Nuclear Instruments and Methods in Physics Research B 269 (2011) 1346–1349

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Micro-Raman investigation of X or γ irradiated Ge doped fibers

A. Alessi [a,*], S. Girard [b], C. Marcandella [b], M. Cannas [c], A. Boukenter [a], Y. Ouerdane [a]

[a] Laboratoire H. Curien, UMR CNRS 5516, Université Jean Monnet, 18 rue du Pr.Benoît Lauras 42000, Saint-Etienne, France
[b] CEA, DAM, DIF, F-91297 Arpajon, France
[c] Dipartimento di Scienze Fisiche ed Astronomiche, Università di Palermo, I-90123 Palermo, Italy

ARTICLE INFO

Article history:
Received 14 February 2011
Received in revised form 22 March 2011
Available online 26 March 2011

Keywords:
Optical fiber
Drawing condition
Radiation effects
Raman spectroscopy

ABSTRACT

Micro-Raman spectra have been recorded on Ge doped optical fibers before and after 10 keV-X or γ-ray irradiation up to doses of 1 MGy (X-ray) or 7.8 MGy (γ-ray). Our data provide evidence that, at such dose levels, the glass matrix is not modified in a detectable way. We observed that varying the Ge doping levels from 0 to about 11 wt.%, X or γ radiation sensitivity of the overall matrix remains unchanged. Such results are observed for fibers obtained with drawing conditions within the usual range used for the fabrication of specialty fibers as radiation-tolerant waveguides. Our data support the potentiality of fiber-based sensors using glass properties, e.g. Raman scattering, for applications in harsh environments as those encountered in nuclear power plants.

1. Introduction

Silica glass is widely employed in different technological fields [1,2] and one of the most important is the production of optical fibers. Among the different classes of fibers, the Ge doped ones are the most used types, mainly for telecommunications applications. Ge doping was first used to increase the refractive index of the inner part (core) of the fibers with respect to that of their external part (cladding) to ensure the light guiding [1]. The researcher's attention on this class of fiber increased when the Ge doped fibers revealed to be sensitive to UV exposures, allowing the writing of fiber Bragg gratings [1,3] and when the possibility to induce non linear effects was shown [1,4].

The fiber radiation responses, and particularly the one of Ge-doped waveguides, have been widely investigated [5–11]. Researcher's objective was to improve the comprehension of radiation-induced processes for a better control on the fiber features needed by various applications.

Two main mechanisms are proposed to explain the photosensitivity of the Ge-doped materials: the so called "color center" and that named "compaction effects". In the first one, the photosensitivity is attributed to the generation or the conversion of point defects. These defects can explain, through their optical activities and the Kramers–Kroning relation, the measured refractive index changes in [6]. In the second model, the refractive index changes are attributed to a reorganization of the glass matrix as reported for example in [11].

The material density changes have been observed in UV irradiated Ge-doped fibers [12,13]. Similarly, modification of the matrix properties have been pointed out on germanosilicate samples irradiated by 50 keV electron beam [14,15]. This information is not easily extrapolated to X and γ irradiations, since the conversion to X or γ doses is not well known.

In parallel to the studies of radiation effects, the influence of the production parameters (deposition–fabrication temperature of the preform [16], tension, speed and temperature drawing of the fiber [17–19]) on Ge-doped glasses has been widely investigated too.

In the present investigation, we report data regarding the Raman activity, before and after X and γ irradiations, of various Ge-doped optical fibers differing by their doping profiles and drawing parameters. We focused our study on high doses since optical fibers are more and more considered for use in high dose radiation environments, mainly as part of strain or temperature sensors [20]. Many of these sensors and components are based on Raman or Brillouin scattering, so their performances will strongly depend on the glass matrix radiation response.

2. Experimental

We have studied optical fibers produced by iXFiber SAS. The samples have been obtained starting from two preforms realized using the modified chemical vapor deposition technique. The fiber named FGe is doped with different amounts of Ge according to a 4-steps radial distribution (maximum content of ~11 wt.% at the core center, see Fig. 1). The fiber FGe has been produced using a drawing speed of ~40 m/min, a tension of ~70 g, and a furnace-temperature of ~1600 °C. At variance the fibers, named FGeDi (i = 1,2,3),

* Corresponding author.
E-mail address: antonino.alessi@univ-st-etienne.fr (A. Alessi).

0168-583X/$ - see front matter © 2011 Elsevier B.V. All rights reserved.
doi:10.1016/j.nimb.2011.03.021

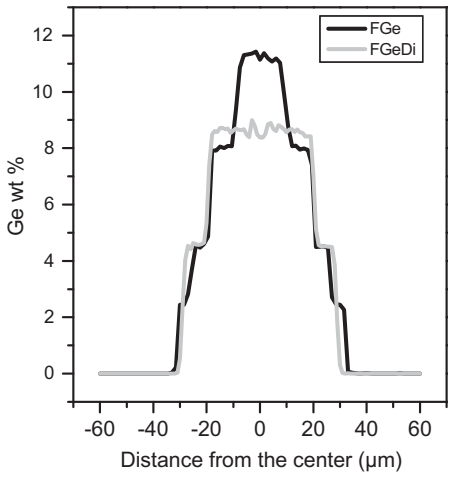

Fig. 1. Ge doping profiles of FGe and of FGeDi (i = 1,2,3) (▬). The center of the fiber core is at the 0 coordinate.

have been doped according to a 2-steps radial distribution of the Ge content (maximum content 9 wt.%, see Fig. 1). All FGeDi fibers have been produced starting from the same original preform. They differ only by the drawing conditions.

FGeD1 fiber has been obtained with a drawing speed of 70 m/min under a tension of 135 g. These parameters were 40 m/min and 70 g for FGeD2, 22 m/min and 33 g for FGeD3.

We have recorded Raman spectra at room temperature using an Aramis (Jobin–Yvon) spectrometer, that is supplied by a CCD camera, an Argon laser (λ = 488 and power ~3 mW), step motors and by a 50× objective. All the spectra have been acquired using the same experimental conditions ensuring a spatial resolution of ~5 μm.

The samples have been X (10 keV) irradiated at room temperature using the ARACOR facility at the French atomic energy center (CEA); whereas the γ-ray irradiations were performed at room temperature using the Brigitte facility at SCK-CEN (Belgium).

3. Results

It is important to underline that the Raman spectra of the FGeD1, FGeD2 and FGeD3 samples recorded before irradiation are very similar as previously reported in [21].

In Fig. 2, we report the Raman spectra (range 350–950 cm^{-1}) recorded in the core center of the FGe and FGeD3 fibers before and after X-ray irradiation at 1 MGy. The presence of different bands appears clearly in all the spectra. The most intense band, peaking at around 440 cm^{-1} (called R band), is associated to the symmetric stretching of bridging oxygen atoms (Si–O–Si) [22,23]. Another one, called D2, peaked at ~600 cm^{-1}, is related to the three-members rings [22,23]. At variance, the band located at 800 cm^{-1} is due to the network [22], TO and LO modes of the silica [24,25]. We also note that the so-called D1 band associated to the four-member rings [22,23,25] can not be clearly distinguished in the different spectra. FGe sample measurements reveal, at 700 cm^{-1}, the presence of a band with low amplitude. Such a band has been associated to Ge–O–Si vibration modes in [25].

For both types of fibers, the comparison between the spectra recorded before and after the irradiation does not reveal significant differences. Same results are observed for all the fibers transverse cross sections. As an example, in Fig. 3a, we report the relative amplitude (with respect to the R band) of the D2 band as a function of the radial distance before and after irradiation in the sample FGeD3. These curves have been obtained by subtracting a tangent

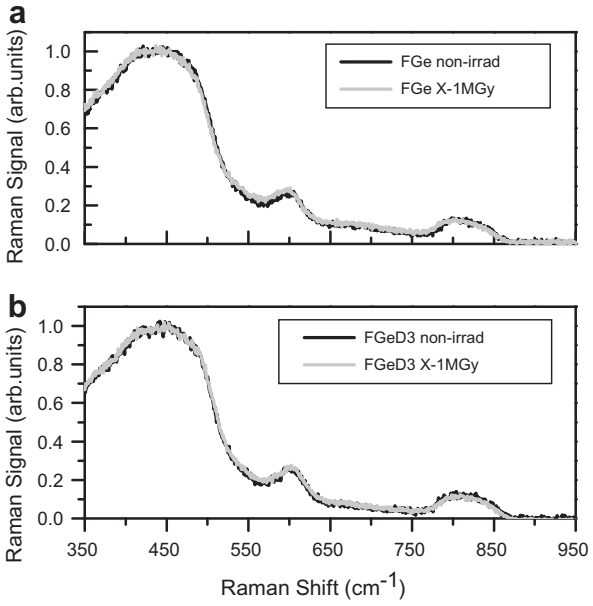

Fig. 2. (a) Raman spectra of FGe fiber and (b) FGeD3 fiber before (▬) and after (▬) X-ray irradiation at 1 MGy.

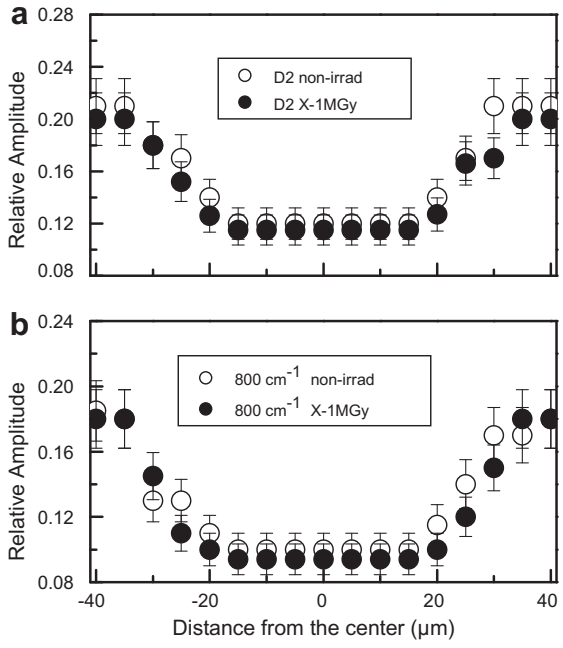

Fig. 3. Relative amplitude of D2 band (panel a) and 800 cm^{-1} band (panel b) recorded as a function of the distance from the FGeD3 core center before (○) and after (●) X-irradiation at 1 MGy.

to the minima of the band. We also note that the D2 band amplitude varies along the fiber diameter as expected from the fact that the presence of Ge atoms modifies the concentration of the three-member rings [25,26]. Similarly, the fact that the D1 band can not be distinguished in the core spectra agrees with previous results [25,26].

The invariance of the Raman spectra after irradiation at the dose of about 1 MGy is supported by Fig. 3b that shows the radial dependence of the relative amplitude of the 800 cm^{-1} band for FGeD3 fiber before and after the irradiation. As for the D2 band, the 800 cm^{-1} band behavior before and after irradiation does not

provide evidence for significant changes of the glass matrix properties. Furthermore, we note that its amplitude depends on the location along the fiber diameter. As for the D2 band this result is related to the Ge content changes and it is in agreement with previous data [25].

We observe very similar results for FGeD1 and FGeD2 fibers. In Fig. 4, we report the Raman spectra of the central part of the three fiber's cores after the 1 MGy X-ray irradiation.

Fig. 5 illustrates the Raman spectra of the FGe central part core before and after γ irradiation at 1.1 and 7.8 MGy.

The three spectra are very similar. This similarity is also observed for the other parts of the fiber differently Ge-doped or made of pure silica.

To highlight this result, we report, in the inset of Fig. 5, the radial distribution of the relative amplitude of the D2 band. We observe that its amplitude decreases when increasing Ge content as observed in FGeDi samples.

To investigate the effects of γ-rays on the FGeDi fibers, we have irradiated the FGeD2 fiber up to the dose of 7.8 MGy. As for FGe fiber, no significant changes in the Raman spectra are observed after irradiation. The radiation hardness of the glass matrix, respect to Raman scattering, is observed along all the fiber diameter regardless of the chemical composition of the different layers.

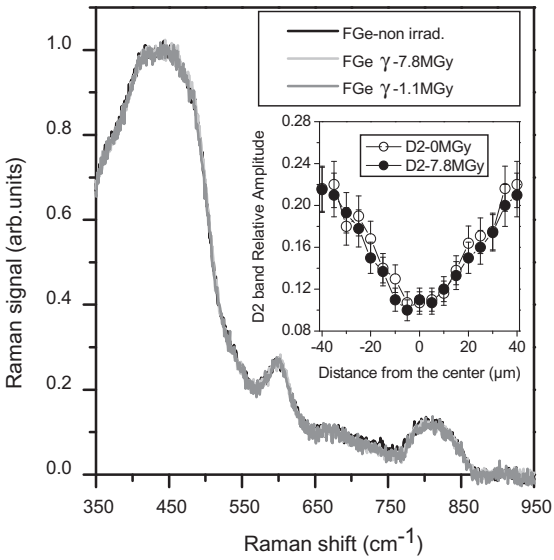

Fig. 5. Raman spectra of the FGe sample before irradiation (▬), at 1.1 MGy (γ-irradiation) (▬) and at 7.8 MGy (γ-irradiation) (▬). The inset reports the D2 relative amplitude as a function of the distance from the fiber center.

4. Discussion

The Raman results obtained on the FGeDi fibers show that within the range of drawing speed and tension no significant differences are observed before (previously reported in [21]) and after irradiation up to doses of 1 MGy. This latter finding indicates that varying these drawing parameters cannot lead to a significant modification of the matrix Raman response to radiations. This consideration can be extrapolated to all the germanosilicate specialty fibers as the studied range covers the domain of tension and speed usually employed for the production of fibers like radiation-tolerant optical fibers. However, this work cannot be directly extrapolated to the case of Telecom-type Ge-doped fibers as these fibers can be produced using larger variation of these parameters.

For higher irradiation doses (up to ~8 MGy), we have measured no change in the Raman spectra acquired for FGe and FGeD2 fibers. The data of the FGe γ irradiated samples confirm that the slight differences observed in the 550–600 cm^{-1} range (Fig. 2a) are negligible. The FGeD2 fiber was produced using intermediate values of drawing speed and tension (with respect to the FGeDi fibers). So, results obtained for the FGeD2 fiber can be extended to FGeD1 and FGeD3 fibers. In the three samples, γ irradiation up to 7.8 MGy dose does not lead to Raman detectable changes in the glass structure. Our results on the FGe and FGeDi fibers show that the change of the doping profiles does not affect the matrix response to the radiation within the explored range of drawing conditions and doping profile features: doping content (0–11 wt.%), number and dimension of steps, further precise investigations could be devoted to the edge zones between different doping level steps (~20 μm for example) also performing stress measurements before and after irradiation. Our finding differs from results obtained on UV-irradiated-surface of Ge doped silica samples [27]. These different radiation induced effects have to be considered comparing the bulk and surface properties or comparing our data with that of Ref. [12].

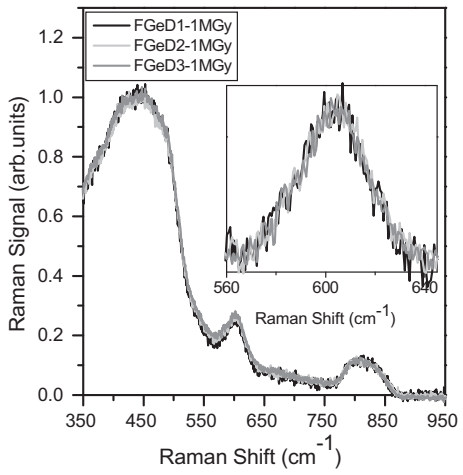

Fig. 4. Raman spectra recorded in the core centers of FGeD1 (▬), FGeD2 (▬) and FGeD3 (▬) 1 MGy X-ray irradiated samples. The inset reports the D2 bands for the three FGeDi fibers.

5. Conclusions

We measured the Raman spectra of different types of pristine, X and γ irradiated Ge doped optical fibers. We found no differences between the Raman spectra recorded before and after irradiation up to doses of 7.8 MGy. These results are observed in all the fibers zones, regardless of the Ge doping levels (up to 11%). This suggests that in the explored ranges of doses and doping levels the Ge content change does not modify the radiation sensitivity of the glass matrix. Moreover, the comparison between fibers obtained from the same original preform shows that within the investigated range of drawing speed and tension, these parameters did not impact the radiation response of the Ge doped silica matrix.

These results support the potentiality of fiber-based sensors using glass properties, e.g. Raman scattering, for applications in harsh environments as those encountered in nuclear power plants.

Acknowledgements

We acknowledge the members of the LAMP group (http://www.fisica.unipa.it/amorphous/) for support with interesting discussions. We also acknowledge B. Brichard from SCK-CEN for providing gamma-ray irradiations.

References

[1] Structure and Imperfections in Amorphous and Crystalline Silicon Dioxide, edited by R.A.B. Devine, J.P. Duraud, and E. Dooryhée Wiley, New York, 2000.
[2] R. Kitamura, L. Pilon, M. Jonasz, Appl. Opt. 46 (2007) 8118.
[3] K.O. Hill, Y. Fujii, D.C. Johnson, B.S. Kawasaki, Appl. Phys. Lett. 32 (1978) 647.
[4] U. Osterberg, W. Margulis, Opt. Lett. 11 (1986) 516.
[5] E.J. Friebele, D.L. Griscom, G.H. Sigel Jr., J. Appl. Phys. 45 (1974) 3424.
[6] D.P. Hand, P.St. Russell, Opt. Lett. 15 (1990) 102.
[7] H. Shigemura, Y. Kawamoto, J. Nishii, M. Takahashj, J. Appl. Phys. 85 (1999) 3413.
[8] S. Girard, I. Trenteseaux, Y. Ouerdane, J.-P. Meunier, A. Boukenter, J.-L. Derep, L. Thonnérieux, Appl. Phys. Lett. 83 (2003) 219.
[9] B. Brichard, O.V. Butov, K.M. Golant, A.F. Fernandez, J. Appl. Phys. 103 (2008) 054905.
[10] A. Alessi, S. Agnello, S. Grandi, A. Parlato, F.M. Gelardi, Phys. Rev. B 80 (2009) 014103.
[11] H.G. Limberger, P.-Y. Fonjallaz, R.P. Salathé, F. Cochet, Appl. Phys. Lett. 68 (1996) 3069.
[12] M. Takahashi, T. Uchino, T. Yoko, J. Am. Ceram. Soc. 85 (2002) 1089.
[13] E.M. Dianov, V.G. Plotnichenko, V.V. Koltashev, Yu.N. Pyrkov, N.H. Ky, H.G. Limberger, R.P. Salathe, Opt. Lett. 22 (1997) 1754.
[14] A.-S. Jacqueline, B. Poumellec, J.C. Chervin, S. Garcia-Blanco, S. Esnouf, Mater. Sci. Eng. B 107 (2004) 46.
[15] S. Garcia-Blanco, A.-S. Jacqueline, B. Poumellec, J.S. Aitchison, J. Non-Cryst. Solids 351 (2005) 2085.
[16] S. Girard, Y. Ouerdane, A. Boukenter, J.P. Meunier, J. Appl. Phys. 99 (2006) 023104.
[17] H. Hanafusa, Y. Hibino, F. Yamamoto, Phys. Rev. B 35 (1987) 7646.
[18] J. Hong, FTIR investigation of amorphous silica fibers and nano-size particles, ProQuest Dissertations and Theses; Thesis (Ph.D.) (Rensselaer Polytechnic Institute, Troy, 2003).
[19] K. Tsujikawa, K. Tajima, M. Ohashi, J. Lightwave Technol. 18 (2000) 1528.
[20] K.T.V. Grattan, T. Sun, Sensors and Actuators 82 (2000) 40.
[21] A. Alessi, S. Girard, C. Marcandella, M. Cannas, A. Boukenter, Y. Ouerdane, J. Non-Cryst. Solids 357 (2011) 24.
[22] F.L. Galeener, A.E. Geissberger, Phys. Rev. B 27 (1983) 6199.
[23] A.E. Geissberger, F.L. Galeener, Phys. Rev. B 28 (1983) 3266.
[24] F.L. Galeener, G. Lucovsky, Phys. Rev. Lett. 37 (1976) 1474.
[25] G.S. Henderson, D.R. Neuville, B. Cochain, L. Cormier, J. Non-Cryst. Solids 355 (2009) 468.
[26] X. Nian, X. Zhisan, T. Decheng, J. Phys.: Condens. Mater. 1 (1989) 6343.
[27] F.X. Liu, J.Y. Qian, X.L. Wang, L. Liu, H. Ming, Phys. Rev. B 56 (1997) 3066.