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Application of spectroscopic techniques in the radiation dosimetry of glasses: An update

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Abstract. The colorimetry and thermoluminescence properties of gamma irradiated glass were reported in as early as 1920. The utility of radio-photoluminescence (RPL) of silver activated metaphosphate glass for monitoring high doses of accidental and routine gamma radiation was reported in the 1960s. Since then considerable amount of research work has been carried out to study the thermoluminescence (TL), optical absorption (OA), electron paramagnetic resonance (EPR) and optically stimulated luminescence (OSL) of different commercially available glasses for high as well as low dose applications. A brief review of the progress made in the spectroscopic studies of glasses during the past few decades and the application of glasses for radiation dosimetry has been given in this paper.

1. Introduction

Glasses play an essential role in a variety of scientific fields and in industry due to the ease of their preparation and shaping and their long term stability. Some of the common applications are flat glass, container glass, optics and optoelectronics material, laboratory equipment, thermal insulator, reinforcement fiber and art. When an ionising radiation such as X or gamma ray interacts with glasses, many physical and/or chemical changes occur, which can be used for quantifying the amount of energy deposited in these glasses. The changes result in breaking of bonds such as Si-O, Na-O or P-O and formation of new bonds with impurity atoms (e.g. Pb, Ga, B) in the glass matrix. The colorimetry and thermoluminescence properties of corning glass produced by gamma rays from radium were reported in as early as 1920 [1], wherein the induced bluish / violet color was found to be proportional to intensity of the radiation and also to the thickness of the glass. The utility of radio-photoluminescence (RPL) of silver activated metaphosphate glass for monitoring high doses of accidental and routine gamma radiation was reported long ago [2, 3]. Colorimetry of glasses was also reported to be useful to determine high doses of Gamma irradiation [3, 4]. Since then considerable amount of research work has been carried out to study the thermoluminescence (TL), optical absorption (OA), electron paramagnetic resonance (EPR) and optically stimulated luminescence (OSL) of different commercially available glasses in the high dose as well as low dose applications such as industrial irradiation facilities [4-10]. Recently RPL phosphate glass dosimeters have been reported to be useful in medical physics and cosmic ray dosimetry [11]. The progress made in the spectroscopic studies of glasses for radiation dosimetry in the past few decades are described in this paper.

2. Radiophotoluminescence

Radiophotoluminescent glass dosimeters are accumulation type solid state dosimeters. The material used is silver activated phosphate glass in the form of small glass rods. When this glass is exposed to radiation, stable luminescence centres (Ag^0 , Ag^{2+}) are created in silver ions. Using pulsed ultraviolet laser excitation, the orange fluorescence emitted by the glass is detected. The RPL signal is not erased during the readout and hence the dosimeter can be reanalysed many times. Accumulation of the dose is possible and can be used for registration of the lifetime dose. In an early version of low sensitive environmental glass dosimeter, layers of lead, brass and Teflon were used to flatten its over-response to low energy photons, while the round shape was chosen to reduce the dosimeter's angular dependence. RPL dosimeter has been successfully used in a large scale personal monitoring and in clinical dosimetry in Japan since 2001 [12]. In a recent version, called Piesch dosemeter sphere, a capsule with phosphate glass dosemeter (8 mm x 8 mm x 4.7 mm) is used. The response is practically independent of the direction of incident radiation. After irradiation, the dosemeter is stimulated by UV light, emitting visible fluorescence. The stored dose information is not erased during the read out, making it suitable for long-term dosimetry. The dosimeter can be regenerated by heating at 400 °C. The fading is less than 10 % after 10 years. The measuring range is from 0.4 mSv to 30 Sv. The accuracy of routine measurements is better than $\pm 15\%$. The energy dependence is $\pm 8\%$ in the range from 45 keV to 1.2 MeV. An admixture of boron makes it possible to measure thermal neutrons of nuclear reactions besides the measurement of gamma radiation [13]. In clinical applications, the glass rod dosimeter (GRD) has many advantages such as small size, ease of handling, linearity of response in the required dose region. Recently postal dose inter-comparison experiments for the absorbed dose evaluation with GRD have been done and the potential of GRD for external dosimetric audit program in radiotherapy in Korea has been established [14]. Further comparison of performance of environmental RPL dosimeters with that of TLD-200/TLD-100 showed that both are comparable for environmental dose measurements [15].

3. Colorimetry /Optical absorption

The darkening of lead alkali silicate glasses has been used for evaluation of high irradiation doses [16]. The color changes are attributed to trapping of electrons/holes at glassy network. For high gamma dose measurements and dose mapping inside gamma chamber in the range 100 Gy to 50 kGy, optical absorption of commercial glasses have been used [17, 18]. The increase in oxygen hole center and Si-bond breakage in irradiated window glasses were determined by electron spin resonance studies [10]. In a recent report, optical absorption of Nd doped yttrium aluminoborate and yttrium calcium borate glasses has been studied [9].

4. Thermoluminescence

In irradiated $\text{MgO-P}_2\text{O}_5$ glasses, the intensity of the TL peaks was found to be dependent on gamma dose (2-120 Gy) and Mg content [6]. In X-irradiated CdS doped Asahi Y-44 filter glass, linearity between TL intensity and X-ray dose was observed in the low dose region, 0.05 to 0.8 Gy [8]. Though the sensitivity of its TL was less compared to commercial TLDs, fading effect was very small. The utility of commercial colored glasses for applications such as water purification, pasteurization, sterilization and disinfestations, requiring monitoring of high gamma doses (50Gy-100 kGy, using their TL peaks, has been demonstrated by Caldas et al. [7]. The potential of using the TL peaks from fused silica glasses collected from Indian houses for accidental gamma dosimetry has been explored by Narayan et al. [18] in the dose range 1-20 Gy. Potassium aluminum phosphate (KAP) glasses doped with Mn displayed good TL sensitivity for γ -rays, X-rays and UV light [19]. A recent report on the TL from UV irradiated Dy, Li doped calcium borosilicate glass-ceramic [20] showed linear response under UV irradiation (2.1 Jcm^{-2} to 30.1 Jcm^{-2}) with good signal reproducibility. Topaz-glass composites have been reported to be useful for dose measurements down to 10cGy -1Gy [21]. In the recent years, TL and EPR properties of watch glass [22] and display windows of mobile phones [23]

have been examined for accidental dosimetry, where rapid and accurate dose measurement is essential. Especially EPR signal from irradiated watch glass was found to be linear in the dose region 2-50Gy.

5. Optical fiber dosimetry

Another area of recent interest is the optical fiber dosimetry, wherein the radiation induced defects cause an increase in the light attenuation, leading to reduction in the transmitted light intensity. This reduction is proportional to the dose delivered. Luminescence from native or radiation induced defects in the optical fibres has also been measured and correlated with dose [24]. Commercial SiO₂ optical fiber has been reported to be an excellent candidate for use in TL dosimetry of ionizing radiation [25]. This fiber showed a well defined glow peak at 230°C, with the TL response being linear in the range 0.1-3 Gy.

6. Summary

The advances made in the development of glass dosimeters during the past 2-3 decades using spectroscopic have been reviewed in this paper. Currently the thrust areas in the field of radiation dosimetry of glasses are development of novel glass phosphors exhibiting OSL for dosimetric application in medical, industrial and sensing, such as verifying of dose during radiotherapy, environmental cumulative dose measurement and remote ground water monitoring.

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