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Optical properties of thin films monitored in real-time at high gamma radiation doses using long period fiber gratings

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ABSTRACT

In this work, we report an all-optical-fiber-based method to monitor in real-time the changes in thin film optical properties when exposed to harsh environments such as high doses of gamma radiation. The method is based on a long period grating (LPG) inscribed in a radiation resistant optical fiber, which is coated with a thin film to be analyzed. Due to impact of the environment on the film properties, the transmission spectrum of the LPG changes. To validate this methodology, we deposited oxides of various metals, i.e. aluminum (Al_2O_3), titanium (TiO_2), and tantalum (Ta_2O_5), as well as a polymeric film, namely polystyrene (PS), on the LPG surface and measured the resonant wavelength shift induced by high doses of gamma exposure (dose rate of 2.3 kGy/h, up to a total dose of 46 kGy). The changes could be observed in real-time up to maximum reached dose and during the recovery period. The results are significantly dependent on thin film material. Among the oxides, TiO_2 thin film demonstrated the highest susceptibility to irradiation, whereas Al_2O_3 exhibited the least impact. Additionally, a good recovery potential for Ta_2O_5 thin film was observed, hinting at its promising application in dosimetry. Conversely, the PS film exhibited a permanent effect induced by irradiation. This is the first real-time experimental study of the impact of high dose gamma radiation on thin film optical properties using a universal optical-fiber-based device. The findings pave the way for assessing radiation hardness of thin film materials or utilizing such a method to optimize material treatment involving ionizing radiation.

1. Introduction

Thin films are commonly used elements of almost every technical device manufactured nowadays. Depending on properties, they may offer different functionalities, such as electrical contact, barrier or isolation in electronic elements, electromagnetic field distribution in photonic devices, or simply enhanced mechanical or chemical protection in a large variety of applications. For example, thin films are often used in critical components of aerospace and nuclear systems, such as coatings on spacecraft surfaces, or protective layers in nuclear reactors. In both aerospace and nuclear applications, thin films are used as radiation shielding materials to protect sensitive electronic equipment, instruments, and personnel from harmful radiation exposure. However, the properties of the films, as well as for bulk materials, may change in time, mostly due to environmental conditions. While the change is typically not crucial for bulk material performance, it may be of high importance for devices based on thin films [1–5]. The ability to change with certain parameters is a key property in sensing applications, but in all the others, the change needs to be avoided, limited or at least predicted. This is the case when the films are exposed to ionizing radiation during, e.g., space missions, high-energy physics experiments, or operating in nuclear facilities. Interaction between the radiation which includes alpha, beta, gamma, and X-rays, as well as electrons, protons, and neutrons may cause changes in properties of materials, also in their thin film form, and thus may have a curtail impact on performance of the devices based on them during their lifetime [6]. Among key properties of

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the films, not only in photonic applications, are those optical that can be investigated using non-destructive methods. Such optical parameters as refractive index (n) and extinction coefficient (k) on top of the film thickness determine transmission/reflection properties of the materials. Moreover, the parameters correspond to electrical permittivity and density, which are critical for electrical and mechanical performance of devices based on the films, respectively [7]. Among the thin film materials, metal oxides are very often applied. They can be obtained with different deposition methods and tailored in a wide range of properties at the stage of their deposition or post-processing [8]. The other group of materials widely exposed to high-energy particles in many applications are organic polymers. They increasingly find applications in memory devices, and other optical, as well as electronic applications [9,10].

To investigate optical properties of thin films typically spectroscopic ellipsometers or interferometers are used, but the measurement setups contains both electronic and precise optical elements designed for operation in well-defined laboratory conditions and are hardly transferrable, to the harsh environmental conditions, such as nuclear reactors. Thus, the effect of the radiation of the film properties is typically measured post-factum, what highly limits the information about the impact of radiation. There can be found a few studies reporting effects of irradiation of optical coatings with ionizing radiation, especially at its high dose [11–15]. According to our best knowledge up to date there has been reported very few experimental setups making it possible to analyze optical properties of thin films in real time [16] whereas most of the works are focusing on off-line measurements [17–22]. In [16] the real time measurements are based on a photonic integrated circuit coated with thin films to be investigated, supported by lead-in and lead-out optical fibers for the readout. As shown by this measurement concept, even if thin films are analyzed using a planar device, optical fibers are essential for obtaining the information externally in real time.

Optical fibers and sensors based on them are often employed in environments exposed to harsh conditions, due to their small size and weight, high multiplexing capability, long distance monitoring and resistance to corrosion and electromagnetic interferences. Several studies focused on the assessment of the behavior of optical fibers when exposed to different sources of radiations with several characteristics, highlighting the possibility to have radiation resistant fibers, as well as radiation sensitive models permitting the development of dosimeters [23–25]. Some optical fiber sensors are suitable for the real-time monitoring of thin film properties when exposed to radiations [26–28]. Among them are long period gratings (LPG) obtained as a periodic modulation of n and/or geometry of a piece of optical fiber. The period of the modulation ranges within hundreds of micrometers and produces a resonant coupling between the core mode and co-propagating cladding modes. The transmitted spectrum of LPG shows a series of attenuation bands, each one associated to the coupling with a different cladding mode. The spectral position of the bands is mainly dependent upon the grating period and the effective refractive indices of core and cladding modes [29]. Consequently, the device is highly sensitive to changes occurring both in fiber properties and surrounding medium. In this context, LPG has been proven as a valuable tool to probe the changes occurring in the optical properties (n and thickness) of the thin film deposited around the grating region [30,31]. Recently, LPGs have drawn attention for their application in environments exposed to radiations [32]. The investigations focused on the measurement of LPG response under different sources of radiation (i.e., gamma, neutrons, and protons) when fabricated in commercially available optical fibers of different compositions. The results highlighted a strong dependence of the LPG wavelength shift upon the fiber type, highlighting a significant sensitivity in some cases (e.g., fibers doped with Ge, B, P, etc.) and resistance in others (pure silica core and F-doped) [33–36]. Moreover, a few reports focused on the investigation of LPG coated with nano-sized materials for the measurement of humidity in radiation exposed environments [37–39]. Therefore, the LPG has been demonstrated so far to be a valuable platform for the real-time monitoring of optical fiber properties, also in harsh conditions as those when the radiation occurs.

This paper presents, for the first time, experimental studies focusing on the real-time monitoring of the effects of gamma radiation on the optical properties of thin films deposited on LPGs. The gratings have been fabricated in radiation-resistant pure silica core fiber and were coated with different thin film materials including a selected polymeric film, i.e. polystyrene (PS), and three different metal oxides namely, aluminum (Al_2O_3), titanium (TiO_2), and tantalum (Ta_2O_5) oxides, which are expected to be considered for devices working in radiation exposed environments. Evolution of the films' optical properties has been observed during gamma irradiation, as well as at the stage of recovery. For some of the materials irradiation has been repeated to identify their possible multiple application in high-dose radiation environments.

2. Materials and methods

In this section, the fabrication procedure of LPGs based on electric arc discharge method is first described (Section 2.1), subsequently the attention is focused on the thin film deposition methods and their analysis (Section 2.2), finally the gamma irradiation experimental setup is illustrated (Section 2.3).

2.1. LPG fabrication and measurement

For the fabrication of the LPGs a pure-silica core fiber was selected. The Nufern (USA) S1310 fiber is designed for harsh environments and manufactured to be radiation resistant. It has the following properties: pure-silica core with a diameter of 8 μm, F-doped inner cladding with diameter of about 95 μm, pure-silica outer cladding with a diameter of 125 μm, numerical aperture NA = 0.12 and mode field diameter MFD = 10.4 ± 0.8 μm at λ = 1550 nm. Next, an electric arc discharge approach was selected for the LPG writing as detailed in our previous work [40], since it permits the fabrication of grating in any kind of fiber including non-photosensitive ones with greate flexibility. In our technique, the discharge is periodically repeated and combined with a mechanical action, such as pulling the fiber by constant axial tension, to ensure a repeatable process. In the specific case, a periodic perturbation is achieved by subjecting the optical fiber under test to the arc discharges provided by a Sumitomo Type-39 (Japan) fusion splicer, with typical arc discharge parameters: power in the range of 10–20 step (proprietary unit), arc duration of 500–750 ms and electrode gap of about 0.8 mm, resulting in devices with final length up to 30 mm. During the procedure, one end of the fiber was fixed onto a precision translation stage while the other end was kept under constant axial tension by means of a 12 g weight. The desired spectral features of the LPG are obtained after the application of a sequence of arc discharges to the uncoated region of the fiber, with the discharges being alternated with fiber displacement by the grating period (Λ). For our purpose, the grating period was designed to have the resonance corresponding to the 6th order of cladding modes in the working wavelength range (1510–1590 nm), i.e., Λ = 430–440 μm.

The LPG spectra were acquired using a broadband source as input light and an optical spectrum analyzer as detector (Yokogawa AQ6370B, Japan), during the fabrication and deposition phases. For the work, we focused on the wavelength range 1500–1600 nm. Whereas the opto-electronic acquisition system employed during the irradiations consisted of a sm125 interrogator (Micron Optics, USA), operating in the wavelength range 1510–1590 nm, with a resolution of 1 pm and acquisition time set to 30 s. In particular, the wavelength shift of the LPGs as well as the raw spectral data were acquired in real-time during the exposure to gamma radiation.

2.2. Thin film deposition and analysis

The PS layer was deposited on the LPG surface by dip coating technique, starting from a 10 % PS solution in chloroform and using a

withdrawal speed of 30 mm/min. Metal oxides, i.e. Al_2O_3, TiO_2 and Ta_2O_5 films were in turn deposited using atomic layer deposition (ALD) method using Veeco Savannah S100 system, making possible to achieve great uniformity of the coating around the fiber [41]. The precursors used for the deposition were deionized water in tandem with trimethylaluminum (TMA), titanium tetrachloride ($TiCl_4$) and pentakis (dimethylamino)tantalum(V) (PDMAT) in the case of Al_2O_3, TiO_2 and Ta_2O_5 depositions, respectively. The precursors were working at room temperature, except PDMAT that was heated up to 105 °C. All the depositions were conducted at 80 °C. The depositions of Al_2O_3, TiO_2 and Ta_2O_5 were interrupted after 1211, 627 and 440 cycles, respectively. The number of cycles has been adjusted for each of the material to obtain fixed 10 nm shift in the resonance wavelength of the LPG.

The optical properties of thin films, i.e., n and k, as well as their thickness, were assessed before and after the irradiation on the reference silicon wafers coated in the same processes as LPGs using HORIBA Jobin Yvon UVISEL spectroscopic ellipsometer.

2.3. Gamma irradiation setup

The setup for the gamma irradiation is described in detail in our previous works [36,42]. Briefly, we used a compact irradiator, model GC-5000 (BRIT, India), where the sensors are exposed to gamma radiation produced by ^{60}Co rods. The LPGs were arranged onto a custom metallic holder to keep their strain state fixed during the experiment. A total dose of about 46 kGy was reached during the experiment, with an average dose rate of 2.3 kGy/h for the total of 20 h. Subsequently the recovery effects were observed in the next 5 h, i.e. long enough to observe any permanent change.

3. Results and discussion

3.1. Thin-film deposition

For this experiment, the LPGs were induced in pure-silica core fiber exhibiting the attenuation band corresponding to 6th order cladding mode in the wavelength range 1580–1600 nm. Such LPGs were subsequently coated with different thin films. The pristine and coated spectra of the devices are comparatively plotted in Fig. 1(a), 1(b), 1(c), 1(d), respectively for PS, TiO_2, Ta_2O_5 and Al_2O_3. For each case a blue shift of the resonance wavelength can be seen, which is in agreement with theoretical behavior and proves the effectiveness of the deposition [41]. In all the cases, due to higher n of the thin film materials than the one of the fiber claddings, effective refractive index of the cladding mode increases bringing the resonance towards shorter wavelengths [30]. Coating by PS induced the highest shift reaching about 35 nm, as

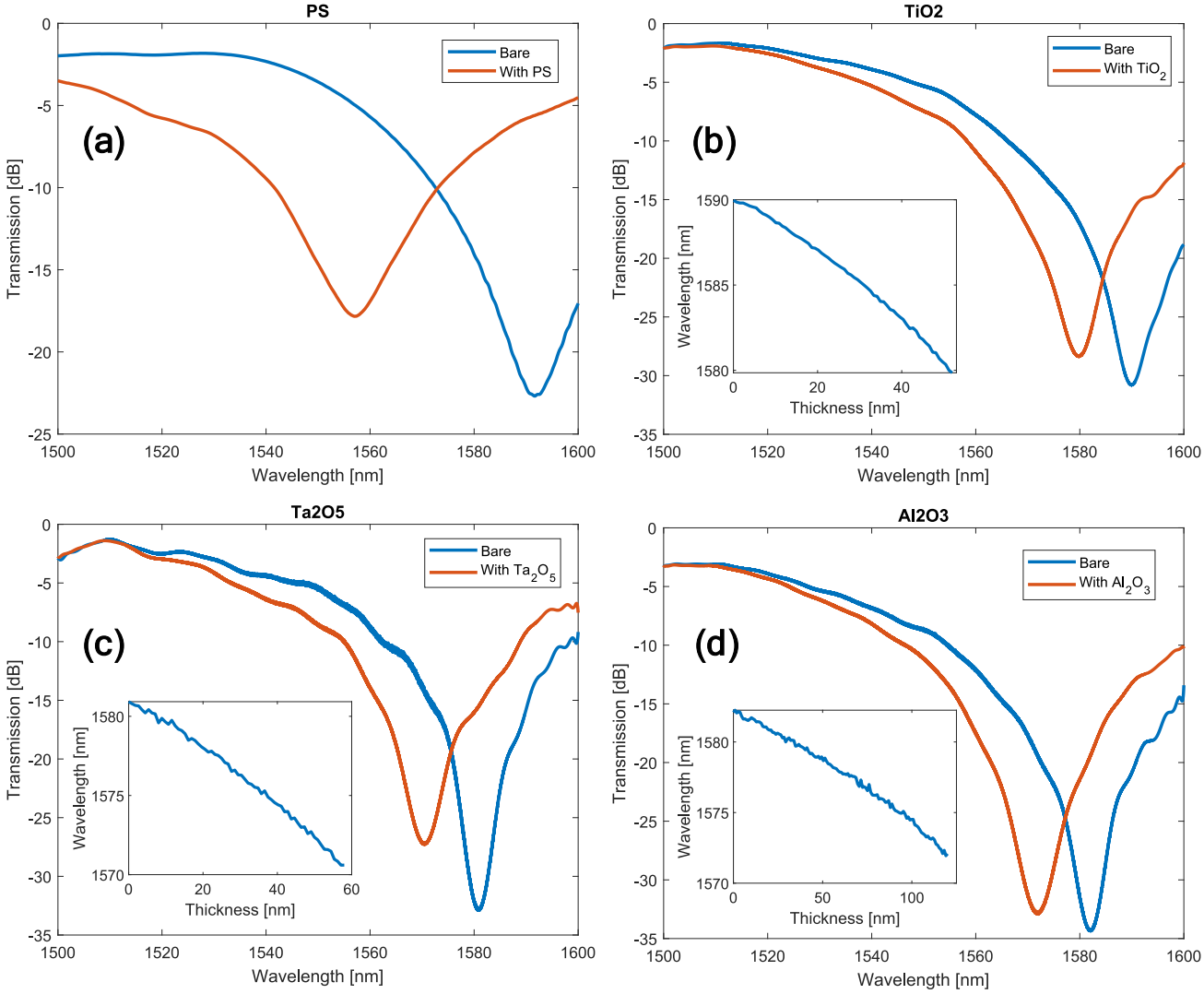

Fig. 1. Effect of different thin film deposition on the spectra of LPGs in the case of: (a) PS, (b) TiO_2, (c) Ta_2O_5, and (d) Al_2O_3. The insets report the evolution of the resonance wavelength with film thickness when monitored during ALD process.

reported in Fig. 1(a). Using a numerical design tool [43] and $n = 1.58$, we estimated PS coating thickness to 270 ± 20 nm. During the deposition of TiO_2, Ta_2O_5, and Al_2O_3, the evolution of the resonance wavelength was monitored in real-time and reported as inset in the corresponding subfigures in Fig. 1. For each metal oxide, the deposition was stopped each time as the shift reached 10 nm. According to measurements of reference samples a thickness of 120 nm, 52 nm and 58 nm was reached for Al_2O_3, TiO_2 and Ta_2O_5 depositions, respectively. The difference in thicknesses of the metal oxides for fixed resonance wavelength shift originates from n of the thin film materials [44]. Among the used oxides the one of Al_2O_3 is the lowest and for TiO_2 is the highest. Thus, to induce the same shift it is required thicker film for the material showing lower n.

3.2. Gamma irradiation

Next, the coated LPGs and additionally bare one as a reference were together irradiated in gamma chamber. In order to highlight the difference between bare and coated LPGs, the evolution of spectra for bare LPG and exemplary chosen coated LPGs (PS and Ta_2O_5 film) is shown in Fig. 2. Here, the spectra marked in yellow, blue and orange correspond to readout before the irradiation, irradiation at the maximum dose and the recovery after irradiation, respectively. The variation in the resonance wavelength is mainly observed, i.e., the attenuation bands for the bare LPG exhibited a blue shift of 0.40 nm at maximum dose of irradiation (Fig. 2(a)), while there was no significant shift after maximum dose till the recovery, i.e., no recovery was observed. For bare LPG, we might attribute the trivial change in n of the inner cladding region since core and outer cladding are made of pure-silica composition. Differently, for PS-coated LPG, the attenuation band shifted of −2.51 nm when reaching the maximum dose with a decrease in visibility, moreover after the irradiation was turned off the recovery was negligible (Fig. 2(b)). Finally, for Ta_2O_5-coated LPG, the attenuation band exhibited a red shift of 0.51 nm after maximum dose of irradiation with a full recovery of the resonance wavelength when the irradiation was over (Fig. 2(c)). As evident here, the coated LPGs exhibited not trivial shifts after maximum dose of irradiation that acted differently than bare LPG. In other words, the coatings had an impact on the performance of the devices.

The real-time response, i.e. resonant wavelength shift with gamma irradiation exposure time (h)/total dose (kGy) is shown in Fig. 3(a). The gap in one of the data is related to unexpected turn off of the equipment due to a power outage at the facility, which occurred when access was not allowed. Whereas in Fig. 3(b), the resonant wavelength changes at maximum irradiation dose and after recovery period are reported, with respect to the value before irradiation. As expected, and shown in Fig. 2 (a), bare LPG is resistant to the radiation. It seems the irradiation might only slightly increase the n of the F-doped cladding. It induced an increase in the effective refractive index of the cladding modes and thus producing a slight blue shift of resonant wavelength. When LPGs coated with Al_2O_3, and PS are considered, they also exhibited a blue shift. For those coated with Ta_2O_5 and TiO_2 in turn a red shift has been noted. In particular, the spectra for Ta_2O_5 coated LPG experienced a dynamic increase in the resonance wavelength which subsequently saturated and reached the value of 0.51 nm at the highest dose, with a full recovery of the resonance wavelength when the irradiation was over. Differently, the TiO_2 coated LPGs showed a higher shift of 1.92 nm at highest dose, whereas it decreased to 0.84 nm when the irradiation was over, i.e., a partial recovery was observed. Even higher (blue) shift of −2.51 nm was then observed for PS coated LPG with negligible recovery. Finally, for Al_2O_3 LPG the shift at highest dose was negligible whereas a shift of −0.58 nm was observed during the recovery. Finally, through numerical modelling of the LPG behavior [43], we could associate the wavelength shifts during irradiation to a change in the n of the metal oxides within ± $5 \bullet 10^{-2}$. Whereas in the case of PS such change is an order of magnitude lower, because of higher thickness of the film moving the working point of the device to a slightly higher sensitivity region.

3.3. Discussion

The observed effects need to be discussed separately for polymer and metal oxides. In the first case ionizing radiation treatment is known as a specific way to modify the chemical, structural, optical, mechanical, and electrical properties of polymers by causing irreversible changes in their macromolecular structure [45,46]. Such irradiations may produce electrons and lower energy photons which are responsible for the modification of the polymer material. The possible modification is due to the breaking of polymer chains and creating free radicals. Moreover, these free radicals can also recombine to create crosslinks between the adjacent molecules. The studies reveal a variety of modifications including main chain scission, intermolecular crosslinking, creation of unsaturated bonds, formation of volatile fragments, and creation of carbonaceous clusters. Moreover, also the polymer dielectric property has been found to depend strongly on the degree of crystallinity as well as on the manner in which a particular degree of crystallinity has been attained [47–50]. In this context, PS is used to fabricate many commercial products and the material can also find applications in dosimetry [51]. Since PS is rich in electrons, damage induced by gamma irradiation is relatively limited because of the electrons available in the vicinity of the radiation damage [52]. Our results show that the modification is highly effective in the initial stage of exposure, i.e. up to 1 kGy dose, while later the effectiveness is significantly lower and rate of change drops to 20 % of the initial one. Based on literature evidence, the radiation induced mechanisms responsible of the changes we have observed could be related to both cross-linking in the polymer chains and chain scissions to smaller fragments as well as formation of color centers or other chromophores [53].

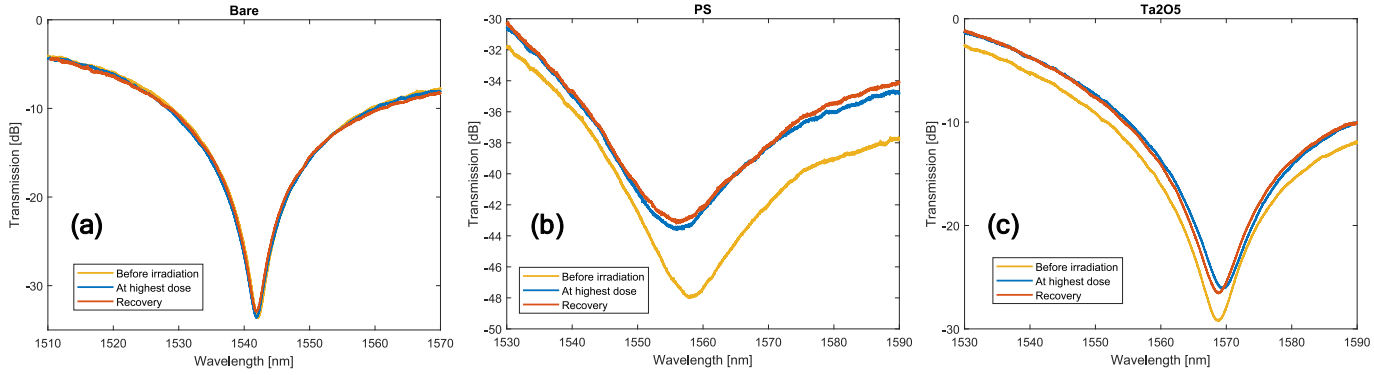

Fig. 2. Effects of gamma irradiation on LPG spectra (before irradiation, at highest dose and after recovery). The spectra for (a) bare LPG, (b) PS and (c) Ta_2O_5 nanocoated are compared as an example.

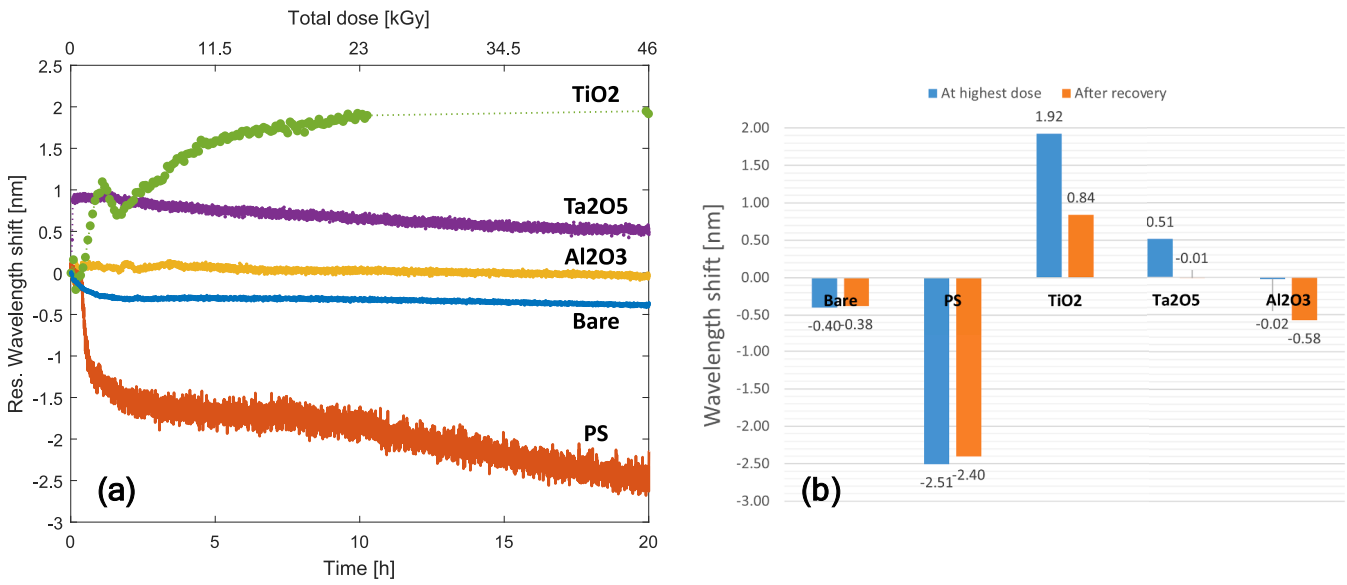

Fig. 3. Effect of gamma irradiation on resonance wavelength of the LPGs, where (a) shows real-time tracing with time/dose (the gap in TiO_2 data is due to a power outage), and (b) comparison of the shifts at highest dose and after recovery for each of the investigated cases.

In the case of metal oxides, the radiation induced vary different effects depending on the thin film material applied. In this group affected the most was TiO_2 film, while the least Al_2O_3. Interestingly, in contrast to other materials Ta_2O_5 has shown a full recovery. In all cases, the irradiation was also the most effective at the initial stage of the irradiation procedure. On top of the composition, properties of these materials are directly or indirectly dependent on the presence of defects and oxygen vacancies. The defects determine the optical, electronic and transport properties of the material and usually dominate the chemistry of its surface. Oxygen vacancies are naturally present in every oxide in the form of Schottky or Frenkel defects, and their concentration can be changed in several ways. Point defects play a fundamental role in determining physical and chemical properties of inorganic materials. This holds not only for bulk properties, but also for the surface of oxides, where several kinds of point defects exist and exhibit rich and complex chemistry. Depending on the material electronic structure, the nature of oxygen vacancies changes dramatically. Examples include non-metal vacancies at metal/non-metal sites, neutral vacancies, positively/negatively charged non-metal vacancies, free positive holes, etc. [54–56]. Several thin-film materials (including Ta_2O_5, HfO_2 and SiO_2) were investigated for their response to gamma rays and protons revealing that, with the exception of SiO_2, all materials demonstrated resilience to high doses of both gamma rays and low-energy protons [6]. In [16], the radiation damage in integrated photonic devices was evaluated in real-time in-situ using optical fibers for the interrogation, revealing RI changes of SiC and SiO_2 thin films within 10^{-5}-10^{-4}. In [14] thin films of Ta_2O_5 irradiated with gamma and beta rays exhibited efficient photon absorption, leading to reduced transmittance in tantalum, moreover the grain boundaries of the film diminished after the irradiation. Atanassova et al. [57] reported that the main source of electrically active defects in irradiated films is associated with oxygen vacancies and the broken Ta-O and Si-O bonds. Zhang et al. [58] studied the behavior of titanium oxide films prepared by reactive sputtering method, after the electron and gamma-ray radiation, and they found that the number of Ti^{4+} ions decrease and turn to Ti^{3+} according to the reaction: $2TiO_2 \rightarrow Ti_2O_3 + O$ [59]. Miyakawa et al. [60] in turn studied the effects of high-energy ions on the $12CaO{\cdot}7Al_2O_3$ films. They observed that the conductivity of the film was enhanced, and the film became colored by irradiating with ultraviolet light due to the formation of F+-like centers. Overall, based on literature the mechanisms associated to radiation induced changes in these metal oxides are related to the creation of different kinds of defects (vacancies, interstitials, or defect complexes), at same time radiation can also remove defects and restore crystalline order (annealing) [54].

3.4. Ellipsometric measurements

The results obtained with the LPG-based method were compared to the results coming from ellipsometric measurements of reference samples, i.e. thin films deposited on Si wafers in the same process and irradiated together with the LPGs by gamma. The influence of irradiation on n and k of TiO_2, Ta_2O_5, and Al_2O_3 is shown in Fig. 4(a), 4(b) and 4(c), respectively. Due to limitations of the applied ellipsometer the results were presented in 260–830 nm wavelength range. However, the fluctuations of n and k for examined thin films are rather negligible in near infrared region. Analyzing graphs presented in Fig. 4, a slight increase in n is visible when k remains rather unchanged for all the films. It must be noted that the ellipsometric measurements are done post-factum, so those after irradiation may refer only to the results after the recovery. The results do not fit well with those measured using LPGs, where an increase in n of the films would induce blue shift of the resonance wavelength. The reason for the difference may lie in applied substrate that may affect the properties of the films [61].

3.5. Multiple irradiations of selected material

Since Ta_2O_5 has shown a full recovery and rapid response at the initial stage of the procedure, the results indicate suitable properties of this material for dosimetry application. That is why the sample underwent three consecutive irradiation cycles and the corresponding results are shown in Fig. 5. It can be clearly seen that the rapid response is observed at initial stage of each of the exposures, the performance drops. The material keeps changing its properties with the dose [14] what is indicated by blue shift of the resonance wavelength. While maximum shift with radiation decreases with each radiation cycle, the recovery keeps almost the same amount inducing an overall blue shift with cycles.

4. Conclusions

In this work a universal all-optical-fiber-based solution for real-time monitoring of optical properties of thin films at ionizing radiation conditions is shown. When a structure such as long period grating is induced in the optical fiber that shows limited sensitivity to the radiation, mainly

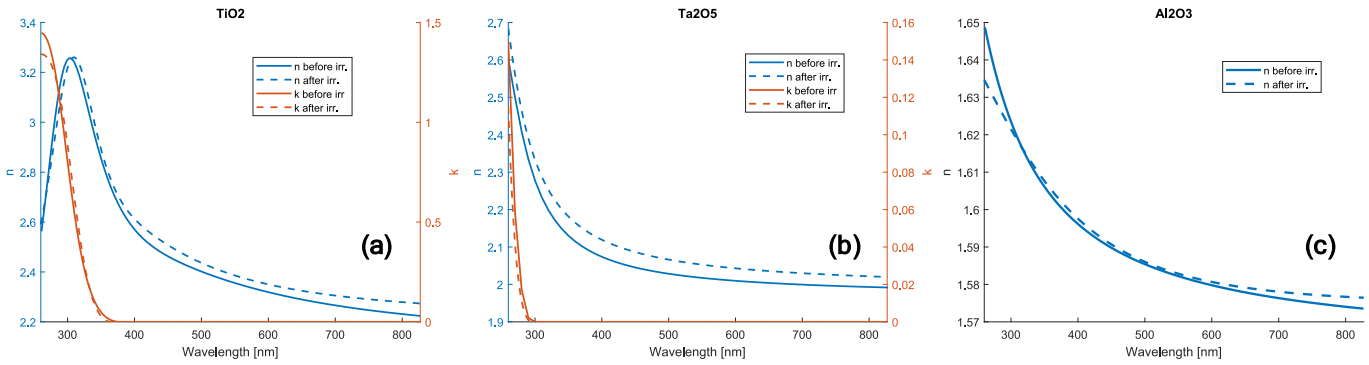

Fig. 4. The *n* and *k* of the thin films measured before and after the irradiation using spectroscopic ellipsometry, where (a) shows TiO_2, (b) Ta_2O_5, and (c) Al_2O_3. The k for Al_2O_3 is not reported as it was too low to be identified.

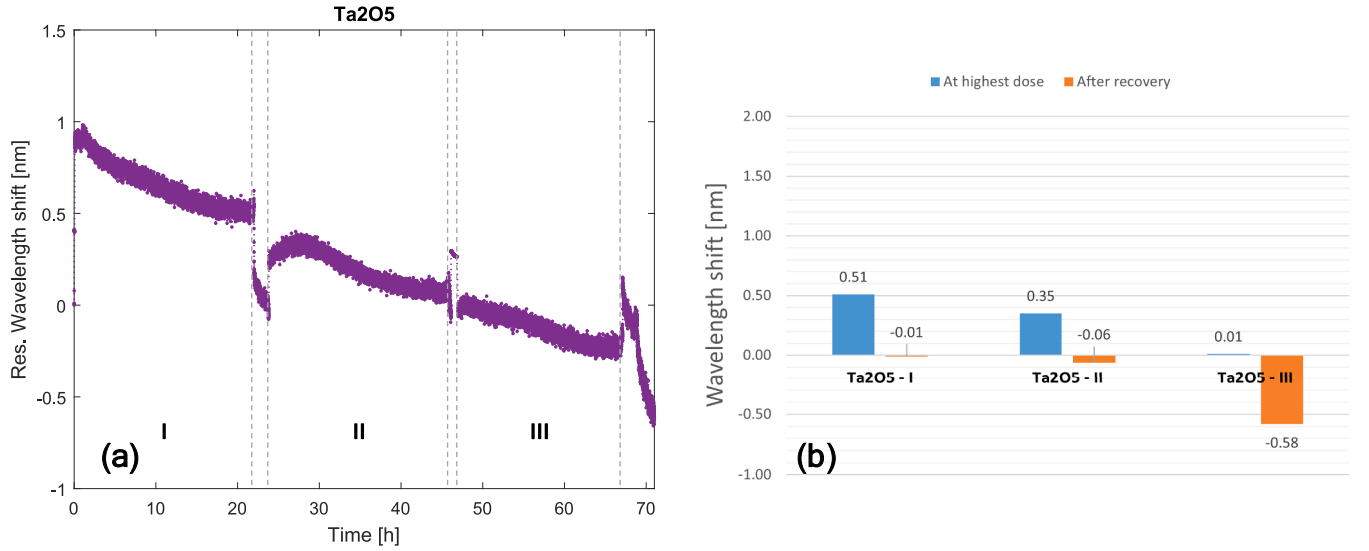

Fig. 5. Effect of multiple gamma irradiation cycles on the resonance wavelength for Ta_2O_5-coated LPG, where a) shows real-time tracing and b) shifts at highest dose and after recovery after each irradiation cycle.

properties of the surrounding medium can be monitored. To verify the method, we have deposited a series of metal oxides and a polymeric film on the LPG surface and tracked the changes induced by high doses of gamma exposure. It can be clearly seen that the changes are dependent on applied thin film material. Moreover, due to real-time monitoring capability, we were able to identify the difference in properties at the higher dose and after recovery. Out of the investigated metal oxides, TiO_2 thin film has been shown to be affected the most by the irradiation, while Al_2O_3 has been affected the least. Moreover, an interesting full-recovery capability for Ta_2O_5 thin film has been found, what can be considered as an indicator for application of this material in dosimetry. For the investigated polystyrene film, the irradiation induced permanent effect. The dynamics of the change was the highest at the initial stage of exposure. This finding makes it possible to apply the LPG-based method for optimization of the treatments, when ionizing radiation is considered as post-processing solution.

CRediT authorship contribution statement

Flavio Esposito: Writing – original draft, Investigation, Conceptualization. **Dariusz Burnat:** Investigation. **Razvan Mihalcea:** Investigation. **Daniel Negut:** Investigation. **Anubhav Srivastava:** Investigation. **Stefania Campopiano:** Supervision, Conceptualization. **Lucia Sansone:** Resources, Investigation. **Michele Giordano:** Supervision, Conceptualization. **Andrei Stancalie:** Writing – review & editing, Supervision, Conceptualization. **Agostino Iadicicco:** Writing – review & editing, Supervision, Conceptualization. **Mateusz Smietana:** Writing – review & editing, Supervision, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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