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Short communication

Intrinsic dosimetry of glass containers used to transport nuclear materials: Potential implications to the fields of waste management and nuclear forensics

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ABSTRACT

Thermoluminescence (TL) and Electron Paramagnetic Resonance (EPR) dosimetry were used to measure dose effects in borosilicate glass with time, from 10 min to ~60 days following exposure to a dose of up to 100 Gy. TL and EPR results were consistent and performed similarly, with both techniques capable of achieving an estimated limit of detection of between 0.5 and 1 Gy. Three peaks were identified in the TL glow curve at roughly 110 °C, 205 °C, and 225 °C. The intensity of the 205 °C peak was the dominant peak over the time period of this study. The stability of all of the peaks with time since irradiation increased with their corresponding temperature and no significant variation was observed in the glow curve response to a specified total dose attained at different dose rates. The intensity of the 205 °C peak decreased logarithmically with time regardless of total dose. Based upon a conservative limit of detection of 3.3 Gy, a 100 Gy dose would still be detected 2.7E3 years after exposure. Here, we introduce the concept of intrinsic dosimetry, the measurement of the total absorbed dose received by the walls of a container containing radioactive material. The foreseen advantage of intrinsic dosimetry comes from considering the measured absorbed dose received by containers in concert with the characteristics (amount, type) of the source of that dose, the radioactive material contained within the walls of the container, in order to provide enhanced information about the history of an unknown sample in question. Three hypothetical scenarios are presented to introduce this method and to illustrate how intrinsic dosimetry might benefit the fields of nuclear forensics and waste management.

1. Introduction

Glass is well known for its resistance to damage from high radiation fields (Lutze and Ewing, 1988). It is no surprise then that glass has become the storage matrix of choice for containing highly radioactive material, from samples in the laboratory to high level radioactive waste at long term disposal sites (MacNeill, 1991). This resistance to radiation is due to the noncrystalline solid/liquid structure of the material, a characteristic that also lends itself to the random incorporation of a variety of contaminants (Leverenz, 1950). While radiation does not cause dislocations within the glass per se, defects within the lattice, particularly at sites where impurities are present, do result. These defects, or electron holes, act to locally trap electrons within the lattice. The buildup (or deficiency) of charge in this manner results in electronic states with absorption bands in the visible or ultraviolet regions of the spectrum. Simple heating, accompanied by light output, typically returns the glass back to its normal electronic state. This process forms the basis of thermoluminescence (TL) dosimetry by quantifying the light output of irradiated materials during heating (Horowitz, 1984). More advanced dosimetry techniques have also been developed, such as Optically Stimulated Luminescence (OSL) (Huntley, et al., 1985) or Electron Paramagnetic Resonance (EPR) (Wertz and Bolton, 1972), which provide either greater sensitivity that TL or are nondestructive and do not require heating.

Dosimetry has been applied to the study of irradiated surfaces in the past with application towards nuclear forensics and emergency response (Aitken, 1985; Göksu, 2003; Larsson et al., 2005; Inrig et al., 2008). In these cases dosimetry was used to measure dose on surfaces, independent of the radiation source and open to the environment. However, one might envision instances where dose could be measured on the walls of glass containers housing radioactive materials. In these instances, measured dose and characteristics of the radioactive materials (i.e., amount and type) could be considered in tandem to extract greater information on samples of unknown origin. Such a tool might find application in waste management or even nuclear forensics, as glass is a common container material of choice used by radiochemists and interdictions of glass vials of trafficked nuclear materials have been documented in the past (Moody et al., 2005). By simultaneously

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considering both the accumulated dose on the walls of these containers and the type and amount of the radioactive material producing that dose, rather unique information regarding the history of an interdicted or unknown sample might be uncovered, including:

- the residence time of an unadulterated sample of radioactive material within a container;
- the amount of radioactive material that once resided in an "empty" container;
- evidence of sample splitting.

Here, we study radiation defects in glass and investigate how these potential signatures might be exploited for intrinsic dosimetry.

2. Experimental

2.1. High exposure facility

All irradiations were done at the High Exposure Facility at Pacific Northwest National Laboratory. The gamma fields produced in this facility are generated by two ^{137}Cs sources (nominal activities of 10 and 100 Ci) and two ^{60}Co sources (nominal activities of 660 and 5890 Ci) used within a collimated geometry (30 collimation angle). The calibration range extends to approximately 6 m, with the beam unobstructed until it encounters a concrete wall at a distance of approximately 8.2 m.

2.2. Sample preparation

Borosilicate glass samples were obtained from microscope slides cut into 1 cm^2 pieces maintaining the nominal thickness of the microscope slide. A total dose of between 3 and 500 Gy was delivered to the glass samples using ^{137}Cs and ^{60}Co gamma sources prior to measurement by TL and EPR. A Harshaw TL reader was used to measure the TL signals in the glass samples. The samples were protected from light exposure during and after the ionizing radiation exposure using black plastic packaging. Signal fade tests were conducted by irradiating glass samples at different total doses (3–100 Gy) and measuring the decay in the defect signal in the glass with time since irradiation, from ∼10 min to 60 days post irradiation. TL response was also studied as a function of dose rate (from 2–500 Gy/h).

Electron paramagnetic resonance (EPR) or electron spin resonance (ESR) spectroscopy, which is analogous to nuclear magnetic resonance (NMR) spectroscopy but sensitive to electron spins rather than nuclear spins, is used to study the local environments of paramagnetic chemicals (e.g., those having one or more unpaired electrons). Because the interaction of high energy radiation with materials results in the formation of free radical species, EPR spectroscopy can be used to track the formation and ultimate annihilation of free radicals. In the present work, continuous wave X band (9.4 GHz) EPR spectra were collected for borosilicate glass slides within 1 day, and approximately 46 days, after exposure to a total dose of 100 Gy. Measurements were conducted at room temperature using a Bruker ESP300E spectrometer.

3. Results

Glow curve response from irradiated borosilicate glass was found to be directly related to the amount of radiation dose received. Fig. 1 shows TL glow curves for glass samples receiving 3, 20, and 100 Gy dose, respectively, along with results from an unirradiated glass sample. Based upon a comparison of these glow curves with that of the unirradiated sample at time zero, the minimum detectable dose in borosilicate glass using TL was estimated to be around 0.5–1 Gy.

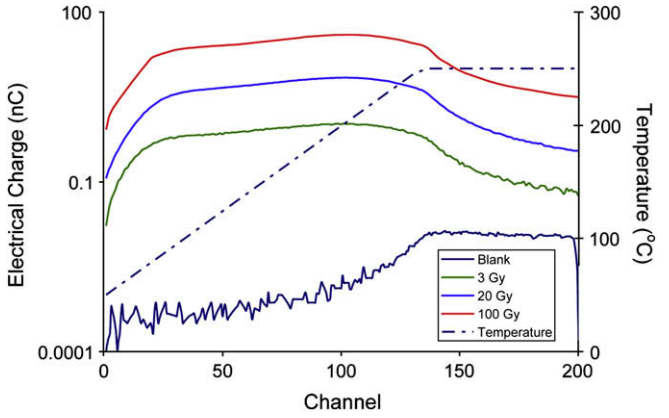

Fig. 1. TL glow curves for borosilicate glass samples immediately after receiving 0, 3, 20, and 100 Gy total absorbed dose.

Glow curve measurements were also affected by time since irradiation. Fade tests showed glow curve response in irradiated borosilicate glass predictably decreases with time, regardless of initial dose received by the glass. Fig. 2 illustrates results from fade tests for TL measurements taken on irradiated glass samples immediately following, 24 h after, and ∼60 days after receiving a dose of 20 Gy. Results (not shown) from fade tests conducted on samples receiving 3 and 100 Gy dose showed fade trends similar to those shown in Fig. 2.

Three separate peaks are evident within the glow curves in Fig. 1. A relatively unstable peak is located around 150 °C, while two more stable peaks are located at around 205 °C and 225 °C. The stability of each of the peaks increases with their glow discharge temperature, as is evident from the rather rapid decay of the 150 °C peak and the shift of the maximum glow peak towards higher temperatures with time since irradiation (see Fig. 2).

The TL signal response of the 205 °C was studied as a function of dose rates. A total dose of 20 Gy was applied to glass slides at differing dose rates, ranging from 3 Gy/h to 500 Gy/h. Results from these studies indicated dose rate had a negligible effect (within the ±3% error of the measurement) on the TL signal.

Observable in Fig. 2, peak response was found to decrease with time after irradiation. The relative fade in signal response with time was found to be predictable, regardless of the total dose applied to the glass surface. Peak heights of the 205 °C peak (relative to those

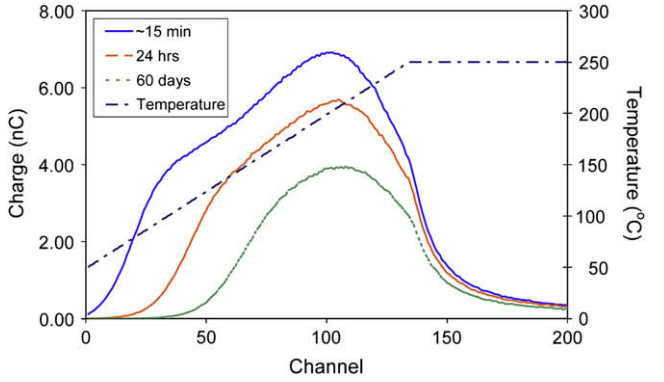

Fig. 2. Glow curve for irradiated glass samples immediately following, 24 h after, and 60 days after receiving a total dose of 20 Gy.