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LIGHT – AN IDEAL INSTRUMENT TO HIGHLIGHT AND QUANTIFY THE EFFECTS INDUCED BY IONIZING RADIATION TO OPTICAL MATERIALS: A REVIEW

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Abstract. In this work, we review the recent developments in the area of using light as instrument to highlight and quantify the effects induced by ionizing radiation to optical materials. It is known that the light, either we refer to the visible wavelengths range of the spectrum or to the other wavelengths, is an ideal vehicle to transport information. Due to its unique characteristics, it can be used in a wide range of applications. Beside other emergent research domains such as medical physics, in the last years, the light started to be more and more involved in the state-of-the-art nuclear physics experiments, as those involved in the frame of Extreme Light Infrastructure – Nuclear Physics (ELI-NP) state-of-the-art European Research Facility, where Laser Technology and Nuclear Physics show their combined potential. One of the most important usability of light in relation to nuclear physics, as characterization instrument, is in dosimetry. It was shown that in specific cases, as for example those when strong electromagnetic pulses are involved, the dosimetry based on light and solid state detectors, which do not contain electronic parts, is the indicated solution. Beside dosimetry, the light is an outstanding instrument for analyzing and characterizing the structural defects induced by ionizing radiation to optical materials.

Key words: light, optical materials, ionizing radiation, “Glass browning” phenomenon, dosimetry, structural characterization.

1. INTRODUCTION

It is known that any material, natural or anthropogenic, is characterized by a series of physical properties such as: structural, mechanical, electrical, magnetic, thermal, and optical [1, 2]. In the last years, the development of laser technique as diversity of applications and performances showed outstanding improvements in a wide range of fields in science, technology, and also in social life [3]. Along with the classical optics, these aspects led to new possibilities and methods for the characterization of materials, but also to the discovery of new improved ones. These discoveries led naturally to the development of new technologies for obtaining materials having special properties, with a great impact in the social life.

The optical properties of materials represent an important aspect that can be used for studying a wide range of their characteristics. One of the most important means to perform this task is represented by the use of coherent (laser) or incoherent light sources in order to highlight and quantify the phenomena occurred in the frame of light-mater interaction. Studying these phenomena can lead to the knowledge of material’s band gap structure, the content of wanted/unwanted impurities, local defects, electrical and magnetic properties etc. [1, 2]. In these kinds of experiments, direct physical observables, such as reflectivity, transmittance, and absorption are measured. From these direct measurements, derived quantities can be determined, such as: dielectric function, optical conductivity, optical magnetic permittivity, energies of excitation etc.

In this work, we discuss the effects that occur into optical materials due to their exposure to electromagnetic (*i.e.* X-rays and gamma-rays) and corpuscular (*i.e.* alpha particles and protons) ionizing radiation. This paper might be welcomed more than ever due to the new developments of the nuclear techniques, involving new types of particle accelerators and high energy bright gamma-ray sources, such as those based on extreme power ultra-short pulses laser systems (ELI-NP) [3–7]. To date, many scientific works related to this topic were published in literature [8–16]. The problems of using optical materials in ionizing radiation environments are quite complex due to the fact that electromagnetic and corpuscular ionizing radiation lead to different effects in the structure of the same materials [12].

In this paper, an overview of several applications of optical materials after being exposed to ionizing radiation fields was done. Among these applications, the most important are: the ones related to the quantities that are proper to be used as dosimetric parameters [17], the ones related to analysing and quantifying the effects produced into the images captured through optical materials affected by ionizing radiation [18, 19], the ones related to analysing the effects produced at the surface of optical materials by accelerated particles using fractal techniques [20], the ones related to the determination of gamma-ray energy occurred in the frame of nuclear reactions [21], the ones related the determination of the distribution of a collimated ionizing radiation beam [22] etc.

2. LIGHT AND OPTICAL MATERIALS

When interacting with optical materials, the incident light I_0, is either transmitted, absorbed, or reflected as it is shown in Fig. 1, where I_T, I_A, and I_R are the intensities of the transmitted, absorbed, and reflected light, respectively. Besides I_0, I_R, and I_T, which are directly measurable quantities, I_A can be determined from the energy conservation law.

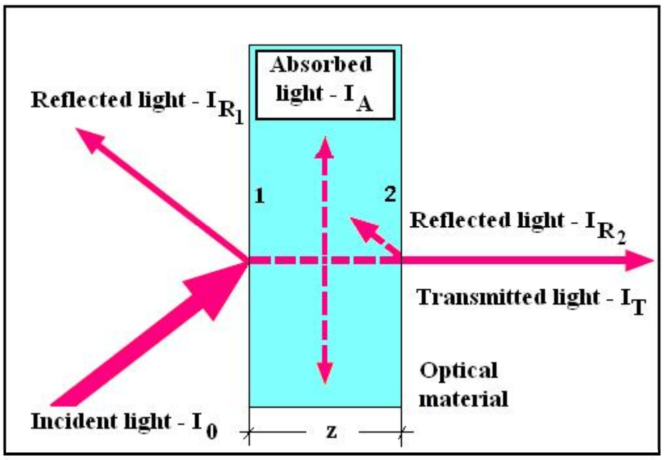

Fig. 1 – Interaction of light with optical materials.

In order to determine and quantify the properties of an optical material passed through by light, starting from the optical phenomena described above, a series of derived quantities can be obtained, such as: R (reflectivity), T (transmissivity), A (absorptivity), α (absorption coefficient), k (extinction coefficient), σ (optical conductivity), ε_1 (real dielectric constant), ε_2 (imaginary dielectric constant), μ (magnetic permeability), E_g (optical band gap energy) [23] etc. The reflection at the surface is described by the coefficient of reflection or reflectivity (R), and it is defined as the ratio between the reflected intensity and the incident intensity [16]. The coefficient of transmission or transmissivity (T) is defined as the ratio between the transmitted intensity and the incident intensity [11]. If there is no absorption or scattering, then, by following the energy conservation law, the following relation is valid: $R + T = 1$. If $T \sim 1$ and $R \sim 0$ condition are satisfied, the optical material is considered to be transparent (light is passing thru it). If $T \sim 0$ condition is satisfied, the optical material is considered to be opaque (light is not passing through it).

The absorption of light or absorptivity (A) by an optical material is quantified by its absorption coefficient, α, and it is defined as the fraction of the absorbed intensity in a unit length within the material [24]. The relation between the absorptivity, reflectivity and the transmissivity within an optical material is given by the Lambert-Beer's Law, as follows:

$$T = (1 - R_1) \cdot (1 - R_2) \cdot e^{-\alpha \cdot z}, \tag{1}$$

where z is the thickness of the optical material and R_1 and R_2 are the reflectivities of the front and back surfaces, respectively. For transparent optical materials, the following relation is valid:

$$T = \mathrm{e}^{-\alpha \cdot z} = \mathrm{e}^{-A}, \tag{2}$$

where $A = \alpha \cdot z$, represents the absorbance.

The absorption coefficient is described by the following relation:

$$\alpha = \frac{1}{z} \cdot \ln \frac{1}{T}. \tag{3}$$

The extinction coefficient (k) represents a kind of absorption coefficient, describing the diffusion of the light passing through a material [23, 25, 26], and is defined by the following relation:

$$k = \frac{\alpha \cdot \lambda}{4 \cdot \pi}, \tag{4}$$

where λ represents the wavelength of the light.

The optical conductivity (σ) is a material property that links the current density to the electric field. In this sense, this linear response function is a generalization of the electrical conductivity. The optical conductivity always remains finite in some frequency intervals (above the optical band gap in the case of insulators) [23], and it is described by the following relation:

$$\sigma = \frac{c \cdot \alpha \cdot n}{4 \cdot \pi}, \tag{5}$$

where n represents the refraction index and c is the speed of light.

The real dielectric constant (ε_1) and the imaginary dielectric constant (ε_2) represent the real and the imaginary part of the complex dielectric function, respectively. The real part indicates how the speed of light in a material can be slowed down while the imaginary part deals with the absorption of energy by a dielectric from electric field due to dipole motion [23, 27]:

$$\varepsilon_1 = n^2 - k^2, \quad \varepsilon_2 = 2 \cdot n \cdot k. \tag{6}$$

The magnetic permeability (μ) indicates the permanent magnetization state of a material [23], and can be expressed as:

$$\mu = \frac{\lambda \cdot \alpha^2}{c \cdot \pi \cdot \sigma}, \tag{7}$$

where μ_r and μ_0 represents the relative permeability and the permeability of free space, respectively.

The optical band gap energy (E_g) of an optical material shows the type of optical transition of a material and the dependence of absorption coefficient on the photon energy. The energy of the optical band gap can be determined from the fundamental absorption of the material, which corresponds to the excitation of the electrons from the valence band to the conduction band, and it can be expressed as follows:

$$\alpha \cdot E_\lambda = \mathrm{const} \cdot (E_\lambda - E_g)^n, \tag{8}$$

where “const” is a material’s specific constant and n is a number that characterizes the transition process and is theoretically equal to ½ for direct transitions and 2 for indirect transitions [23, 27].

3. OPTICAL MATERIALS IN THE PRESENCE OF IONIZING RADIATION

When interacting with ionizing radiation, the optical materials are affecting by the high energy deposition into their volume, leading to the so called “glass browning” phenomenon or RIA (radiation induced absorption) [17], as it is shown in Fig. 2 (gamma-rays). The effects induced in the optical materials by the ionizing radiation (electromagnetic, corpuscular or mixed) are given by the specific interactions between the radiation and the microscopic constituents of the exposed material (molecules, atoms, orbital electrons, nuclei). In the interaction processes the energy transported by the radiation is transferred to the material's constituents. According to the magnitude of energy and the type of the ionizing radiation, different types of effects can occur such as: color centers production (RIA) [17], induced structural defects [20], thermal transfer or nuclear reactions (activation processes) [12]. All these effects may appear individually or simultaneously. The energy transferred to the exposed material propagates gradually in the material's volume, initially to electrons, and, from them, further, to atoms and molecules. This surplus of energy leads, by physical and chemical mechanisms, to the formation of free radicals, atomic and molecular ions, new molecules etc., but also to modifications in the structure of the material. The main quantity of energy transferred to the material is transformed in thermal energy [22]. All the other effects are representing only a small quantity of the transferred initial energy. In the case of the corpuscular ionizing radiations, the effects induced to the exposed optical materials are mostly of structural nature, unlike the electromagnetic type ones, which are mostly described by the induction of supplementary absorption bands. It was shown [12], that the presence of impurities (wanted or not) in the structure of the optical material exposed to ionizing radiation leads to poor performances in terms of radiation hardness.

As it was shown in literature [28, 29], an exposure of the irradiated optical materials to a controlled heating treatment will lead to a regain of their initial optical performances (Fig. 2).

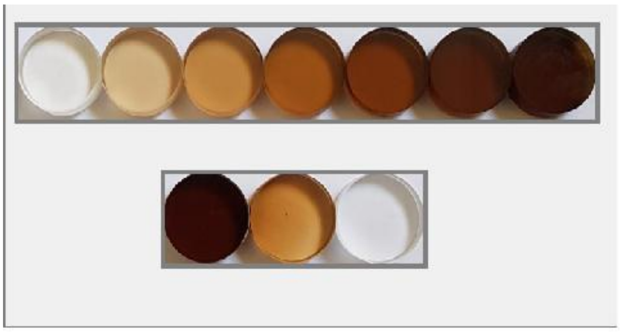

Fig. 2 – "Glass browning" phenomenon due to exposure to gamma-rays (darker tones representing higher absorbed dose values) and the thermal reversing processing of exposed optical material (different stages during the process).

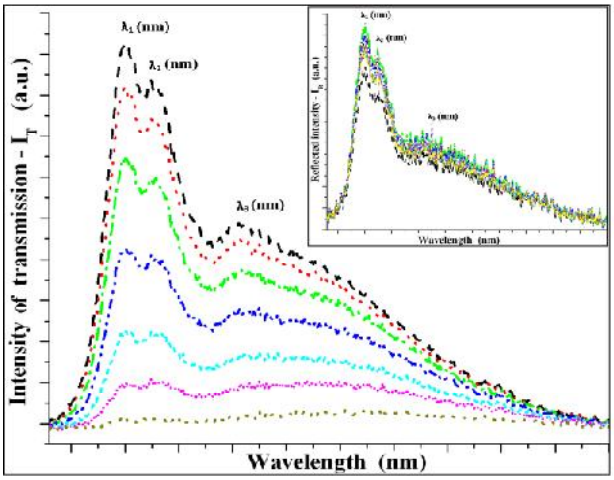

Fig. 3 – Representation of the intensity of transmission/reflection of the exposed optical materials as a function of the wavelength (different absorbed dose values).
Adapted from Ref. [16].

As it can be seen form Fig. 2, the magnitude of the "glass browning" phenomenon, is proportional to the absorbed dose values, on certain intervals, which makes it suitable to be used in dosimetry. In Fig. 3, a plot of the intensity of transmission/reflection of the exposed optical materials as a function of the wavelength (different absorbed dose values) is shown. In Fig. 4, a plot of the intensity of transmission associated to the exposed optical materials as a function of the absorbed dose value, for the three wavelengths values corresponding to maximal intensities (λ_1, λ_2, λ_3), is presented.

The variation of the absorption coefficient with the absorbed dose value (Fig. 5) is described by the following relation:

$$\alpha(D) = m - n \cdot \mathrm{e}^{-k \cdot D}, \tag{9}$$

where m, n, and k are the fitting parameters.

As it was shown in literature, using laser light it is highly recommended due to its unique characteristics: directionality, mono-chromaticity, and coherence [30–34]. In this case, when the initial and the transmitted laser powers are measured, the Lambert-Beer's Law can be used in the following form:

$$P_T(z) = P_0 \cdot \mathrm{e}^{-\alpha \cdot z}, \qquad (10)$$

where P_0 and P_T are the initial and the transmitted laser powers, respectively.

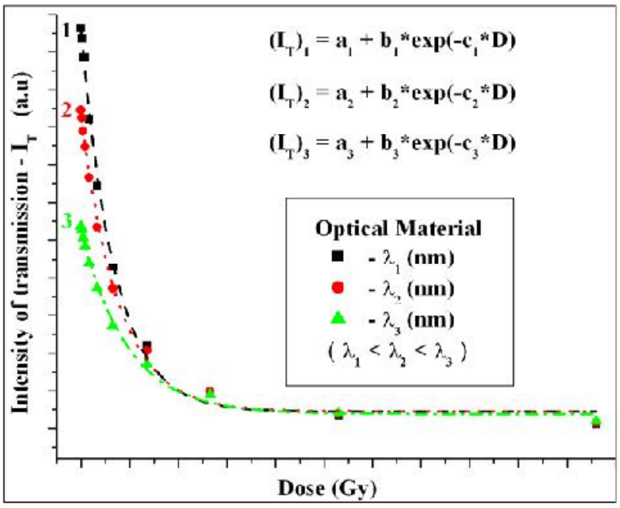

Fig. 4 – Intensity of transmission associated to the exposed optical materials as a function of the absorbed dose. Adapted from Ref. [26].

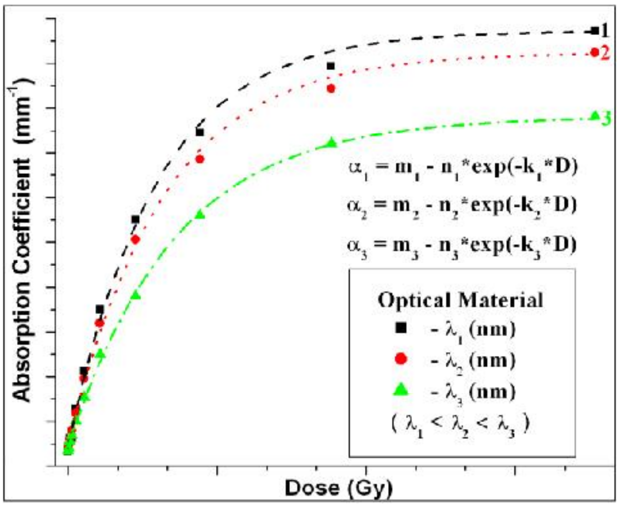

Fig. 5 – Absorption coefficient of the exposed optical materials as a function of the absorbed dose.

As it was shown in literature [17, 19, 22, 24–26, 35–41], all the parameters presented in the previous part of the present paper show a specific variation related to the exposure of an optical material to ionizing radiation. This variation is linear on certain dose intervals, as it is shown in Figs. 6 and 7, which allows the possibility of using them in gamma-ray dosimetry. In Figs. 6 and 7, the slopes (b_1, b_2, b_3) represent the rates of change of the dosimetric parameter values as a function of the absorbed dose.

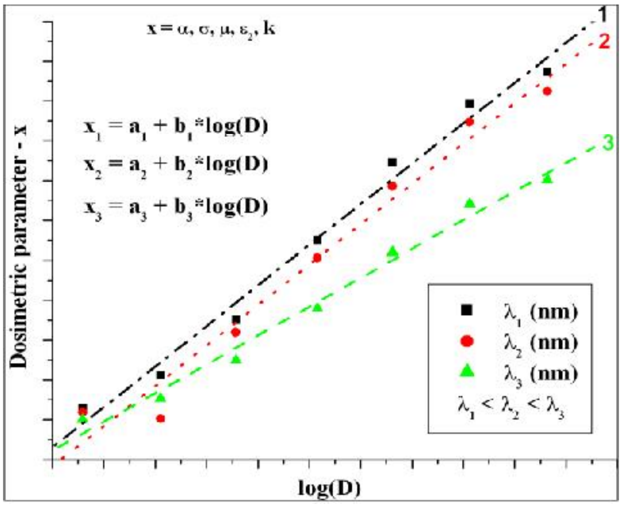

Fig. 6 – Linearized interval of the dosimetric parameters associated with the exposed optical materials as a function of the absorbed dose (calibration curves) – direct proportional parameters. Adapted from Ref. [26].

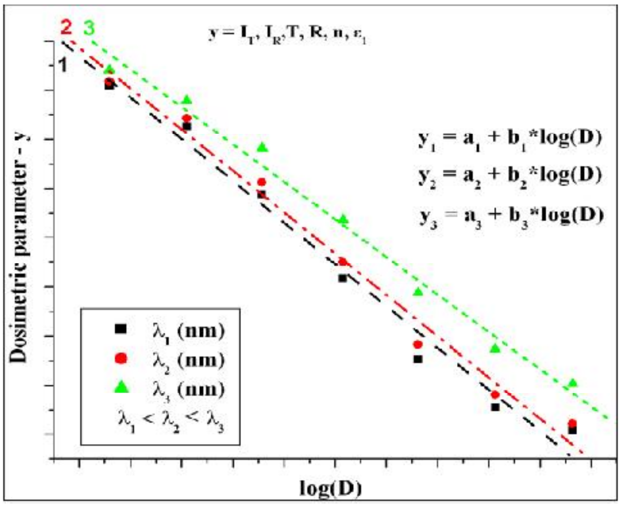

Fig. 7 – Linearized interval of the dosimetric parameters associated with the exposed optical materials as a function of the absorbed dose (calibration curves) – inverse proportional parameters.

To be able to use all of the previous mentioned parameters in the field of gamma-ray dosimetry, a calibration protocol is needed to be performed. This calibration procedure means the exposure of a set of quasi-identical optical materials to different well-known absorbed dose values. The absorbed dose values can be measured using validated classical dosimeters in order to provide traceability to the results and/or by complementary computational Monte Carlo validated methods [42–46]. By determining the values corresponding to one or more of the parameters presented above and by correlating them to the corresponding dose values, calibration curves can be obtained. By determining the value of the chosen dosimetric parameter corresponding to an "unknown" absorbed dose value and by using these calibration curves, the "unknown" dose value can be obtained [17].

The general expression of the quantities that can be used as dosimetry parameters, obtained by fitting the experimental data, is:

$$x_i = (a_x)_i + (b_x)_i \cdot \log(D_{d_i}), \tag{11}$$

where $x = (I_T, T, R, I_R, \alpha, k, \sigma, \varepsilon_1, \varepsilon_2, \mu, E_g)$ are the dosimetric parameters, $(a_x)_i = ((a_k)_i, (a_\alpha)_i, (a_\sigma)_i, (a_{\varepsilon 2})_i, (a_\mu)_i, (a_T)_i, (a_{IT})_i, (a_{IR})_i, (a_{Eg})_i)$ and $(b_x)_i = ((b_k)_i, (b_\alpha)_i, (b_\sigma)_i, (b_{\varepsilon 2})_i, (b_\mu)_i, (b_T)_i, (b_{IT})_i, (b_{IR})_i, (b_{Eg})_i)$ are the fitting parameters, d is the criterial number of the absorbed dose and $i = (\lambda_1, \lambda_2, \lambda_3)$ nm represent the maximal values from the transmission/reflection spectrum of the chosen optical material.

In order to obtain the dependence of the absorbed dose to the fitting parameters, the equation (11) is transformed as follows:

$$D_{di} = 10^{\frac{x_i - (a_x)_i}{(b_x)_i}}. \tag{12}$$

As any determined value associated to directly measurable quantities, the results are affected by uncertainties. In order to evaluate the uncertainty associated to equation (12), the error propagation law was applied, leading to the following relation:

$$\sigma^2_{D_{di}} = \left(\frac{D_i}{(b_x)_i}\right)^2 \cdot \left[2 \cdot \sigma_{(a_x)_i} + \left(\left(\log(D_{di})\right)^2 + \left(\frac{x_i - (a_x)_i}{(b_x)_i}\right)^2\right) \cdot \sigma^2_{(b_x)_i} + \left(\frac{(b_x)_i}{2.3 \cdot D_{di}}\right) \cdot \sigma^2_{D_{di}}\right]. \tag{13}$$

All the quantities from Eq. (13) are known [17].

As it was shown in literature, beside its usability in dosimetry, the light can also be used in the characterization and quantification of the effects induced by ionizing radiation (electromagnetic and/or corpuscular) to optical materials. As an example, it was shown that it can be used to characterize the structural defects induced to optical materials by analyzing digital images of the studied samples using complex fractal techniques [20]. Another application of light in characterizing optical materials exposed to electromagnetic ionizing radiation is the one based on the study, by various techniques [18, 19, 47–50], of digital RGB images taken through the studied samples. It was shown that the distribution of a collimated ionizing radiation beam can be also determined, by studying the digital images taken to an optical material placed into it [22]. Another very important application of the light and of the optical materials in nuclear physics is for determining the gamma-ray energy occurred in the frame of nuclear reactions, see Ref. [21].

4. CONCLUSIONS

In this paper, we have reviewed the recent studies and developments in the area of using light as an ideal instrument to highlight and quantify the effects induced by ionizing radiation to optical materials. A few of the most important aspects regarding the possible applications of optical materials after being exposed to ionizing radiation fields were presented such as: the ones related to the observable quantities that are suitable to be used as dosimetric parameters, the ones related to analyzing and quantifying the structural defects induced to optical materials by analyzing the digital images of the studied samples using complex fractal techniques, the ones related to characterizing optical materials based on the study of digital RGB images taken through the studied samples, the ones related to determining gamma-ray energy occurred in the frame of nuclear reactions, and the ones related to determining the distribution of a collimated ionizing radiation beam. Also, the main quantities used in dosimetry were thoroughly presented in this paper: reflectivity, transmissivity, absorptivity, absorption coefficient, extinction coefficient, optical conductivity, dielectric constant, magnetic permeability, and optical band gap energy.

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