_3/TiO_2$ can improve the water diffusion problem in polymeric structures and the response time of the device is expected to shorten significantly. Introducing an additional thin layer of $TiO_2$ is expected to reduce the surface roughness that results from the dry etching process in this structure and to have a negligible influence on the sensitivity of the structure. It can also act as an additional diffusion barrier for water molecules. The optimized geometry provides the highest sensitivity in polymer slot waveguides and the additional processing steps needed are simple and compatible with polymers. The proposed device has potential for fabrication of low-cost waveguide biosensor chips for applications in aqueous sensor configurations. Our future goal is to demonstrate the proposed waveguide structure in an interferometric configuration and operating as a biosensor. In addition to the interferometer, the device will include integrated input/output couplers and strip-to-slot waveguide couplers.

## References


1. C. A. Barrios, "Optical slot-waveguide based biochemical sensors," Sensors **9**, 4751–4765 (2009).
2. V. R. Almeida, Q. Xu, C. A. Barrios, and M. Lipson, "Guiding and confining light in void nanostructure," Opt. Lett. **29**, 1209–1211 (2004).
3. R. G. Eldridge, "Water vapor absorption of visible and near-infrared radiation," Appl. Opt. **6**, 709–713 (1967).
4. P. Bettotti, A. Pitanti, E. Rigo, F. Leonardis, V. Passaro, and L. Pavesi, "Modeling of slot waveguide sensors based on polymeric materials," Sensors **11**, 7327–7340 (2011).
5. M. Hiltunen, J. Hiltunen, P. Stenberg, E. Heinonen, P. Vahimaa, and P. Karioja, "Polymeric slot waveguide at visible wavelength," Opt. Lett. **37**, 4449–4451 (2012).
6. G. Testa and R. Bernini, "Slot and layer-slot waveguide in the visible spectrum," J. Lightwave Technol. **29**, 2979–2984 (2011).
7. S. W. Kwon, W. S. Yang, H. M. Lee, W. K. Kim, G. S. Son, D. H. Yoon, S. D. Lee, and H. Y. Lee, "The fabrication of polymer-based evanescent optical waveguide for biosensing," Appl. Surf. Sci. **255**, 5466–5470 (2009).
8. J. Hiltunen, S. Uusitalo, P. Karioja, S. Pearce, M. Charlton, M. Wang, J. Puustinen, and J. Lappalainen, "Manipulation of optical field distribution in layered composite polymeric inorganic waveguides," Appl. Phys. Lett. **98**, 111113 (2011).
9. K. Solehmainen, T. Aalto, J. Dekker, M. Kapulainen, M. Harjanne, K. Kukli, P. Heimala, K. Kolari, and M. Leskelä, "Dry-etched silicon-on-insulator waveguides with low propagation and fiber-coupling losses," J. Lightwave Technol. **23**, 3875–3880 (2005).
10. T. Alasaarela, T. Saastamoinen, J. Hiltunen, A. Säynätjoki, A. Tervonen, P. Stenberg, M. Kuittinen, and S. Honkanen, "Atomic layer deposited titanium dioxide and its application in resonant waveguide grating," Appl. Opt. **49**, 4321–4325 (2010).
11. T. Alasaarela, D. Korn, L. Alloatti, A. Säynätjoki, A. Tervonen, R. Palmer, J. Leuthold, W. Freude, and S. Honkanen, "Reduced propagation loss in silicon strip and slot waveguides coated by atomic layer deposition," Opt. Express **19**, 11529–11538 (2011).
12. T. Kwon, D. Moon, Y. Moon, W. Kim, and J. Park, "Al2O3/TiO2 multilayer passivation layers grown at low temperature for flexible organic devices," J. Nanosci. Nanotechnol. **12**, 3696–3700 (2012).
13. C. Zhou and L. Li, "Formulation of the Fourier modal method for symmetric crossed gratings in symmetric mountings," J. Opt. A **6**, 43–50 (2004).
14. J. Hugonin and P. Lalanne, "Perfectly matched layers as nonlinear coordinate transforms: a generalized formalization," J. Opt. Soc. Am. A **22**, 1844–1849 (2005).
15. N. N. Feng, J. Michel, and L. C. Kimerling, "Optical field concentration in low-index waveguides," IEEE J. Quantum Electron. **42**, 885–890 (2006).
16. K. Tiefenthaler and W. Lukosz, "Sensitivity of grating couplers as integrated-optical chemical sensors," J. Opt. Soc. Am. B **6**, 209–220 (1989).
17. M. Wang, S. Uusitalo, L. Hakalahti, C. Liedert, R. Myllylä, and J. Hiltunen, "Polymeric dual-slab waveguide interferometer for biochemical sensing applications," Appl. Opt. **51**, 1886–1893 (2012).