LOW-ENERGY X-RAY DOSIMETRY STUDIES (7 to 17.5 keV) WITH SYNCHROTRON RADIATION*

N. E. IPE, H. BELLAMY, J. R. FLOOD, K. R. KASE, and P. PHIZACKERLEY Stanford Linear Accelerator Center Stanford University, Stanford, California 94309

and

K. J. VELBECK AND R. TAWIL

HARSHAW/BICRON

6753-I Cochran Road, Solon, Ohio 44139

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Abstract

Unique properties of synchrotron radiation (SR), such as its high intensity, brightness, polarization, and broad spectral distribution (extending from x-ray to infra-red wavelengths) make it an attractive light source for numerous experiments. As SR facilities are rapidly being built all over the world, they introduce the need for low-energy x-ray dosemeters because of the potential radiation exposure to experimenters. However, they also provide a unique opportunity for low-energy x-ray dosimetry studies because of the availability of monochromatic x-ray beams. Results of such studies performed at the Stanford Synchrotron Radiation Laboratory are described. Lithium fluoride TLDs (TLD-100) of varying thicknesses (0.015 to 0.08 cm) were exposed free in air to monochromatic x-rays (7 to 17.5 keV). These exposures were monitored with ionization chambers. The response (nC/Gy) was found to increase with increasing TLD thickness and with increasing beam energy. A steeper increase in response with increasing energy was observed with the thicker TLDs. The responses at 7 and 17.5 keV were within a factor of 2.3 and 5.2 for the 0.015 and 0.08 cm-thick TLDs, respectively. The effects of narrow (beam size smaller than the dosemeter) and broad (beam size larger than the dosemeter) beams on the response of the TLDs are also reported.

INTRODUCTION

Synchrotron radiation (SR) is produced as a natural consequence of bending the trajectory of energetic charged particles. These charged particles radiate energy as they are being deflected in the magnetic fields of bending magnets or special devices called insertion devices.

Synchrotron radiation emitted by high-energy electrons or positrons bent in the magnetic fields of storage rings is extremely intense. It spans a broad range of wavelengths, extending from infrared, through the visible and ultraviolet range, and into the hard x-ray region of the electromagnetic spectrum. The entire spectrum is referred to as white light. Synchrotron radiation is transmitted through beam ports in the storage ring shielding wall to the experimental floor. Because of its unique properties (such as high intensity, brightness, broad spectral range, collimation, polarization, etc.), synchrotron radiation has become a powerful tool for basic and applied research in the fields of physics, biology, chemistry, medicine and technology.

There are currently over thirty SR facilties in operation around the world, with approximately another twenty-eight in the construction, design, or proposal stage. As SR facilities are rapidly being built all over the world, they introduce the need for low-energy x-ray dosemeters because of potential radiation exposure to experimenters who work in close proximity to the SR beamlines and experimental enclosures. However, they also provide a unique opportunity for low-energy x-ray dosimetry studies because of the availability of monochromatic (mono) x-ray beams. Preliminary studies were conducted at the Stanford Synchrotron Radiation Laboratory (SSRL): (a) to investigate the low-energy response of Harshaw/Bicron lithium fluoride thermoluminescent dosemeters (TLDs), and (b) to investigate the feasibility of using synchrotron radiation for low-energy x-ray dosimetry studies. Results of these studies are presented.

EXPERIMENTAL CONDITIONS

Experiments were performed at SSRL Beamline 1-5 using synchrotron radiation from the 3 GeV electron storage ring SPEAR (a schematic of the beamline is shown in Fig. 1). Beamline 1-5 accepts 0.75 milliradians of synchrotron radiation from a bending magnet. The synchrotron radiation enters the beamline through a pair of beryllium windows, followed by another beryllium window.¹⁾ The approximate spot size of the beam is 2 cm × 0.3 cm. The entire spectrum is referred to as white light. The dimensions of the white x-ray beam can be defined by a pair of vertical slits and a pair of horizontal slits. The beam is then incident on a downward reflecting monochromator, which can be used to select the desired x-ray energy. The monochromator, comprised of two silicon crystals cut with their surfaces paralled to the (111) plane, provides an energy resolution ($\Delta E/E$) of about 2×10⁻⁴ at 8 keV. A white beam stop in the monochromator assures that white light does not enter the downstream experimental enclosure. A piezoelectric crystal adjustment is used to detune the monochromators to eliminate the higher energy harmonics (lower in intensity) that normally accompany the x-ray beam of selected energy. The x-ray beam is then transported into two experimental enclosures that are in tandem (called hutches, not shown in the figure). This experiment used the downstream hutch. These hutches are electronically interlocked to ensure safe access. The pair of mono beam shutters at the entrance to the hutch are closed during access. A second set of horizontal and vertical beam-defining slits are placed downstream of the mono beam shutters. Because the beam is essentially parallel, these slits define the beam size at the sample position (24.8 m from the point x-ray source).

Two parallel plate ion chambers are mounted between the slits and the sample. A shutter and a series of filters are mounted in a holder between the ion chambers. The shutter regulates the x-ray dose to the sample. The filters are used to calibrate the energy of the mono beam by scanning across an absorption edge (7.112 keV for iron, 8.979 keV for copper, and 11.919 keV for gold), while measuring the beam intensity with the ion chambers both before and after the filter. A collimator after the second ion chamber acts as a guard slit. The ion chambers are operated at a high voltage of 310 volts and are filled with slowly flowing nitrogen at atmospheric pressure. The parallel plates are made of aluminum and spaced 1 cm apart. Digital LED readout of the ion chamber currents is displayed by a NIM module above the operator's computer terminal, located outside the hutch. The sample is placed in a sample holder mounted on a goniostat that has three independent rotation and three independent translation axes. A microscope mounted at 135° to the incident beam direction is used to center the sample. The entire system is under the control of a computer that drives

the monochromator to the desired energy and exposes the sample to the selected radiation dose. The lowest usable energy for Beamline 1-5 is 4.5 keV because of the absorption of the x-ray beam in the berryllium windows, the helium filled beam pipes, and the monochromator housing and kapton windows of various components. The highest attainable energy is < 20 keV and is limited by the silicon (111) crystals.

The lithium fluoride TLDs obtained from Harshaw/Bicron were chipstrate dosemeters. These dosemeters consist of TLD-100 chips hermetically bonded to a polyimide substrate, to which an ID bar code is attached. Three sets of TLD-100 with different sizes of 0.1 cm \times 0.1 cm, 0.2 cm \times 0.2 cm, and 0.3175 cm \times 0.3175 cm were used. The first set (0.1 cm \times 0.1 cm) was composed of TLDs with different thicknesses: 0.01524, 0.02, 0.04, 0.06, and 0.08 cm. The second and third sets were of the same thickness, 0.01524 cm.

The Harshaw/Bicron TLD System 8800 was used to read out the TLDs. The TLD System 8800 workstation is a fully automated state-of-the-art system for TLD measurement. It combines high capacity and throughput capabilities with noncontact heating A precisely temperature-controlled stream of hot nitrogen gas is used to heat the TLD elements.

The TLD readout consists of a 50°C preheat for zero seconds, followed by heating at a temperature ramp rate of 15°C/s for twenty seconds, and heating at 300°C for 3 1/3 seconds. The total acquisition time is 23 1/3 seconds. The TLD is then allowed to return to ambient temperature.

EXPERIMENTAL PROCEDURE

To obtain a horizontal beam profile, the vertical slits downstream of the mono beam shutters were narrowed to an aperture of 0.15 cm, while the horizontal slits were narrowed to an aperture of 0.025 cm. The vertical slits were centered at 5.82 cm, while the horizontal slits were scanned across the beam and the ion chamber current noted at each position. Figure 2 shows the horizontal beam profile for 7 keV x-rays where the intensity (proportional to ion chamber current) is plotted as a function of the position of the center of the slits. The beam is relatively uniform over a horizontal distance of about 2 cm. The horizontal slits were then centered at 3.44 cm and the aperture widened to 0.15 cm. The vertical slit aperture was narrowed to 0.025 cm and the slits were scanned across the beam. The vertical profile for 7 keV x-rays is shown in Fig. 3. Due to limiting apertures, the vertical profile deviates significantly from the flattened Gaussian shape (full width at half maximum \cong 0.3 cm) that it should assume. The vertical slits were centered to the right of the maximum in Fig. 3.

The horizontal and vertical slits upstream of the beam stoppers were adjusted so that the beam size at the sample position was $0.15 \text{ cm} \times 0.15 \text{ cm}$. This was verified by measuring the size of the beam from a photograph. Experiments were also performed with the beam size set to $0.13 \text{ cm} \times 0.13 \text{ cm}$. The monochromator was calibrated using an iron or copper filter for the low energies and the gold filter for the higher energies. The monochromator was then set at various energies ranging from 7 to 17.5 keV. At each energy, the harmonics were monitored by studying the spectrum

scattered by a thin kapton foil using a Bicron NaI scintillator (Model 1XMP040B-X). The NaI scintillator was mounted vertically at 90° to the incident beam direction. The monochromator was detuned so that the intensity of the mono beam was reduced by 20%. This amount of detuning was sufficient to eliminate completely the higher harmonics at each energy (the harmonics were most significant at 7 keV).

The TLDs were mounted on the sample holder and centered using the microscope. Three TLDs were irradiated free in air at each energy for approximately 10 to 20 seconds. The temper-ature and ion chamber integrated currents were noted at each irradiation. The ion chamber read-ings were converted to kerma and corrected for attenuation in the nitrogen and air paths to obtain the kerma at the sample position. The kermas varied from approximately 0.1 to 0. 2 Gy.

The TLDs were read out at Harshaw/Bicron. Control readings (six controls were used per energy) were subtracted out. For calibration purposes, a fade factor was applied to normalize responses to Battelle data (Battelle, Pacific Northwest Laboratories, Battelle Boulevard, P.O. Box 999, Richland, Washington 99352). The Batelle data consisted of TLD readings from exposure to ⁶⁰Co and 17.5 keV x-rays (k-fluorescent x-rays from molybdenum), both free in air and on a 30 cm \times 30 cm \times 15 cm polymethyl methacrylate phantom.

The 60 Co exposures were performed with sufficient build up material. The TLDs were exposed to kermas of 0.1 Gy at each energy.

RESULTS AND DISCUSSION

Figure 4 shows the measured and predicted responses in nanoCoulombs/Gray (nC/Gy) as a function of energy for different thicknesses of TLD-100 (0.1 cm \times 0.1 cm). The measured response (average of three TLDs) increases with both energy and thickness. There is a steeper increase in response with energy for the thicker TLDs. Measured responses at 7 and 17.5 keV are within a factor of 2.3 and 5.2 for the 0.015 and 0.08 cm-thick TLDs, respectively. The error bars are not visible. The light output of the TLD is proportional to the energy absorbed E_{ab}.

$$\mathbf{E}_{ab} = \Phi \mathbf{E} \left(1 - \exp\{-\mu_{en} \mathbf{d}\} \right) \mathbf{A} ,$$

where ϕ is the photon fluence in cm⁻², E is the energy of the photon in eV, μ_{en} is the energy absorption coefficient of the TLD in cm⁻¹, d is the thickness of the TLD in cm, and A is the area of the beam in cm⁻². The kerma (K) in air is given by

$$K = \phi E \left(\frac{\mu_{tr}}{\rho}\right)_{air}$$

where $(\mu_{\rm tr} / \rho)_{\rm air}$ is the mass energy transfer coefficient of the air in cm² g⁻¹

For x-rays of energy less than 50 keV, the photon mean free path is much greater than the range of the liberated electrons.²⁾ Thus, electronic equilibrium is achieved. Since the electrons lose only a negligible fraction of their energy by the bremsstrahlung process, the mass energy transfer coefficient is equal to the mass energy absorption coefficient. Under these conditions, the kerma is equal to the absorbed dose D.

$$K = D = \phi E \left(\frac{\mu_{en}}{\rho}\right)_{air}$$

where $(\mu_{\rm tr} / \rho)_{\rm air}$ is the mass energy absorption coefficient of air.

The predicted response is therefore proportional to

$$\frac{\mathrm{E}_{\mathrm{ab}}}{\mathrm{k}} = \frac{\left(1 - \exp\{-\mu_{\mathrm{en}}\mathrm{d}\}\right)\mathrm{A}}{\left(\mu_{\mathrm{en}}/\rho\right)_{\mathrm{air}}}$$

The energy absorption coefficients from Ref. 3 were used.³⁾ The predicted response at each thickness was normalized to 7 keV. For TLD thicknesses of 0.01524 cm and 0.02 cm, the predicted and measured responses are fairly close. There are however significant differences between the predicted and measured responses for thicknesses \geq .04 cm. These differences may be attributed to the attenuation of the light, and the attenuation of the low-energy x-rays in the thicker TLDs. Further, the predicted response does not take into account any scattering from the substrate that the TLD is bonded to.

Figure 5 shows the measured response of the TLDs as a function of thickness for different energies. Also shown are the responses to 60 Co and 17.5 keV (free in air) x-ray exposures performed at Battelle. The SSRL data at 17.5 keV are about 11 to 17% greater than the Battelle data. The differences are not fully understood.

Figure 6 shows the response of the TLDs relative to 60 Co, free in air.

Based on the above results, the 0.01524 and 0.02 cm-thick TLDs have the least energy dependence. However, there is still a factor of 2 increase in response between 7 and 17.5 keV.

To determine the feasibility of using synchrotron radiation for low-energy x-ray dosimetry studies, the limitations must be identified. The small spot size of the synchrotron radiation beam, which is dictated by the vertical beam size, necessitates the use of dosemeters smaller than the beam, unless it is possible to predict the response of larger dosemeters using the results from smaller dosemeters. Tables 1 and 2 show the predicted response of large dosemeters using narrow beams (beams smaller in size than the dosemeter) and broad beams (beams larger in size than the dosemeter).

Table 1 shows the measured and predicted responses for the narrow beam at each energy for the SSRL exposures. Columns 2, 3, and 4 show the average of the measured responses for 3 TLDs, and the standard deviation of the average responses for the three different dosemeter sizes. Column 5 shows the predicted response for the large dosemeters obtained by multiplying the response of the 0.1 cm \times 0.01524 cm dosemeter by the ratio of the beam area to the dosemeter area (1.5²). The predicted responses are within the uncertainities of the measured responses except at 17.5 keV where the difference is 15%. This large difference may be attributed to the errors in centering the sample, especially in the vertical plane where the dose rates fall off rapidly as shown in Fig. 3.

Columns 2 and 3 in Table 2 show the measured responses of $0.1 \text{ cm} \times 0.1 \text{ cm} \times 0.1524 \text{ cm}$ and $0.3175 \text{ cm} \times 0.3175 \text{ cm} \times 0.01524 \text{ cm}$ TLDs when exposed to broad beams of ⁶⁰Co and 17.5 keV x-rays at Battelle. Column 4 shows the calculated response of the 0.3175 cm $\times 0.3175 \text{ cm} \times 0.01524 \text{ cm}$ TLD obtained by

multiplying the response of the 0.1 cm \times 0.1 cm \times 0.01524 cm by the ratio of the areas (3.175²). The predicted responses are about 5% higher than the measured responses.

Columns 2 through 5 in Table 3 shows the measured responses for two different beam sizes, $0.15 \text{ cm} \times 0.15 \text{ cm}$ and $0.13 \text{ cm} \times 0.13 \text{ cm}$. Columns 6 and 7 show the predicted response for the 0.13 cm $\times 0.13$ cm beam obtained by multiplying the response for the 0.15 cm $\times 0.15$ cm beam for each dosemeter size by the ratio of the areas of the two beams (0.75). These predicted responses are within 2 to 13% of the measured responses. As expected, the measured responses for the two different sized dosemeters are not significantly different.

For x-ray energies of < 10 keV, the skin dose dominates the effective dose.⁴⁾ It may therefore be more appropriate to report the response of TLDs for low-energy x-rays, relative to the dose equivalent at a depth of 0.07 mm, or at depths below 0.1 mm when the calibrations are performed with the dosemeters fixed to the surface of a phantom. This will be the thrust of future studies.

CONCLUSIONS

The need for low-energy x-ray dosemeters has increased because of the increasing number of synchrotron radiation facilities being built around the world. The SSRL Beamline 1-5 was used for low-energy x-ray dosimetry studies (7 to 17.5 keV). The measured responses of Harshaw /Bicron LiF TLDs were found to increase with increasing energy and increasing thickness of the TLD, as expected. A steeper

increase in response with energy was observed with the thicker TLDs. The thinnest TLDs (0.01526 and 0.02 cm) have the least energy dependence; however, the response increases by about a factor of 2 between 7 and 17.5 keV. The predicted responses were much higher than the measured responses for the thicker TLDs (≥ 0.04 cm), possibly because of the attenuation of light and the attenuation of the low-energy x-rays in the TLD.

The small vertical dimensions of the synchrotron radiation beam may appear to place a limitation on the size of the dosemeters that can be used. However, it has been demonstrated that it is possible to expose large dosemeters to narrow beams, and then predict the response for broad beams by correcting the response to narrow beams for the effect of the beam size and dosemeter size. The predicted responses were in general within the uncertainities of the responses, except at 17.5 keV, where a 15% difference was observed.

Future studies will involve the investigation of the use of other dosemeter types, as well as the response relative to the dose equivalent at a depth of 0.07 mm, or at depths less than 0.1 mm, since the skin dose dominates the effective dose at these low energies.

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	Mea	Predicted reponse		
Е		for columns 3 & 4		
(keV)	$(0.1 \text{ cm})^2$	$(0.2 \text{ cm})^2$	$(0.3175 \text{ cm})^2$	(nC/Gy)
7	6.66 ± 0.37	15.27 ± 0.11	14.83 ± 0.69	14.98
9	9.72 ± 0.53	23.11 ± 0.99	19.98 ± 0.33	21.87
12	12.43 ± 0.08	26.75 ± 1.71	27.90 ± 1.59	27.97
15	13.19 ± 0.01	29.59 ± 0.13	28.34 ± 0.97	29.68
17.5	15.18 ± 0.53	30.43 ± 0.92	29.64 ± 0.58	34.15

Table 1. Narrow beam response of LiF TLD-100 as a function of dosemeter size. (Dosemeter thickness is 0.01524 cm and beam size is 0.15 cm $\times 0.15$ cm.)

Table 2. Broad beam response of LiF TLD-100 as a function of dosemeter size. (Dosemeter thickness is 0.01524 cm.)

	Measur (1	red Response nC/Gy)	Predicted Reponse (nC/Gy)	
Е	Dose	emeter size	Dosemeter size	
(keV)	$(0.1 \text{ cm})^2$	$(0.3175 \text{ cm})^2$	$(0.3175 \text{ cm})^2$	
1250	9.31 ± 0.04	90.40 ± 0.07	93.85	
17.5	13.83 ± 0.03	132.44 ± 0.60	139	

		Measured resp	Predicted reponse (nC/Gy)			
	Beam size $(0.15 \text{ cm})^2$		Beam size $(0.13 \text{ cm})^2$		Beam size $(0.13 \text{ cm})^2$	
Е		Doseme	Dosemeter size			
(keV)	$(0.2 \text{ cm})^2$	$(0.3175 \text{ cm})^2$	$(0.2 \text{ cm})^2$	$(0.3175 \text{ cm})^2$	$(0.2 \text{ cm})^2$	$(0.3175 \text{ cm})^2$
7	15.27 ± 0.11	14.83 ± 0.69	11.00 ± 0.48	11.97 ± 0.69	11.47	11.14
9	23.11 ± 0.99	19.98 ± 0.33	15.89 ± 0.46	16.69 ± 0.68	17.35	15.01
12	26.75 ± 1.71	27.90 ± 1.59	18.27 ± 0.64	18.56 ± 0.78	20.09	20.96
15	29.59 ± 0.13	28.34 ± 0.97	21.75 ± 0.48	20.76 ± 0.88	22.22	21.29
17.5	30.43 ± 0.92	29.64 ± 0.58	24.39 ± 0.92	23.15 ± 0.54	22.86	22.26

Table 3. Narrow beam response of LiF TLD-100 as a function of beam size. (Dosemeter thickness is 0.01524 cm.)



Figure 1. Schematic of SSRL Beamline 1-5.



Figure 2. Horizontal beam profile.



Figure 3. Vertical beam profile.



Figure 4. Measured and predicted responses of LiF TLD–100 as a function of energy for various TLD thicknesses.



Figure 5. Measured response of LiF TLD–100 as a function of thickness for various energies.



Figure 6. Measured response of LiF TLD-100 relative to 60 Co as a function of energy for various TLD thicknesses.