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Sunna 535-nm photo-fluorescent film dosimeter response to different environmental conditions

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Abstract

Evaluations on the influence of environmental variabilities on the *red* fluorescence component of the *Sunna Model* γ photo-fluorescent dosimeter^{TM} have previously been reported. This present paper describes the environmental effects on the response of the *green* fluorescence component of the same dosimeter, which is manufactured using the injection molding technique. The results presented include temperature, relative humidity, and light influences both during and after irradiation. The green fluorescence signal shows a significant dependence on irradiation temperature below room temperature at 1%/°C. Above room temperature (approximately 24–60°C), the irradiation temperature effect varies from −0.1%/°C to 1.0%/°C, depending on the absorbed dose level. For facilities with irradiation temperatures between 30°C and 60°C and absorbed dose levels above 10 kGy, irradiation temperature effects are minimal. Light-effects results indicate that the dosimeter is influenced by ultraviolet and blue wavelengths during irradiation as well as during the post-irradiation stabilization period (approximately 22 h), requiring the use of light-tight packaging. Results also show that the dosimeter exhibits negligible effects from ambient moisture during and after irradiation when in the range of 33–95% relative humidity.

Keywords: Dosimetry; Film dosimeters; Food irradiation; Radiation processing; Sterilization

1. Introduction

Becker (1969), Kaufman and Clark (1963), McLaughlin et al. (1979), and Schulman (1979) describe the theory and behavior of the M-color center, which is utilized for the lithium fluoride (LiF)-based Sunna photo-fluorescent dosimeter.[1] Murphy et al. (2003) describe how the Sunna dosimeter, upon irradiation with gamma rays and electrons and excitation by 440-nm light, emit bright fluorescence in two distinct spectral bands that peak at 535 (green) and 645 nm (red). By using broad-band optical filtration, this M-center luminescence can be measured with a visible-light photo-sensitive system (i.e., fluorimeter). This present work describes the methods and results of measurements performed on batch No. 0399–20 of the *Sunna Model* γ dosimeter (see footnote 1), which is a $1\times 3\text{-cm}^2$ polymeric film of 0.5-mm thickness produced using injection molding. Only data on the 535-nm green emission are presented, and include irradiation- and post-irradiation temperature effects, relative humidity effects, and light effects. Other publications have described the dose response and post-irradiation stabilization (Murphy et al., 2002; Miller et al., 2002; Murphy et al., 2003) associated

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with this green emission. Earlier work (Kovács et al., 1999, 2000; McLaughlin et al., 1999a, b) focused on the properties of the *red* emission from the dosimeter.

2. Experimental

All environmental variables associated with each experiment were controlled before, during and after irradiation to ensure that the data reveal the true influence of the parameter being investigated. The estimated combined uncertainty of the dosimeter system used for the experiments was evaluated so as to be able to recognize the limitations of the experiments and to properly interpret the measurement data. Except during readout, all the test dosimeters utilized were kept in light-tight packaging. Unless otherwise indicated, all the dosimeter readouts were performed in a room with lights that were covered with yellow filters to filter out UV and blue wavelengths that were found to influence the signal of freshly irradiated dosimeters (see Section 3).

2.1. Readout instrument

A Turner Designs Model TD-700 fluorimeter[2] was used for measuring the fluorescent signal from the irradiated dosimeter. This instrument consists of a 440-nm blue light source (a blue LED array and a 440-nm optical interference filter) for the excitation light, a sample chamber with green (530 nm) optical interference filters to transmit the desired fluorescence wavelength band, and a red- or green-sensitive photo-multiplier tube (PMT) to count the fluorescence photons. The bandwidth associated with the excitation and emission filters is 10 nm. The light spot area of the dosimeter that is "excited" and "read" is 0.33 cm^2 for the TD-700. The unit displays a digital readout in fluorimeter signal units (fsu), which are proportional to the PMT current.

2.2. Irradiation temperature effects

Irradiation temperature effects data were obtained from dosimeter samples irradiated with ^{60}Co gamma radiation at the National Institute of Standards and Technology (NIST) and the Pacific Northwest National Laboratory (PNNL), and from samples irradiated with electrons at a commercial E-beam radiation processing facility. The samples irradiated at NIST were sandwiched between two $3.7 \times 3.7 \times 0.5$-$\mathrm{cm}^3$ polystyrene blocks and sealed in a plastic bag. The temperature during irradiation was maintained using a temperature-controlled air shower, blowing at 0.142 m^3/min onto the sample assembly. The sample assembly was allowed to reach thermal equilibrium and remain there from 30 min prior to irradiation until 10 min or more after irradiation. The samples were not given the post-irradiation heat treatment, but were allowed to stabilize at room temperature.

The samples irradiated at PNNL were sandwiched between two $5 \times 5 \times 1$-cm^3 aluminum blocks to ensure a known constant temperature during irradiation. The dosimeter/block combination was placed in the desired calibrated temperature environment, allowed to reach equilibrium, immediately irradiated to the desired dose, then placed in a room temperature environment within 2 min after irradiation where the signal could stabilize.

The samples irradiated at the E-beam facility were sandwiched between two $7 \times 10 \times 0.5$-cm^3 polyethylene blocks to ensure a constant temperature prior to irradiation. The dosimeter/block combination was placed in the desired calibrated temperature environment, allowed to reach equilibrium, immediately placed on the conveyor system and irradiated to the desired dose (5 min duration from conveyor start to irradiation), then placed back in the temperature environment for the desired storage time before the dosimeters were read out or given the stabilizing post-irradiation heat treatment. Although the adiabatic heating of the dosimeters was measurable, it was not accounted for. For these tests the heat treatment (68°C for 20 min duration) was started within approximately 30 min after irradiation.

2.3. Light effects

Light effects testing was performed on the dosimeter at three stages: prior to irradiation; freshly irradiated; and after a period in which the dosimeter was allowed to stabilize. The experiment for the irradiated dosimeters was performed in a room brightly lit with bare fluorescent lights (*cool white* type without any covering) at approximately 1.5-m distance in order to model a worst-case industrial laboratory situation. For each stage the groups of dosimeters were divided into four sets: set ♯1 being kept in a light-tight package; set ♯2 being kept under a UV-absorbing filter blocking wavelengths of <400 nm; set ♯3 being kept under a UV- and blue-absorbing filter with yellow appearance and blocking at wavelengths <500 nm; and set ♯4 being bare with no covering.

The testing for the non-irradiated dosimeters was performed for both white fluorescent laboratory illumination and sunlight through a window. In this test, the lab light fixtures had the typical plastic (poly carbonate) shades, which are effective at blocking the UV wavelengths (<400 nm).

Additional testing was performed to determine the influence of leaving dosimeters (sets of both freshly irradiated and fully stabilized dosimeters) exposed to the fluorimeter excitation light (440 nm).

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[2] Turner Designs, Sunnyvale, CA.

2.4. Relative humidity effects

Tests of the effect of relative humidity (r.h.) were performed at NIST. For the test, dosimeters were first placed in open foil pouches and held in dessicators with stable moisture concentrations at 33%, 57%, and 94% r.h. After 40 h the pouches were sealed and irradiated to various doses of 0.4 and 2.0 kGy at a dose rate of 6.5 kGy/h. The temperature of the gamma cell irradiation chamber during irradiation was controlled with a 0.142 m^3/min air shower blowing down on the samples. At 30 min after irradiation, some samples were heat treated at 69°C for a duration of 30 min to determine any influence on humidity effects, and the remaining samples were allowed to stabilize naturally.

Tests were performed at PNNL to determine the effect of placing the dosimeters in environments near 100% r.h. and room temperature prior to irradiation. For this test, a large set of dosimeters were submerged in water at room temperature and, at various times over a period of 25 days, sets of three were removed and irradiated to 2 kGy along with dosimeters that had been left in a normal laboratory environment of 30–50% r.h. and 22°C. The net responses of these dosimeters were then compared to one another to determine the water diffusion rate and the amplitude of the effects. No heat treatment was performed.

An additional test was performed at PNNL to measure the water diffusion rate and resulting change in dosimeter response for newly manufactured dosimeters placed in a normal laboratory environment of 30–50% r.h. and 22°C. A change in response due to absorption of water from the air was expected because the manufacturing process involves heating the resin at very high temperatures, resulting in new dosimeters with very little water content.

2.5. Storage temperature influence on short-term post-irradiation stability

To determine the influence of dosimeter storage temperature on the short-term stability of the dosimeter's fluorescence signals, a short-term growth study was performed at two storage temperatures, 0°C and 22°C, using dosimeters irradiated to 2 kGy. The procedure involved irradiating a set of approximately 20 dosimeters to 2 kGy using ^{60}Co gamma radiation, immediately placing them in the desired temperature environment, then periodically reading one dosimeter at a time until the readings were stable.

3. Results and discussion

3.1. Irradiation temperature effects

Figs. 1–3 show the irradiation temperature response data for various dose and dose rate levels using ^{60}Co gamma rays (7.5 and 13 kGy/h), and 4.5 and 10 MeV E-beam radiation (on the order of thousands of kGy/h). In general, the data show that: (1) for the doses measured, the effect below room temperature is essentially dose independent and is approximately +1%/°C (relative to room temperature); (2) the effect becomes dose dependent at temperatures above room temperature; (3) for dose levels of approximately

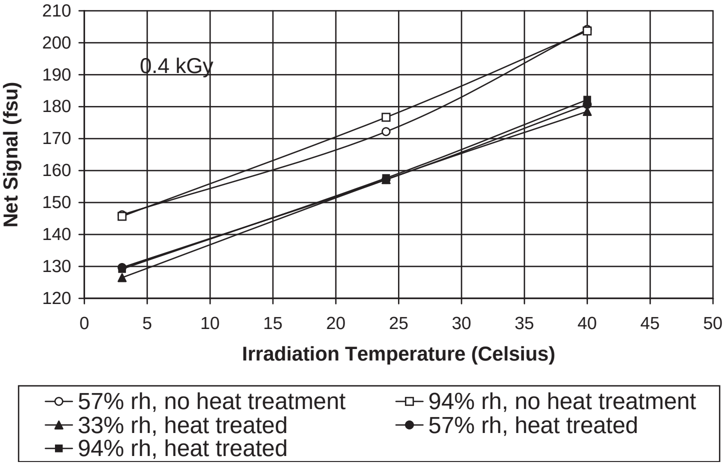

Fig. 1. Influence of irradiation temperature on the *Sunna Model* γ dosimeter's green emission at a absorbed dose of 0.4 kGy using ^{60}Co gamma radiation at 6.5 kGy/h. Results are shown for heat-treated (performed 10 min after irradiation) and non-treated (read out 24 h after irradiation) dosimeters.

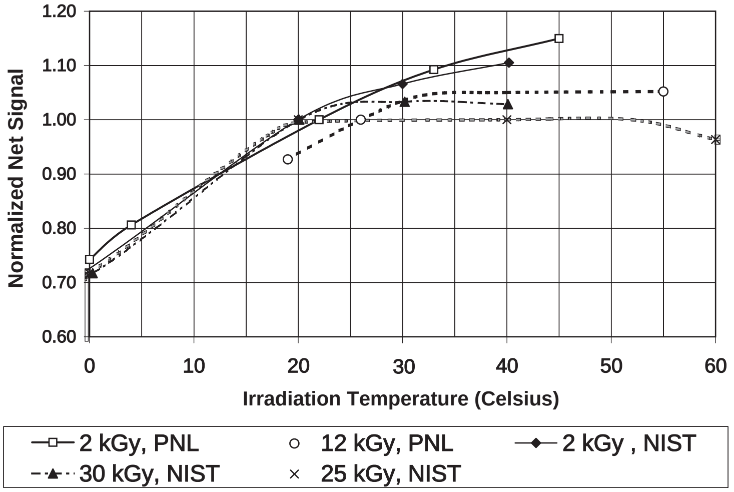

Fig. 2. Influence of irradiation temperature on the *Sunna Model* γ dosimeter's green emission at absorbed doses between 2 and 30 kGy using ^{60}Co gamma radiation at 7 kGy/h (NIST) and 12 kGy/h (PNNL). No post-irradiation heat treatment was performed. Thresholds above room temperature for dashed curves are estimates based on a majority of data.

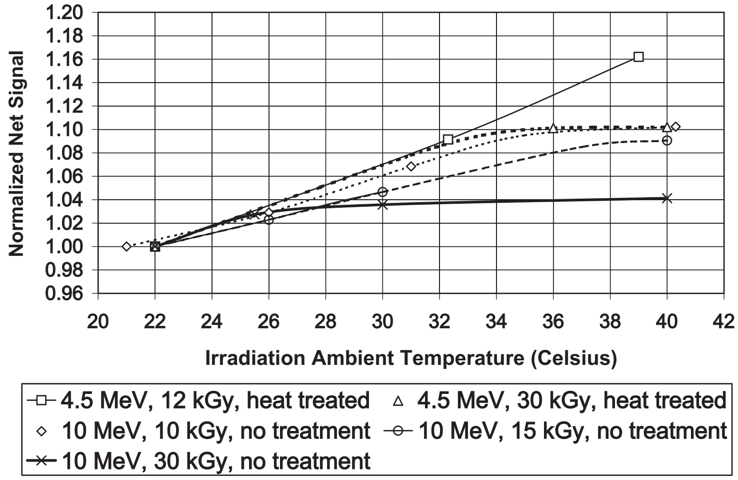

Fig. 3. Influence of irradiation temperature on the *Sunna Model* γ dosimeter's green emission at absorbed doses between 12 and 30 kGy using 4.5 and 10 MeV E-beam. Heat treatment was performed on the 4.5-MeV films 30 min after irradiation. Heat treatment was not performed on the 10-MeV films. Thresholds above room temperature for dashed curves are estimates based on a majority of data.

400 Gy the effect above room temperature stays at +1%/°C; (4) for dose levels of approximately 2 kGy the effect above room temperature is approximately +0.5%/°C; (5) for gamma dose levels of approximately 10 kGy the effect is 0.0 to +0.1%/°C for the temperature range 29–60°C; (6) for gamma dose levels of approximately 15–30 kGy the effect is −0.1 to +0.1%/°C for the temperature range 25–60°C; (7) in addition to the inflection point at about 20°C, there appears to be another dose-dependent inflection point that is located at about 50°C for low doses and shifts to lower temperatures as the dose increases, resulting in the flat curves above 25°C for 30 kGy levels; (8) when E-beam is used, the upper inflection point on the curve is at approximately 35°C for sterilization dose levels; (9) a post-irradiation heat treatment protocol can increase the temperature coefficient and; (10) elevated post-irradiation temperatures

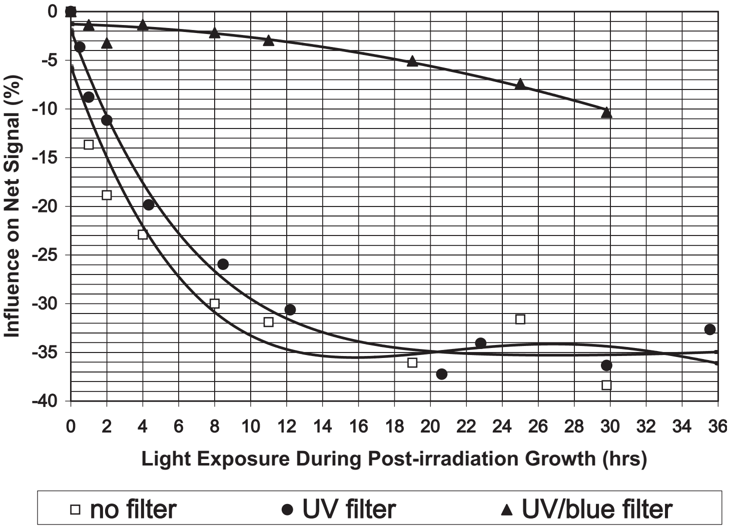

Fig. 4. Percentage change of the dosimeter signal for increasing duration of exposure to the laboratory illumination—with and without various filters covering the light fixtures.

can increase the temperature coefficient. For a detailed report on the effectiveness of post-irradiation heat treatment for the Sunna dosimeter, see Murphy et al. (2003).

3.2. Light effects

Light effects data are shown in Fig. 4 for freshly irradiated (2 kGy) dosimeters. The figure indicates the direct percentage change of the dosimeter signal for increasing duration of exposure to the laboratory illumination—with and without the various filters covering the light fixtures. The data in these figures show that, in the first 60 min after irradiation: (1) the formation of green color centers is affected by a maximum of −0.3%/min by unfiltered light, −0.1%/min by UV-filtered light, and −0.07%/min by UV- and blue-filtered light and; (2) the light effect decreases as the color centers approached their stabilization points (i.e., approximately 22 h), becoming negligible after stabilization. Therefore, freshly irradiated dosimeters can be exposed to typical laboratory fluorescent lighting for approximately 3, 10, and 15 min when no filters, UV filters, and UV/blue filters are used, respectively, before effects approach 1%. If a post-irradiation heat treatment is performed (see Murphy et al., 2003), such light effects are not of concern because dosimeters are thereby stabilized.

In a separate study for non-irradiated (control) dosimeters, typical laboratory light (with the typical plastic shade) caused fading of the green background fluorescence by 1.4%/day, and diffuse sunlight through a north window faded the green background fluorescence by 0.17%/h.

All these light effects can easily be solved by keeping the dosimeters in light-tight packaging, and covering lights at the dosimeter handling and readout stations with the appropriate filters.

The influence of continuous exposure to the fluorimeter excitation light (442-nm LEDs) on the dosimeter's green emission is shown in Fig. 5 for dosimeters irradiated to 2 kGy. Results are shown for both newly irradiated and stabilized dosimeters. The data indicate that the influence on the green emission within minutes after irradiation is approximately +2.5%/min of exposure. Therefore, for newly irradiated dosimeters the influence over a typical readout duration of 7 s can as much as 0.3%, which for most applications would not be a concern. The influence on the green emission after the associated color centers are well stabilized (i.e., after $t = 22$ h or after heat treatment is performed) is effectively zero.

3.3. Humidity effects

Figs. 6 and 7 show the results of humidity testing at NIST for dosimeters stored at ambient humidities ranging from 33% to 95% r.h. (from 40 h prior to irradiation until 0.5 h after irradiation) and irradiated at temperatures ranging from 3°C to 40°C. Dose levels of 0.4 and 2 kGy were delivered for combinations of humidities and temperatures. The results indicate that for all scenarios the effect was <2.5% relative to 57% r.h., with the exception of 2 kGy and 95% r.h., which

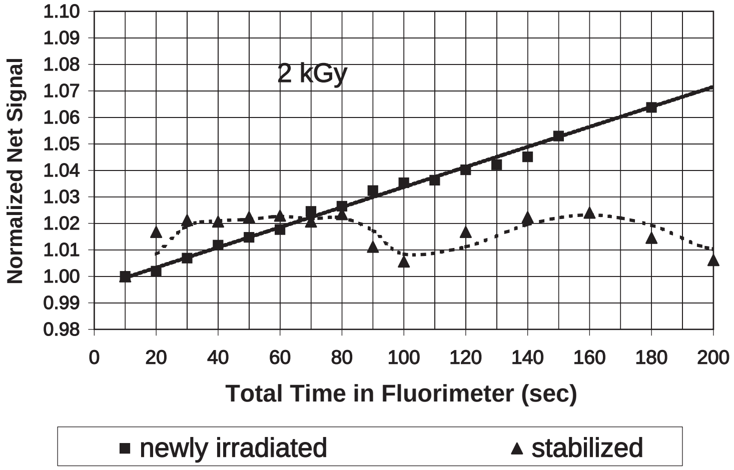

Fig. 5. Effect of fluorimeter excitation light (i.e., 440 nm) on the *Sunna Model γ* dosimeter's green emission during readout process. Data indicates that if newly irradiated dosimeters are left in the fluorimeter readout chamber, the signal is affected by as much as +2.3%/min. A typical readout time is 7–10 s. The effects of readout are negligible after the associated color centers complete their inherent stabilization process at 22 h.

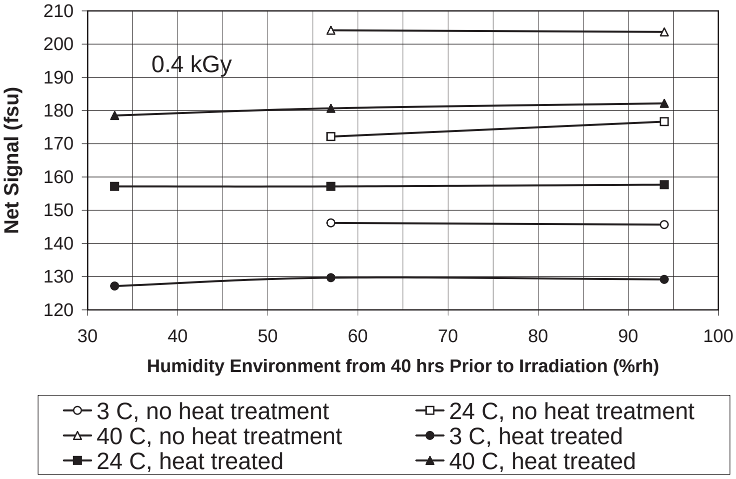

Fig. 6. Influence of ambient relative humidity on the *Sunna Model γ* dosimeter's green emission at an absorbed dose level of 0.4 kGy (using ^{60}Co a gamma radiation at 6.5 kGy/h) and at various irradiation temperatures. Results are shown for heat-treated (performed 10 min after irradiation) and non-treated (read out 24 h after irradiation) dosimeters.

has a maximum effect of –4.5%. Because of the slow water uptake for polyethylene, it is expected that these effects would be significantly lower for dosimeters stored in a humidity-controlled environment (e.g., a refrigerator) until hours before irradiation.

Fig. 8 shows the change in dosimeter response versus the time duration submerged in room temperature water. The figure indicates that it takes approximately 24 h for the water to be absorbed into the dosimeter and influence the response. This response maximizes at approximately +4% at 24 h, then appears to stabilize at approximately +3% out to 25 days.

Fig. 9 shows the change in response of a newly manufactured dosimeter batch over a period of 166 days. The change is considered to be due to absorption of water from the air rather than some other type of batch "aging" effect. The +4% maximum

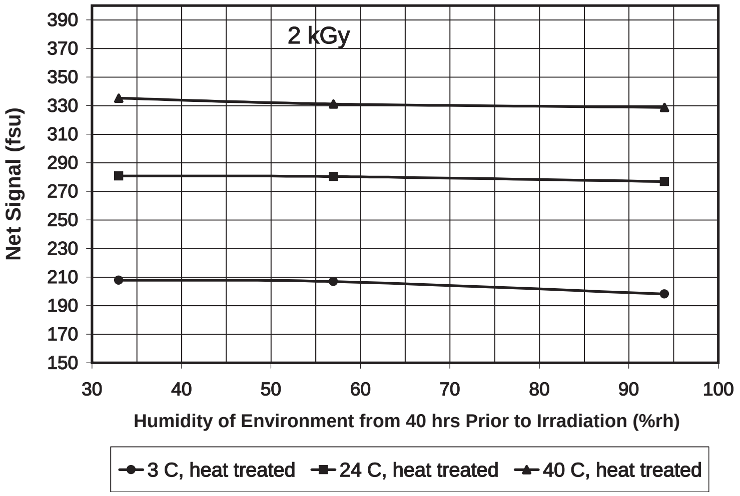

Fig. 7. Influence of ambient relative humidity on the *Sunna Model* γ dosimeter's green emission at an absorbed dose level of 2 kGy (using ^{60}Co gamma radiation at 6.5 kGy/h) and at various irradiation temperatures. Results are shown for heat-treated dosimeters (performed 10 min after irradiation). Absorbed dose level is 2 kGy (using ^{60}Co gamma radiation at 13 kGy/h).

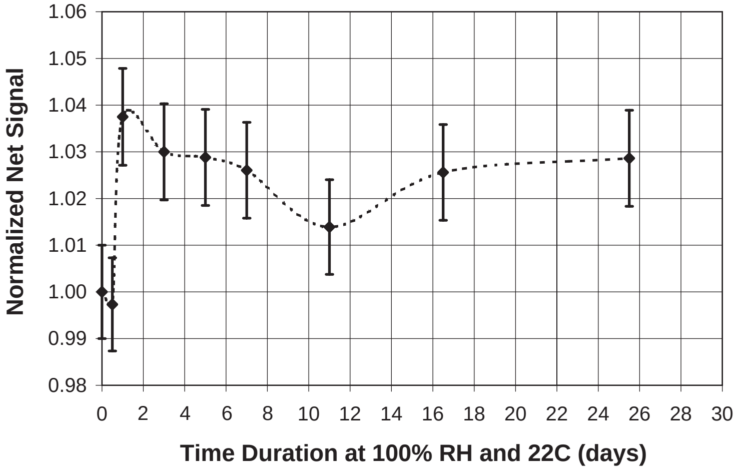

Fig. 8. Dose response versus time duration in 100% humidity environment (at 22°C) for the *Sunna Model* γ dosimeter's green emission at an absorbed dose level of 2 kGy (using ^{60}Co gamma radiation at 13 kGy/h). Response is influenced starting at about 24 h. No heat treatment was performed.

effect shown in the figure, and which appears to stabilize at about 100 days, is consistent with the maximum +4% humidity effect observed in Fig. 8. These results suggest that dosimeters should not be used until approximately 100 days after production (stored under normal laboratory conditions). If they are to be used before this time, they should be hermetically sealed under controlled conditions of humidity.

3.4. Storage temperature influence on short-term stability

Fig. 10 shows the influence of irradiation temperature and post-irradiation storage temperature on the short-term post-irradiation growth of signal from dosimeters irradiated to 2 kGy. The data show that decreasing the irradiation and storage temperature to 0°C increases the stabilization time from 22 h to 4 days for the green emission.

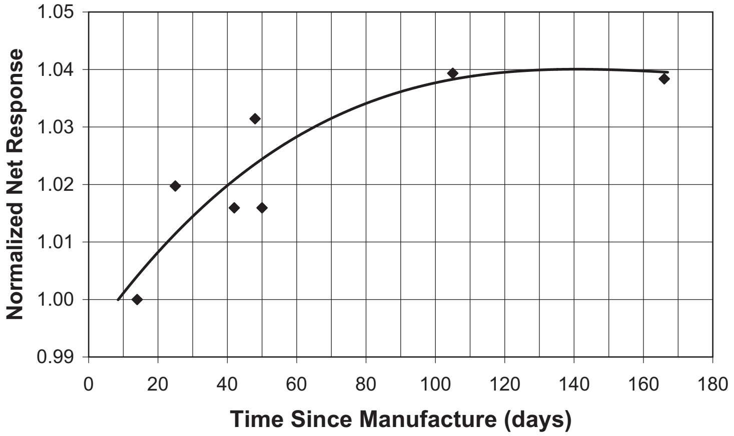

Fig. 9. Change in dose response over time for newly manufactured *Sunna Model* γ dosimeter's in normal laboratory conditions of 30–50% r.h. and 22°C. The effect is considered to be due to absorption of water from the air rather than some other type of "aging" effect. Absorbed dose level is 5 kGy (using ^{60}Co gamma radiation at 13 kGy/h). No heat treatment was performed.

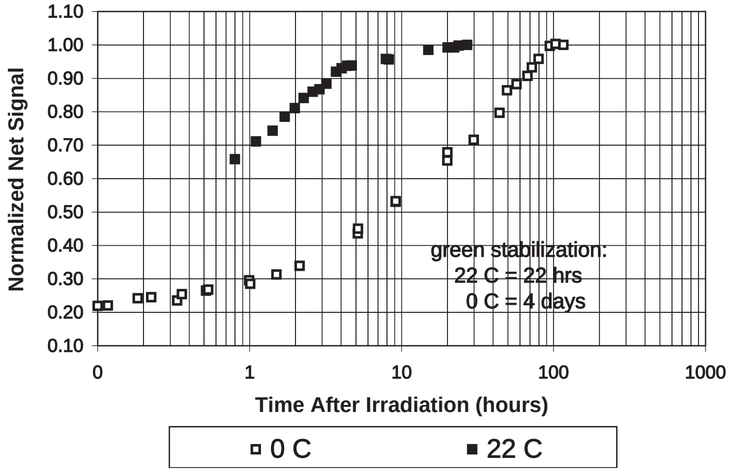

Fig. 10. Influence of irradiation- and post-irradiation storage temperatures on the *Sunna Model* γ dosimeter's post-irradiation stabilization time. Results are shown for an absorbed dose level of 2 kGy (using ^{60}Co gamma radiation at 14 kGy/h) and at irradiation and storage temperatures of 0°C and 22°C.

4. Conclusions

Like most routine high-dose dosimeter systems, the *Sunna Model* γ dosimeter is influenced by certain environmental conditions; however, the all-around performance of the green emission (Murphy et al., 2003) makes it appealing for several applications. The environmental effects on the dosimeter are: (1) UV and blue light effects for newly irradiated dosimeters, (2) irradiation temperature effects, and (3) storage temperature effects. The irradiation temperature effects for the green emission are on the order of +1%/°C for both food and sterilization dose levels at refrigerated temperatures, +0.5%/°C to +1.0%/°C for food dose level irradiations at room temperature or above, and approximately −0.1%/°C to +0.2%/°C for sterilization dose levels at temperatures between 30°C and 60°C. The light effects can easily be controlled by using light-tight packaging and handling bare dosimeters under properly filtered illumination. The irradiation temperature effect can be accounted for by calibrating under temperature conditions of eventual use.

The storage temperature influences the time period needed for the dosimeter signal to stabilize—the colder the storage temperature, the longer for stabilization to occur.

The data covered in this paper indicate that a potential advantage of the green-emission Sunna film

(*Model γ*) over radiochromic film (Abdel-fattah and Miller, 1996) and PMMA (Barrett, 1982) dosimeter systems are a slow water uptake combined with a minimal humidity influence up to 95% r.h., even when combined with high temperatures, allowing use in high humidity environments and dose mapping of wet product using bare dosimeters.

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