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ENERGY RESPONSE OF GR-200A THERMOLUMINESCENCE DOSEMETERS TO ^{60}CO AND TO MONOENERGETIC SYNCHROTRON RADIATION IN THE ENERGY RANGE 28–40 KEV

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The response of LiF:Mg,Cu,P thermoluminescence dosemeters (type GR-200A) to monoenergetic radiation of energy 28, 35, 38 and 40 keV was evaluated with respect to irradiation with a calibrated ^{60}Co gamma-ray source. High-precision measurements of the relative air kerma response performed at the SYRMEP beamline of the ELETTRA synchrotron radiation facility (Trieste, Italy) showed a significant deviation of the average response to low-energy X-rays from that to ^{60}Co, with an over-response from 6 % (at 28 keV) to 22 % (at 40 keV). These data are not consistent with literature data for these dosemeters, where model predictions gave deviation from unity of the relative air kerma response of about 10 %. The authors conclude for the need of additional determinations of the low-energy relative response of GR-200A dosemeters, covering a wider range of monoenergetic energies sampled at a fine energy step, as planned in future experiments by their group at the ELETTRA facility.

INTRODUCTION

Thermoluminescence dosimetry is one of the most important dosimetric techniques with applications in areas such as personnel, environmental and clinical dosimetry. Nakajima et al.[1] were the first to produce, in powder form, LiF doped with Mg, Cu and P impurities. This material passed through a gradual evolution that led to a dosimetric system with good tissue-equivalence, high-sensitivity, linear dose response, low fading rate and low residual signal[2]. Presently, LiF:Mg,Cu,P dosemeters are produced by various manufacturers such as Solid Dosimetric Detector & Method Laboratory in Beijing, China (type GR-200), Nemoto in Japan (type NTL-500), Institute of Nuclear Physics in Poland (type MCP-N) and by Thermo Scientific in USA (type TLD-100H, TLD-600H,TLD-700H)[3].

Many factors affect the energy response of these dosemeters such as type and concentration of dopants and the spatial distribution of the energy deposition events[4]. For X-rays, the photon ionisation density, expressed by mean lineal energy or by linear energy transfer (LET), varies with photon energy because of changes in secondary electron spectra. LiF:Mg,Cu,P shows an anomalous response to low-energy X-rays (,100 keV), due to its high dependence on LET: small changes in this quantity (due to the gradual transition of X-ray interaction from predominantly photoelectric effect to Compton scattering[5]) lead to an observable change in the response.

Figure 1a and b show, respectively, the mass attenuation and mass energy absorption coefficient of LiF:Mg,Cu,P in the energy range 10–50 keV and 27–41 keV, respectively. These coefficients were calculated using the software XMudat(6) for the following relative elemental weight composition: 72 % F, 26 % Li,0.2%Mg,0.7%Pand0.1%Cu.Figure1ashows that Compton interaction in LiF:Mg,Cu,P is prevalent over photoelectric absorption at photon energies greater than 31 keV. This occurrence might determine a spectral variation of the response of LiF:Mg,Cu,P TLD dosemeters at energies across 31 keV.However, the observed data in the low-energy range for relative energy response of LiF:Mg,Cu,P relative to air, normalized to the response at a ^{60}Co source relative to air (this ratio being called 'relative response' in the following), are not explained simply in terms of mass absorption coefficients. Based upon cross-sectional data of ref. (7), Figure 1c shows the ratio of mass absorption coefficient of LiF:Mg,Cu,P to air, in the energy range 27 – 41 keV (μ'_E), normal- ized to the corresponding value at the reference energy of 1.25 MeV ($\mu'_{^{60}Co}$):

$$\frac{\mu'_E}{\mu'_{^{60}\mathrm{Co}}} = \frac{[(\mu_{en}/\rho)^{\mathrm{LiF}}_{\mathrm{air}}]_E}{[(\mu_{en}/\rho)^{\mathrm{LiF}}_{\mathrm{air}}]_{^{60}\mathrm{Co}}}. \qquad (1)$$

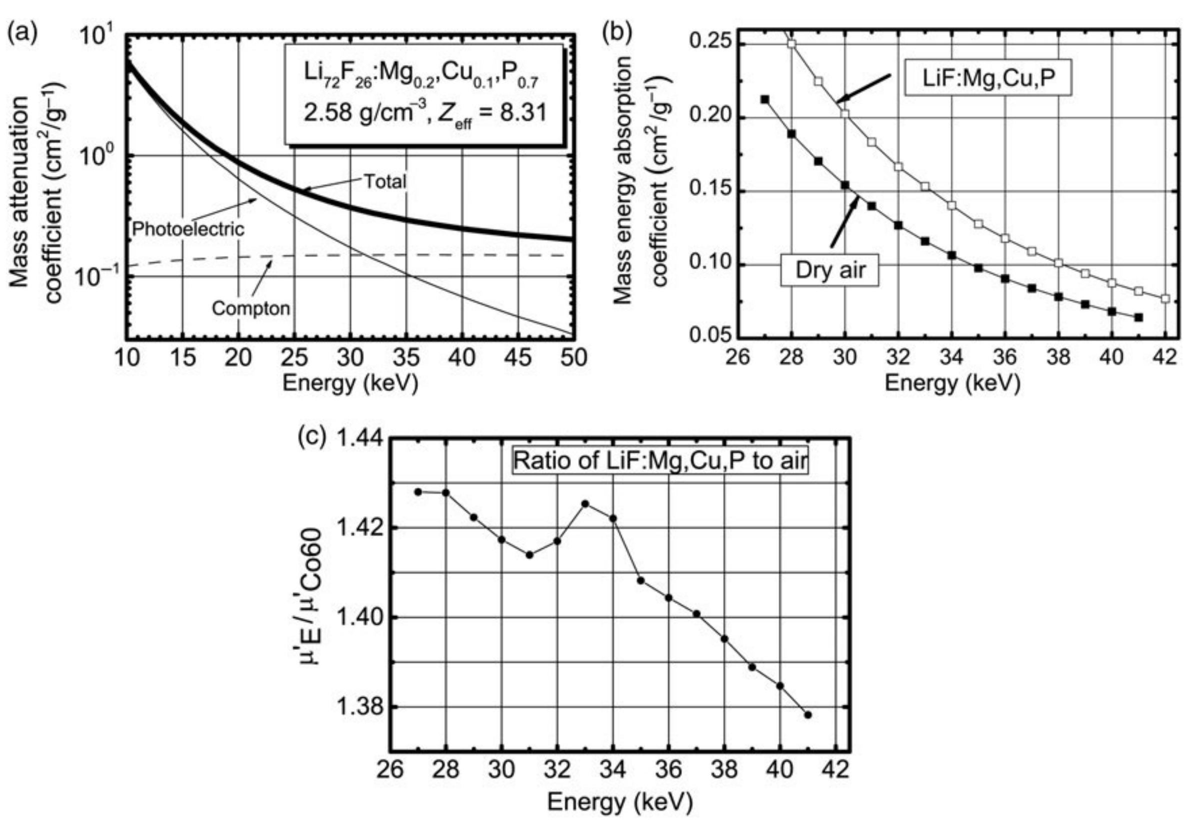

Figure 1. (a) Mass attenuation coefficients and (b) mass energy absorption coefficients for LiF:Mg,Cu,P (with composition given in the text) and air, and (c) ratio of mass absorption coefficients for LiF:Mg,Cu,P (m^0_E) and air, in the energy range 27–41 keV, normalized to corresponding value at the reference energy of 1.25 MeV (m^0_{Co60}). Data have been calculated with the software XMuDat[6] with cross-sectional data from ref. (7).

This figure shows a slight (1.3 % above the baseline trend) spectral feature in the energy range 28 – 40 keV, which might reflect into an energy-dependent response of LiF:Mg,Cu,P TLDs in this range. While the accuracy of available attenuation data for the materials in Figure 1c are not such to exclude that interpolation inaccuracies may be present; however, literature data give a more complex scenario for the energy response of such dosemeters relative to high-energy X-rays (MeV) or gamma rays (^{60}Co), as explained below. Kron et al.[8] studied the energy response of various radiation detectors (including LiF:Mg,Ti and LiF:Mg,Cu,P TLD dosemeters) to monoenergetic and polyenergetic photons and proved that irradiation with monoenergetic X-ray beams permits to avoid the compounding effect of the spectral distribution of X-rays from conventional sources. They showed an enhancement up to about 50 % of the LiF:Mg,Ti response to monoenergetic photon beams in the energy range 10–100 keV, with respect to 6 MV spec- tral X-rays from a medical linear accelerator. On the other hand, the response for LiF:Mg,Cu,P was close to the 6-MV response, considering the experimental uncertainties, with some over-response up to about 20 % at energies around 25 keV (Figure 2).

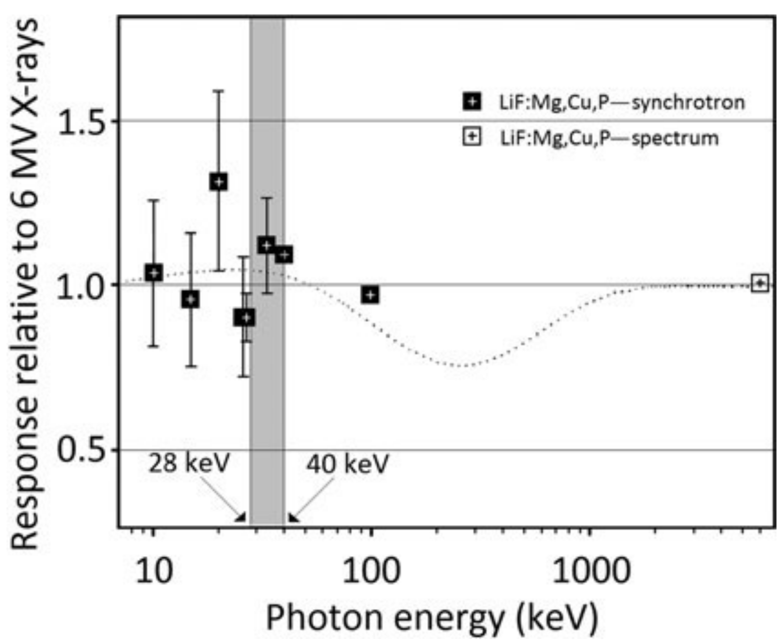

Figure2. Response of LiF:Mg,Cu,P TLDs to monoenergetic synchrotron radiation, relative to 6 MV spectral X-rays from a medical linear accelerator (data points). The dotted line represents the model prediction. The shaded area indicates the region of photon energies investigated in the present study (adapted from Figure 8 of ref. (8)).

Using monoenergetic synchrotron radiation (SR), Duggan et al.[9] irradiated LiF:Mg,Cu,P TLDs from two manufacturers (MCP-N, Poland; GR-200 series, China) and LiF:Mg,Ti (GR-100, China) in the range 10 – 26 keV. Their results confirmed both qualitatively and quantitatively those of Kron et al.[8], showing an increased response up to a factor of 1.5 for LiF:Mg,Ti and a response close to unity for LiF:Mg,Cu,P, relative to 6-MV irradiation. Their experimental findings confirmed the model prediction of Kron et al.[8]. However, considering the observed variations in the mean value of the TLD response in the low energy range around 30 keV and the large experimental un- certainties, no clear indication can be drawn about the spectral feature in the photon energy range 28 – 40 keV of interest for the present study (e.g. as reported in Figure 1c). Hence, the issue of the relative spectral response of TLDs (and in particular of LiF:Mg,Cu,P) in the low energy range below about 100 keV deserves further investigation, with particular interest in the spectral range between 31 and 35 keV.

In the present work the authors studied the energy response of LiF:Mg,Cu,P (GR-200A) dosemeters using monoenergetic beams in the energy range 28 – 40 keV and a reference irradiation to gamma rays from a calibrated ^{60}Co source. The aim was to evaluate experimentally the relative (to ^{60}Co) response of those dosemeters at various monoenergetic photon energies in that range, at an SR source. The interest was motivated by the ongoing project SYRMA-CT at the ELETTRA SYRMEP beamline for in vivo breast tomography at energies approaching 40 keV, where TLDs were planned to be used for beam dosimetry and phantom dosimetry.

MATERIALS AND METHODS

In this work, cylindrical LiF:Mg,Cu,P TLDs (type GR-200A, Solid Dosimetric Detector & Method Laboratory, Beijing, China) of 4.5-mm diameter and 0.8-mm thickness were investigated. Annealing pro- cedure consisted in the heating of the dosemeters at a temperature of 2358C for 20 min. Reading was per- formed with the Thermo Scientific™ Harshaw TLD™ Model 4500 reader that heat the sample to a temperature of 2408C with a linear ramp rate of 88C/s, with no pre-heat step.

GR-200A TLDs were calibrated individually in agamma-rays field from a ^{60}Co source with known cali-brated activity. TLDs were irradiated at a distancefrom the source of 1 m at 5.07 mm depth in a PMMAphantom that provided charged particle equilibrium. Dosemeters were exposed to four different levels of ^{SR} air kerma, obtained by varying the exposure time. For each TLD, experimental data were plotted as air kerma in mGy vs. corresponding reading in nC. A calibration curve was obtained by a linear fit to the data points.

Monoenergetic X-ray beams from SR were used to investigate the energy response of GR-200A. The irradiations were performed at the beam energies of 28, 35, 38, 40 keV at the SYRMEP beam line of the ELETTRA SR facility (Trieste, Italy). The size of the beam was fixed at 170 3.94 mm^2 (H V). TLDs were irradiated free in air with the circular face in a plane transverse to the beam axis. Due to the small vertical size of the beam, a vertical scanning of 20 mm at constant speed was necessary to ensure a uniform irradiation. A calibrated ionization chamber (Radcal 10X6 – 3CT with the Radcal Accu-Pro digital multimeter) was placed free in air near the TLDs to measure the air kerma during irradiations. The value of air kerma measured by the ionization chamber, K_{SR}^{air} , was corrected for the dimension of the scanned area. This value was obtained with an overall accur- acy of 5 % (as specified by the manufacturer). The air kerma rate varied from a maximum of 147 mGy min^{-1}at the beam energy of 28keV, to a minimum of 3 mGy min^{-1} at 40 keV. At each energy, TLDs were irradiated in groups of five at four different values of air kerma.

The air kerma relative response of each TLD, R_n, was calculated as

$$R_n = \left(\frac{K^{\text{air}}_{^{60}\text{Co}}}{K^{\text{air}}_{\text{SR}}} \right)_n \quad (2)$$

Where K^{air}_{60Co} was calculated usign TLD reading (T_n) and calibration parameters obtained from the fit:

$$(K^{\text{air}}_{^{60}\text{Co}})_n = i_n + s_n \cdot T_n \quad (3)$$

where i_n and s_n are the intercept and the slope of the calibration curve, respectively. $(K^{air}_{60Co})_n$ represents the air kerma in the ^{60}Co reference beam that would give the same reader output T_n. Relative uncertainty in this value was estimated by propagating the uncertainties on calibration parameters: overall, they were in the range from 0.4 to 2.5 %. $(K_{SR}^{air})_n$ is the air kerma in the SR beam, measured with the ionization chamber, mea- sured with an overall accuracy of 5 % (as specified by the manufacturer).

The uncertainty on R_n, $\sigma(R_n)$, was estimated by propagating the uncertainties on $(K^{air}_{60Co})_n$ and on measured air kerma $(K_{SR}^{air})_n$:

$$\sigma^2_{R_n} = \left(\frac{1}{K^{\text{air}}_{\text{SR}}} \right)^2 \cdot \sigma^2(i_n) + \left(\frac{T_n}{K^{\text{air}}_{\text{SR}}} \right)^2 \cdot \sigma^2(s_n) + \left(\frac{K^{\text{air}}_{^{60}\text{Co}}}{(K^{\text{air}}_{\text{SR}})^2} \right)^2 \cdot \sigma^2(K^{\text{air}}_{\text{SR}}) + \left(\frac{s_n}{K^{\text{air}}_{\text{SR}}} \right)^2 \cdot \sigma^2(T_n) \quad (4)$$

where $\sigma(i_n)$ and $\sigma(s_n)$ are the uncertainties on intercept and slope of the calibration curve of the nth dosemeter, respectively, calculated using the least mean square method. $\sigma(K_{SR}^{air})$is the uncertainty on K^{air}_{SR} estimated as 5 % of the measured value and $\sigma(T_n)$ was evaluated by repeated measurements with a single TLD and it is about 2 %.

At each energy, the overall air kerma response (R_E) was calculated as the weighted average, with weights w_n and weighted standard deviation σ_R, of the air kerma response, R_n, of TLDs exposed at that energy:

$$R_E = \frac{\sum_n w_n \cdot R_n}{\sum_n w_n}; \quad w_n = \frac{1}{[\sigma(R_n)]^2}; \quad \sigma_{R_E} = \frac{1}{\sqrt{\sum_n w_n}} \tag{5}$$

The weighted standard deviation of the mean, σ_{RE} (calculated out of n = 20 measurements at 28 keV and 35 keV and n = 15 at 38 keV and 40 keV, respectively) was between 1.2 and 1.4 % of the weighted mean value, R_E. The expected trend of the relative response was cal-culated as

$$R_{E,\mathrm{corr}} = \frac{\mu'_E}{\mu'_{^{60}\mathrm{Co}}} \cdot \frac{F_C^E}{F_C^{^{60}\mathrm{Co}}} \cdot \frac{F_L^E}{F_L^{^{60}\mathrm{Co}}} \eta \tag{6}$$

which takes into account light self-absorption (the term F_L^E), attenuation of X-rays in the TLD (F_c^E) and the relative TLD efficiency (η). The values of η used were those (relative to ^{137}Cs irradiation) as a function of E, extracted from Figure 2 in ref. (10). The attenu- ation correction factor at energy E, F_c^E, was defined as

$$F_C^E = \frac{1 - \mathrm{e}^{-(\mu_{\mathrm{en}}/\rho)\rho \mathrm{d}}}{(\mu_{\mathrm{en}}/\rho)\rho \mathrm{d}} \tag{7}$$

where d is the thickness of the dosemeters and r and μ_{en}/r are the density and the mass energy absorption coefficient of LiF:Mg,Cu,P, respectively, for photons of energy E, calculated with the software XMuDat[6] with cross section data from ref. (7). The light correction factor at energy E, F_L^E, has been calculated according to the following expression:

$$F_L^E = \frac{[1 - \mathrm{e}^{(-(\mu_{\mathrm{en}}/\rho)+\cdot\mu_l)\rho \mathrm{d}}](\mu_{\mathrm{en}}/\rho)\rho}{[1 - \mathrm{e}^{(-\mu_{\mathrm{en}}/\rho)\rho \mathrm{d}}]((\mu_{\mathrm{en}}/\rho) + \mu_l)\rho} \tag{8}$$

where μ_1 is the light absorption coefficient.

RESULTS

Figure 3 shows the air kerma response of GR-200A TLDs to monoenergetic SR at 28, 35, 38 and 40 keV, relative to the response to ^{60}Co gamma rays. This figure presents a comparison between data in this work and data from Kron et al.[(8)] and Bakshi et al.[(11)], and data from Gonzalez et al.[(12)]. It is to be noted that normalization is for 6-MV X-rays in ref. (8), and for ^{60}Co gamma rays in refs. (10, 11) and in the present study. In the data of this work it is possible to note a minimum of the relative response at 35 keV. The discrepancy between the response for ^{60}Co gamma rays and that in the range 28 – 40 keV (from 6 to 22 %) is greater than the uncertainties on the corre- sponding data points (about 2 %). Figure 3 (continuous line) shows the trend of the relative response, calculated according to Equation (6).

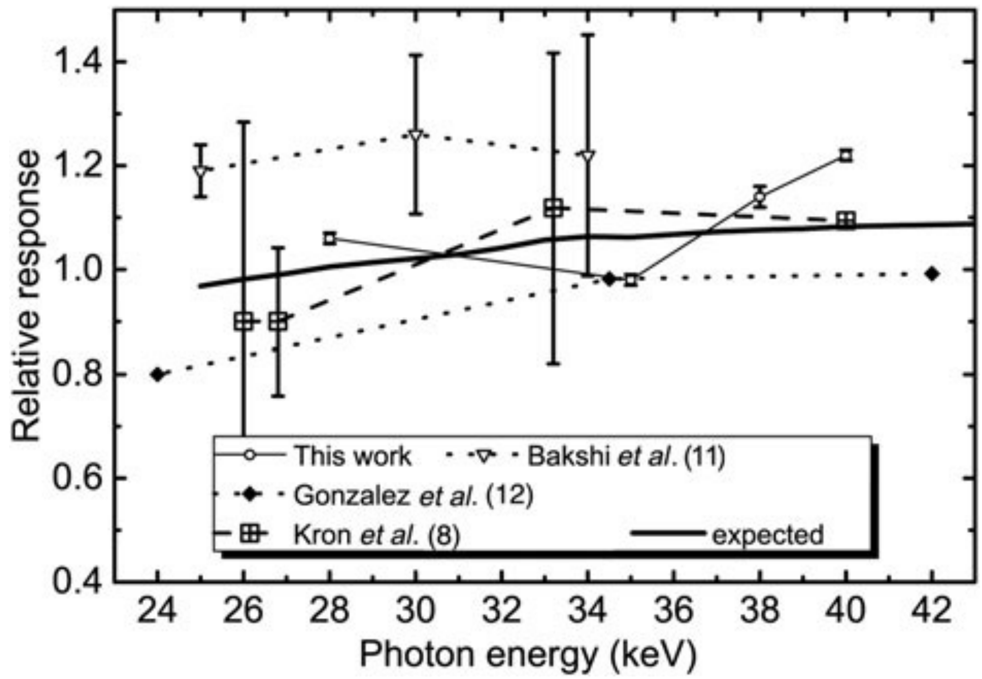

Figure 3. Comparison of energy response of LiF:Mg,Cu,P reported in this work and energy response reported in refs. (8, 11, 12). Note that in ref. (8) and in this work, LiF:Mg,Cu,P TLDs produced in China as GR-200A were studied. In ref. (11) the authors investigated the LiF:Mg,Cu,P produced in Poland and commercially sold as MCP-100. Data from Kron et al.[(8)] are normalized to the response to 6 MV X-rays, while data in the present work and in refs. (11, 12) are relative to ^{60}Co gamma rays. The calculated response follows from Equation (6) in the text.

DISCUSSION

The data in this work showed that the air kerma re- sponse relative to ^{60}Co is significantly different from unity for GR-200A TLDs, in the range 28 – 40 keV, with a maximum over-response of 22 %, while the statistical uncertainty on the relative air kerma re- sponse data was around 2 %. On the other hand, cor- responding data in ref. (8) presented uncertainties from 15 to 42 %, which make those relative response data compatible with unity. In particular, as regards the data of the present work, in Figure 3 there is a little dip in the air kerma response trend vs. photon energy from 28 to 40 keV, which is considered above the experimental error. Indeed, a peculiar air kerma response for LiF:Mg,Cu,P dosemeters is predicted by the normalized mass absorption coefficients shown in Figure 1c in the range 28 – 35 keV. However, when considering correction factors in Equations (7) and (8), the calculated response in Equation (6) presents a relatively smooth trend, predicting an over-response of 5–15 % in the range shown in Figure 3.

Data in the literature for the response of GR-200A TLDs to low-energy monoenergetic X-rays refer to relatively few studies. Bakshi et al.[11] measured the energy response, R, of MCP-100 LiF:Mg,Cu,P rela- tive to ^{60}Co and showed that there is a decrease from 1.26 to 1.23 (data interpolated from Figure 10 in ref. (10)) in going from 30 to 35 keV for SR monoener- getic X-rays. In comparison, for those dosemeters, their Monte Carlo simulations indicated values (cor- rected for the detection efficiency of TLD material) between 1.05 and 1.10 for the relative response, at 30 and 34 keV, respectively. In this range there is a deviation (about 12 %) between their measurements and their corresponding Monte Carlo calculations[11]. Gonzalez et al.[12] presented the relative (to ^{60}Co) energy response of GR-200A dosemeters produced in China, exposed to X-rays from X-ray tubes operated at tube potential in the range 30 – 250 kV. The corre- sponding data points in Figure 3 refer to their data at beam effective energies of 24, 34.5 and 42 keV, for which the estimated uncertainty on the response is 4 %. Their data have a maximum value of 1 (i.e. equal response as to ^{60}Co) at effective energies between 34 and 42 keV and decrease both at lower and higher energies, so showing an under-response of GR-200A of 20 % at 24 keV. However, these results refer to poly- chromatic X-ray spectra, while the data in this work are for monoenergetic X-rays. For example, the 34.5 keV effective energy in ref. (12) corresponded to a photon spectrum extending up to 80 keV. The present data indicated that LiF:Mg,Cu,P GR-200A dosemeters (produced in China) show an over-response in the range 28 – 40 keV. Indeed, two different manufacturers were implied in the two sets of literature data[8, 11]; moreover, the experimental data reported in ref. (11) (R $\cong$ 1.25) are not consistent (within the experimental uncertainties) with the experi- mental data of ref. (8) (R $\cong$ 1). On the other hand, GR-200A exposed to X-ray tube radiation show an under-response in this range of effective energies[12]. The present authors argue that there is an indication of a material-specific and manufacturer-specific air kerma response of LiF:Mg,Cu,P TLDs in the low- energy range explored in the present study, where significant deviations from R $\cong$ 1 (i.e. from the response to ^{60}Co) have been observed with monoenergetic irradi- ation between 28 and 40 keV.

CONCLUSIONS

The authors measured the response (relative to air) of GR-200A TLDs to monoenergetic X-rays in the range 28–40 keV and showed that there is an over- response (relative to air) with respect to irradiation with ^{60}Co gamma rays. The observed deviation in the normalized response (up to 22 %) was considered sig- nificant by taking into account the relative uncer- tainty on their data. These findings are of interest for use of such dosemeters for low-energy X-rays. In the energy range of the present study, and using monoenergetic SR beams, another group[8] measured a slight over-response of about 10 %, but it was compat- ible with unitary response within their experimental un- certainties. The results of this work shown in Figure 3 gave indication of a slight spectral feature in the measured energy range, i.e. the decrease of the relative response, at 35 keV. However, the response (Figure 3, continu- ous line) calculated by the authors on the basis of energy-absorption attenuation of $Li_{72}F_{26}$:$Mg_{0.2}$,$Cu_{0.1}$, $P_{0.7}$ and on suitable correction factors, deviates signifi- cantly from the measured data and predict an under-response. Based on the analysis of all the available data, the authors believe that the issue of the relative energy response of GR-200A (China) TLDs at energies across the transition region from photoelectric-dominated to Compton-dominated interaction (Figure 1a) is still open both experimentally and from the point of view of the available models, though no interpretation has been given here apart from the observation of the calculated spectral feature shown in Figure 1c. To help clarify- ing experimentally this issue, the authors plan to per- form additional measurements with SR monoenergetic beams and GR-200A dosemeters, covering a larger energy range (from 18 to 40 keV) with a fine step of 1 – 2 keV, for finely sampling the spectral air kerma response of GR-200A LiF:Mg,Cu,P TLDs. At the time of writing, following approval of the correspond-ing investigation project, beamtime has been allocated for performing such measurements during 2015 at the SYRMEP beamline at ELETTRA SR facility. These additional data may shed more light on the specific re- sponse of such dosemeters in this low energy range, of specific interest for synchrotron radiation research.

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