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P. A. Eschbach
S. D. Miller

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Pacific Northwest Laboratory
Richland, Washington 99352

A GAMMA/NEUTRON-DISCRIMINATING, COOLED, OPTICALLY STIMULATED LUMINESCENCE (COSL) DOSEMETER

Peter A. Eschbach, Steven D. Miller

Pacific Northwest Laboratory[a]

ABSTRACT

The Cooled Optically Stimulated Luminescence (COSL) of CaF_2:Mn (grain sizes from 0.1 to 100 microns) powder embedded in a hydrogenous matrix is reported as a function of fast-neutron dose. When all the CaF_2:Mn grains are interrogated at once, the COSL plastic dosemeters have a minimum detectable limit of 1 cSv fast neutrons;[1] the gamma component from the bare ^{252}Cf exposure was determined with a separate dosemeter. We report here on a proton-recoil-based dosemeter that generates pulse height spectra, much like the scintillator of Hornyak,[2] to provide information on both the neutron and gamma dose.

We first submitted the plastic dosemeters to an optical bleaching procedure[4] using a high-intensity ultraviolet laser and then exposed the dosemeters to fast neutrons using an unmoderated ^{252}Cf source. The previously used COSL process[3] relied on thermal contact of the cooled dosemeter with a room-temperature slab of metal to promote electrons from the shallow traps. In this case, however, we used the standard COSL cooling and light-stimulation process to phototransfer electrons from deep to shallow traps, but with the use of a CO_2 laser to selectively warm the individual CaF_2:Mn grains within the plastic matrix. Grains within the plastic dosemeter that received large

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localized energy deposition produced larger-than-average COSL response when warmed by the CO_2 laser. The data we present here demonstrate the concept of a neutron-discriminating dosemeter. Future work will concentrate on the refinement of the individual grain readout process until individual grains receiving a single neutron-induced recoil proton can be distinguished.

INTRODUCTION

Previous publications(3,4,5) have shown that CaF_2:Mn (TLD 400) emits light proportional to dose when the Cooled Optically Stimulated Luminescence (COSL) process is used. Sensitivities to gamma dose of 26 nC/Kg (1 μGy)(6) and an optical annealing procedure(4) have been demonstrated. In these cases, the dosemeters showed no appreciable fade after a period of one year (if the exposed dosemeters were kept in the dark).(5) Having demonstrated these three characteristics, we were convinced that a fast-neutron dosemeter could be constructed using CaF_2:Mn grains within a polyethylene matrix. Preliminary work(1) demonstrated a lower detectable limit of 10 cSv fast-neutron dose equivalent when exposed to a bare ^{252}Cf source. In this study, all of the grains were interrogated simultaneously using the COSL process; luminescence of the polyethylene matrix itself decreased the lower limit of detection. Here we report results obtained by reading individual grains in the polyethylene matrix. Warming the individual grains directly avoids the interfering luminescence from the polyethylene, and the lower limit of detection is therefore reduced to 1 cSv of fast-neutron dose equivalent.

EXPERIMENTAL

The COSL method is particularly attractive for use in neutron dosimetry because the light emission occurs as the dosemeter warms from liquid nitrogen to room temperature after exposure to ultraviolet light.[3] Thus, normal plastic matrices, such as those of polyethylene, can be used without fear of melting the dosemeter. Another benefit of the COSL process is the existence of a room-temperature optical annealing procedure,[4] which allows reuse of the same dosemeter.

For this study, CaF_2:Mn (TLD-400) in granular form (nominal particle size, 200 microns) was compounded with polyethylene powder and then injection-molded into 0.3-mm thin sheets. The sheets of dosemeter material were then cut into 12.5-mm by 12.5-mm square dosemeters. The cut dosemeters were then optically annealed with 3 Joules of 351- and 363-nm light from an argon ion laser. The annealed dosemeters were then placed in light-resistant paper envelopes for exposure to fast neutrons from a bare (unmoderated) ^{252}Cf source. Another type of exposure was performed with a ^{60}Co "hot particle," with an activity 5.5 μCuries and a diameter of 50 microns. The hot particle exposure was used to demonstrate the scanning reader's ability to read dose in a small localized area.

Exposed dosemeters were "read out" in a semi-automated reader, designed and constructed at Pacific Northwest Laboratory. In this system, dosemeters are placed on top of a sapphire window that is eventually cooled to -150°C. To prevent the formation of frost on the sapphire window as the dosemeter is cooled, the dosemeter and sapphire window are kept under vacuum. While cold, the dosemeters are illuminated with approximately 100 mJoules of multi-line UV light from an argon ion laser. After stimulation of the dosemeter with the ultraviolet light, individual grains of CaF_2:Mn are warmed up to room temperature with a focused beam from a CO_2 laser. Figure 1 is a pictorial representation of the laser scanning reader showing the location of the various components mentioned above. Typical power from the CO_2 laser was 0.5 W, focused to a spot 100 microns in diameter. The CO_2 laser emission heats the grain(s) of CaF_2:Mn only when the CO_2 shutter is open, typically for 0.5 s. COSL light emitted from the heated grain passes through a highly reflective, square light pipe to homogenize the light. The homogenized light strikes a Burle 8575 photomultiplier tube. The light striking the phototube is photon-counted, using a fast pre-amplifier (Ortec 9301), a discriminator (Ortec 436), and a Multi-Channel Scaling (MCS) system.

The MCS system is turned on by the control software coincident with the opening of the CO_2 laser shutter. Data are acquired by the MCS system for one second. After the MCS system has acquired the full one second of data, the control software moves the CO_2 laser optics to the next location for additional data acquisition. In this manner, 100 sites (10x10 grid) on the 12.5-mm-square dosemeter are interrogated and recorded. The whole readout process takes about 5 minutes. The control software was written to allow the user to select a preset region of interest to integrate. Integration of the region of interest is done "on the fly" and stored in a separate data file, along with the CO_2 laser power and cold finger temperature for all 100 sites.

The integrated values can be recalled and summed at a later date to provide a pulse height distribution of all 100 sites on the dosemeter.

RESULTS

One of the initial concerns of the experiment was the effect of flooding the photocathode of the photomultiplier tube with the CO_2 laser emission. To check the effect we ran a 10-by-10 scan across the sapphire window with no dosemeters in place. Observing no appreciable increase in photon count rate on any of the 100 sites when the CO_2 laser struck the sapphire window, we were satisfied that the CO_2 beam was not interacting with the photocathode directly.

The maximum temperature reached by the CaF_2:Mn during the CO_2 laser heating cycle is unknown. However, as seen in Figure 2, the areas struck by the CO_2 laser beam did melt, indicating that temperatures of at least 120°C were reached in the vicinity of the grain(s).

Figure 3 captures the COSL emission observed at one site on a dosemeter exposed to 1 cSv of dose-equivalent fast neutrons. The emission of light coincides fairly well with the opening of the CO_2 laser. However, since heating rates differ, depending on the proximity of the focused CO_2 spot to the grain, it is not possible to determine whether the decay time of the peak itself is correct. The integrated region from 0.0 milliseconds to 600 milliseconds had a value of 4700 counts for this particular peak. Figure 4 plots the integrated values from all 100 sites in the pulse height distribution. From the distribution, we observed 15 sites of the 100 sites with elevated count rates. The lower panel in Figure 4 displays a pulse height distribution from an unexposed (control) dosemeter. The control dosemeter has only one peak in the 2000-3000 count range, indicating that there is a net response to the 1 cSv fast-neutron exposure.

Figure 5 plots another pulse height distribution for a much higher exposure of fast neutrons, 1 Sv dose equivalent. The higher dose exposure shows that the majority of the integrated areas from the individual grain read-outs are larger than 2000 counts. Again, the lower panel of Figure 5 shows a pulse height distribution for a control dosemeter. The net response to the 1 Sv exposure is overwhelming in comparison with the control (unexposed) dosemeter. The ^{252}Cf source used to irradiate these dosemeters typically produces a gamma-dose equivalent that is 6% of the delivered neutron-dose equivalent. To quantify the response of the dosemeters to the inherent gamma component, a group of the dosemeters was exposed to 6 cSv of dose equivalent gamma from a ^{137}Cs source. The pulse height distribution from one of these gamma-exposed dosemeters appears in Figure 6. As can be seen in the figure, the gamma component from the ^{252}Cf irradiation is a small percentage of the neutron response displayed in the upper panel of Figure 5.

To test the resolution of the scanning readout concept, we placed a 50-micron-diameter particle of ^{60}Co in direct contact with an annealed polyethylene/CaF_2:Mn dosemeter for 1 min. The dosemeter was then read out with the laser scanning reader. The resulting COSL response is represented in Figure 7. One site had a response of well over one million counts in the integrated region of interest; surrounding sites also displayed elevated count rates. The count rate dropped rapidly to less than 2000 counts per region of interest away from the highly dosed area.

DISCUSSION

The net response of polyethylene/CaF_2:Mn dosemeters to fast neutrons has been demonstrated with grain sizes that are not optimal for maximum neutron-to-gamma response. However, a study is currently underway in which finer powder (with smaller than 50-micron grains of CaF_2:Mn), incorporated 20% by weight in polyethylene, is exposed to both gamma and neutron fields. Using pulse height distributions such as those in Figures 4 and 5, it is hoped to determine a neutron-to-gamma response.

In early work Fellinger et al.(7) demonstrated that the fast-neutron-to-gamma sensitivity ratio is increased by incorporating smaller grains of phosphor; in later work, Chassende-Baroz et al.(8) also demonstrated this effect by incorporating thinner layers of phosphor.

Based upon a quick calculation using our best observed specific response of CaF_2:Mn to gamma radiation, 3.6 million counts per gram of CaF_2:Mn per sV, the gamma response of a 50-micron grain of CaF_2:Mn would be 5.45 counts per 10 μSv. Thus, it would take a gamma dose of over 400 μSv to produce any response over the 2000 count/(region of interest) instrument background. As the radius to the third power, the gamma response decreases for spherical grains of CaF_2:Mn. This is the benefit of small powders in plastic matrices:

the neutron-to-gamma response should dramatically increase with smaller powders. As the grains become smaller, however, they are also harder to hit directly with the CO_2 laser beam. In addition, as the grains become smaller, the chance of warming more than one grain with the CO_2 laser increases, making the analysis of data more complex.

CONCLUSION

The response of the polyethylene-CaF_2:Mn dosemeters to fast neutrons has been demonstrated. This response was established by plotting a pulse height spectrum consisting of the integrated counts for a preset region of interest. The dosemeter exposed to 1 Sv of fast-neutron dose equivalent displayed an overwhelming response when compared with the response due to the gamma component from the ^{252}Cf irradiation and the control dosemeter. Another group of dosemeters was exposed to 1 cSv of dose-equivalent neutrons from a ^{252}Cf source; these dosemeters showed a significant response compared with a control dosemeter.

These results were obtained with dosemeters containing CaF_2:Mn grains ranging from 100 to 300 microns. Future work will use dosemeters containing CaF_2:Mn grains that have passed through a sieve and have diameters of less than 50 microns. Using these dosemeters, we plan to generate a neutron-to-gamma response factor, and to complete a study using accelerator-generated neutrons, and possibly thermal neutrons, to quantify the energy response of the dosemeters. It should be possible to substantially decrease the lower limit of detection by improving the collection efficiency of the current phototube/cold finger configuration. We also hope to realize substantial improvement by adjusting the control program's parameters: integration region, CO_2 laser heating-pulse width, and UV-stimulation energy.

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FIGURE CAPTIONS

1. Schematic of laser scanning reader. The scan optic is connected to an x and y translation stage for movement of the focused CO_2 beam across the dosemeter surface. The cold finger and phototube are surrounded by a bell jar and pumped to a rough vacuum to reduce frosting.

2. Photograph of microscope image of a dosemeter that has been read out using the laser scanning reader. The melt pits indicate where the CO_2 beam was incident upon the dosemeter. The lower photograph is a 100x image of the central part of the 40X upper image. The lower image shows an elliptical melt pit; at least one grain is covered by the melt pit.

3. COSL emission from one grain of CaF_2:Mn within a polyethylene matrix. The whole dosemeter was exposed to 1 cSv dose-equivalent fast neutrons from ^{252}Cf.

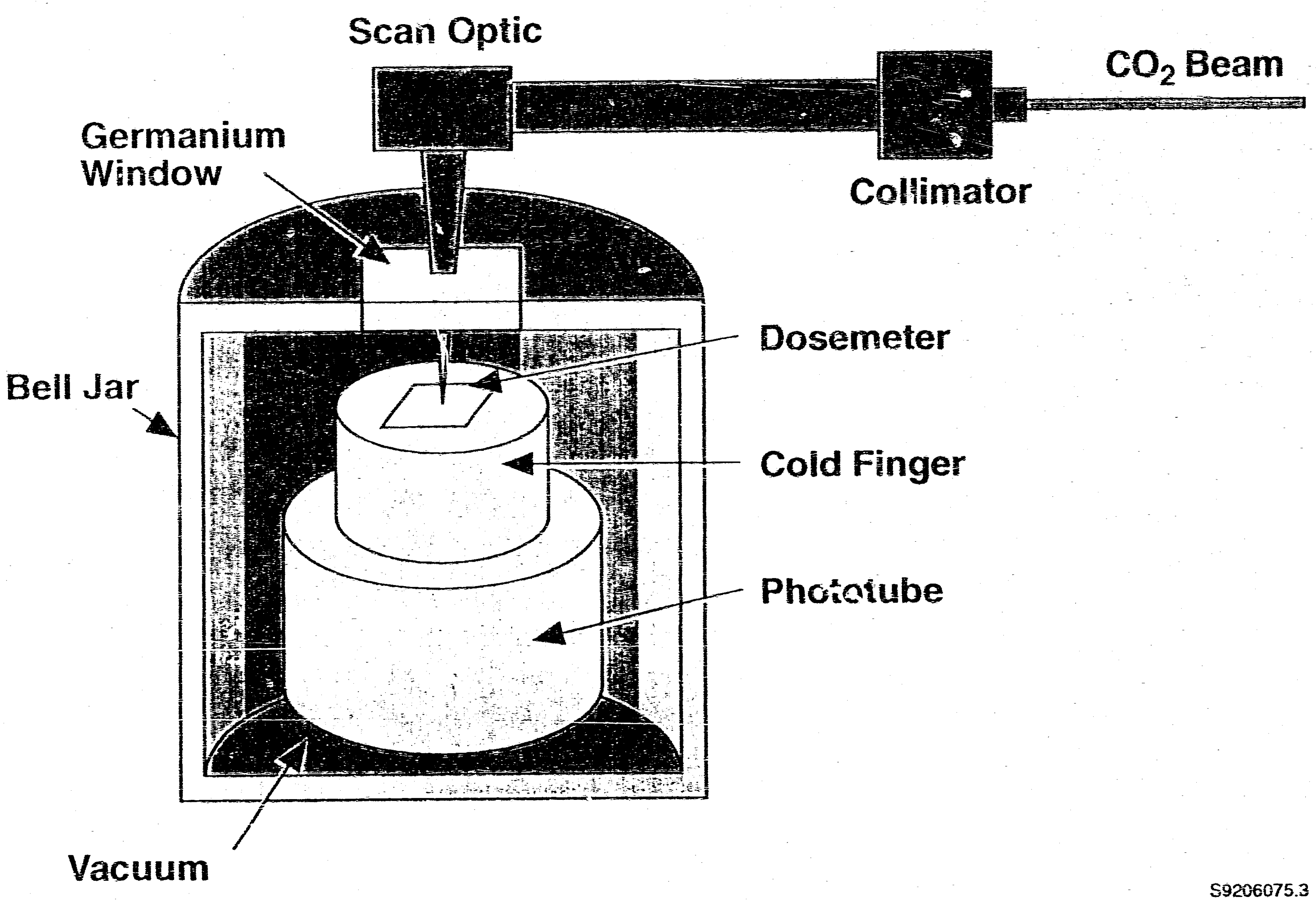
4. The upper plot is the pulse height distribution from 1 cSv dose-equivalent fast neutrons from ^{252}Cf. The lower plot is the pulse height distribution from the control dosemeter.

5. The upper plot is the pulse height distribution from 1 Sv dose equivalent fast neutrons from ^{252}Cf. The lower plot is the pulse height distribution from the control dosemeter.

6. The pulse height distribution from a dosemeter exposed to 6 cSv of gamma dose equivalent from ^{137}Cs.

7. COSL emission from a 10 x 10 scan of a polyethylene/CaF_2:Mn dosemeter exposed to a 5.5 μCurie ^{60}Co particle. The particle had a major axis diameter of 50 microns.

Laser Scanning Reader



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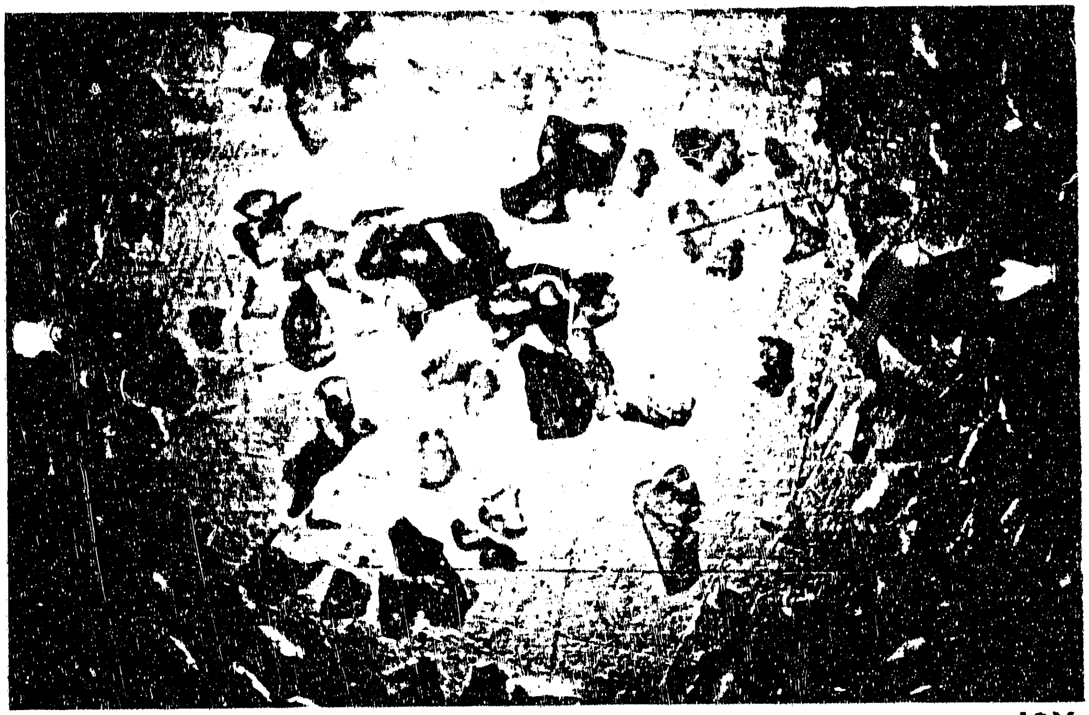
Microscope Images of Polyethylene/CaF_2:Mn Dosemeter Exposed to Emission from a CO_2 Laser



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100X

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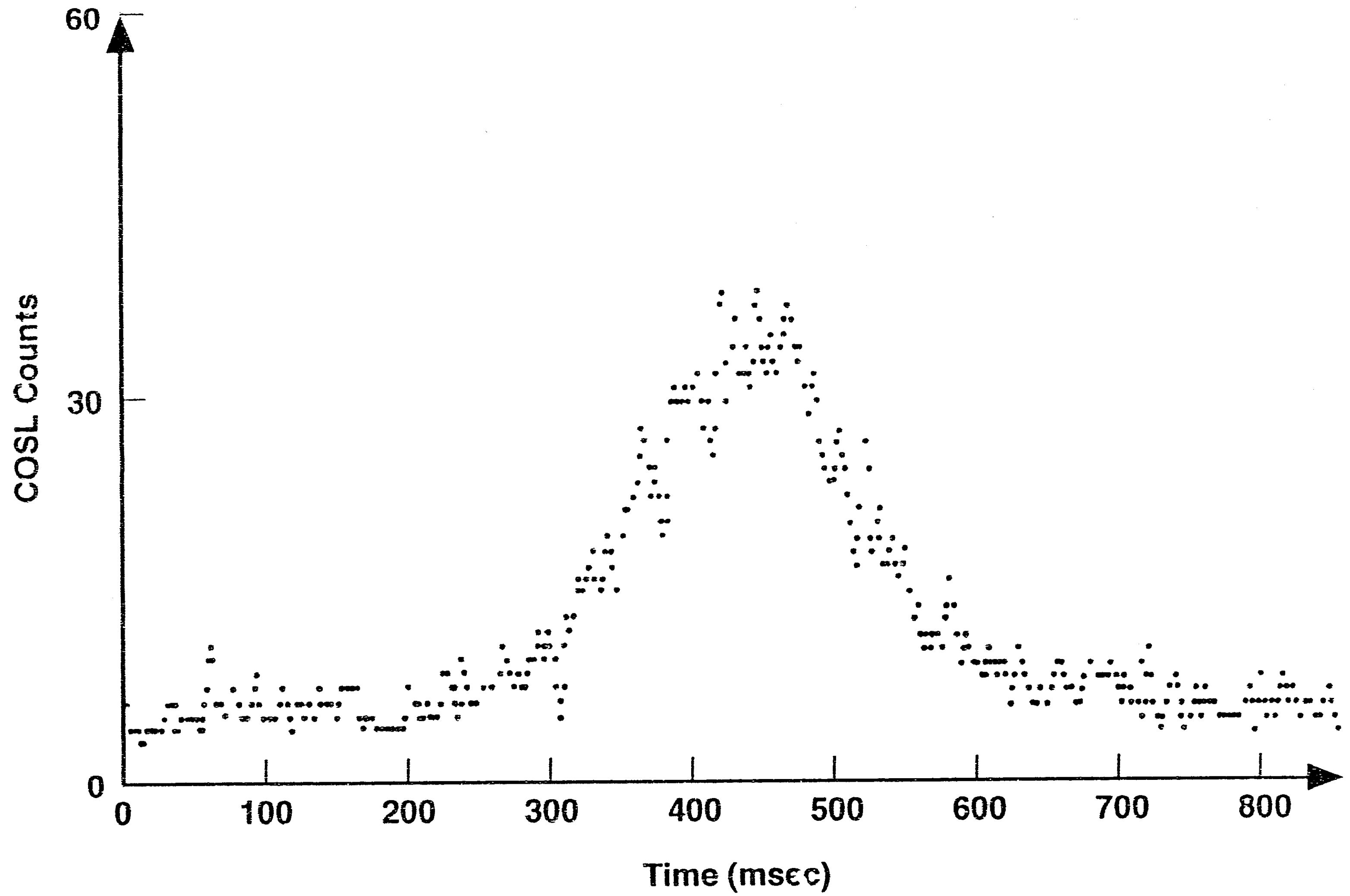
COSL Emission from a Single Grain of CaF_2:Mn



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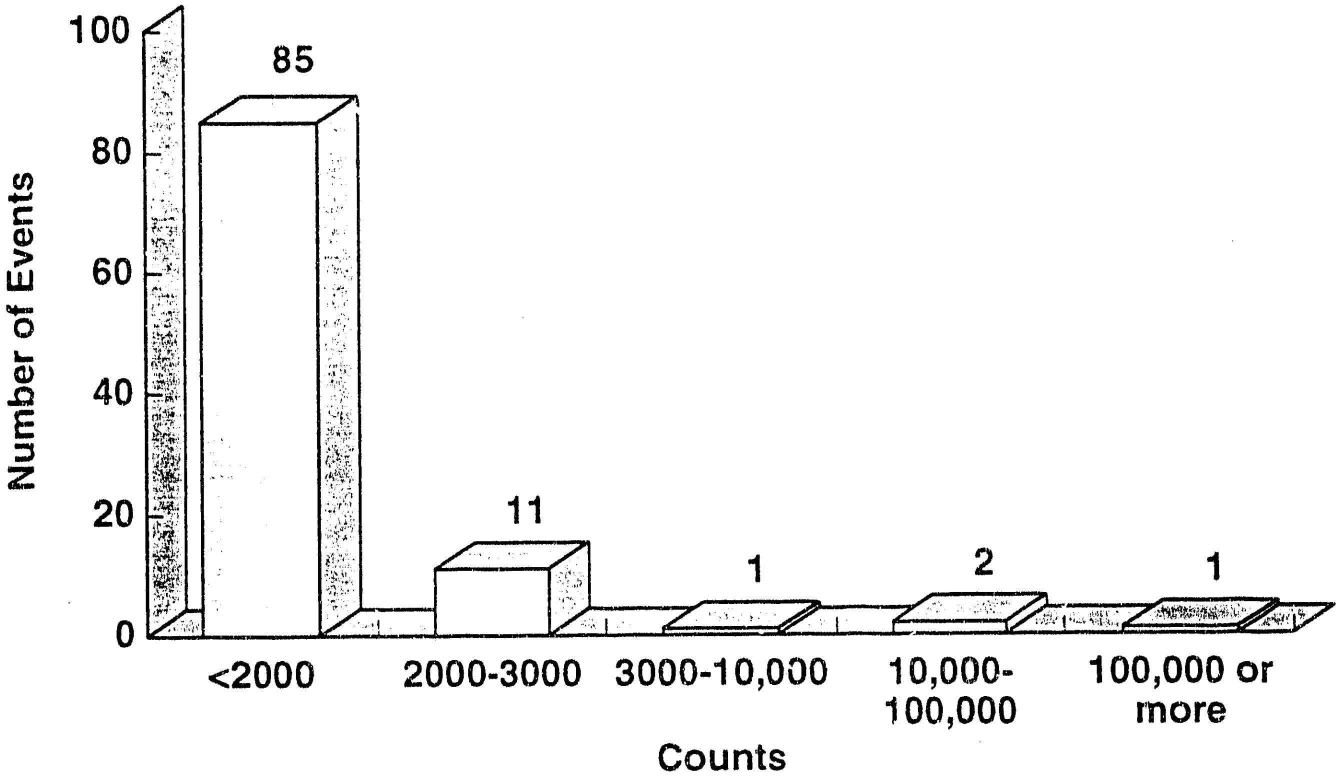
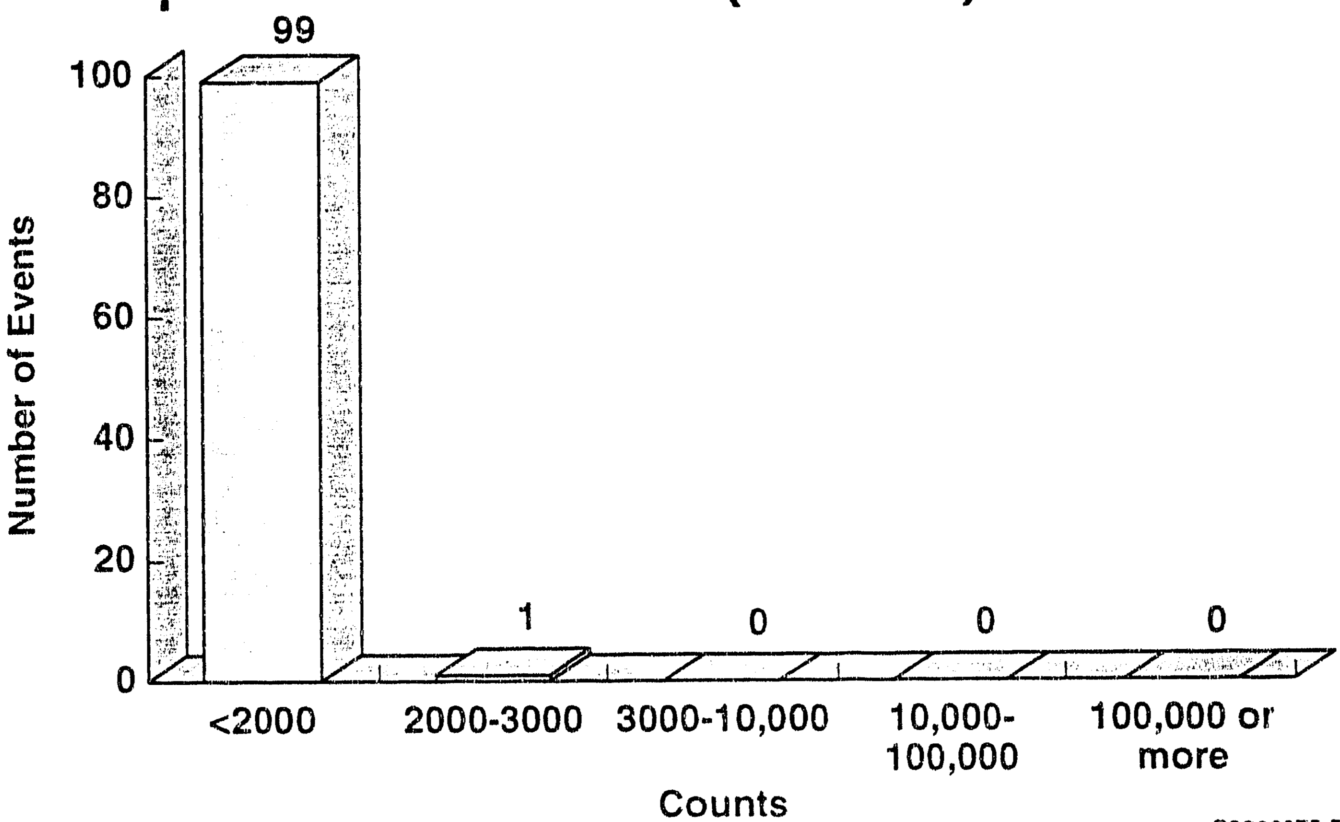
Pulse Height Distribution from 1 cSv (1 Rem) Dose Equivalent of Fast Neutrons



Pulse Height Distribution from an Unexposed Dosemeter (control)



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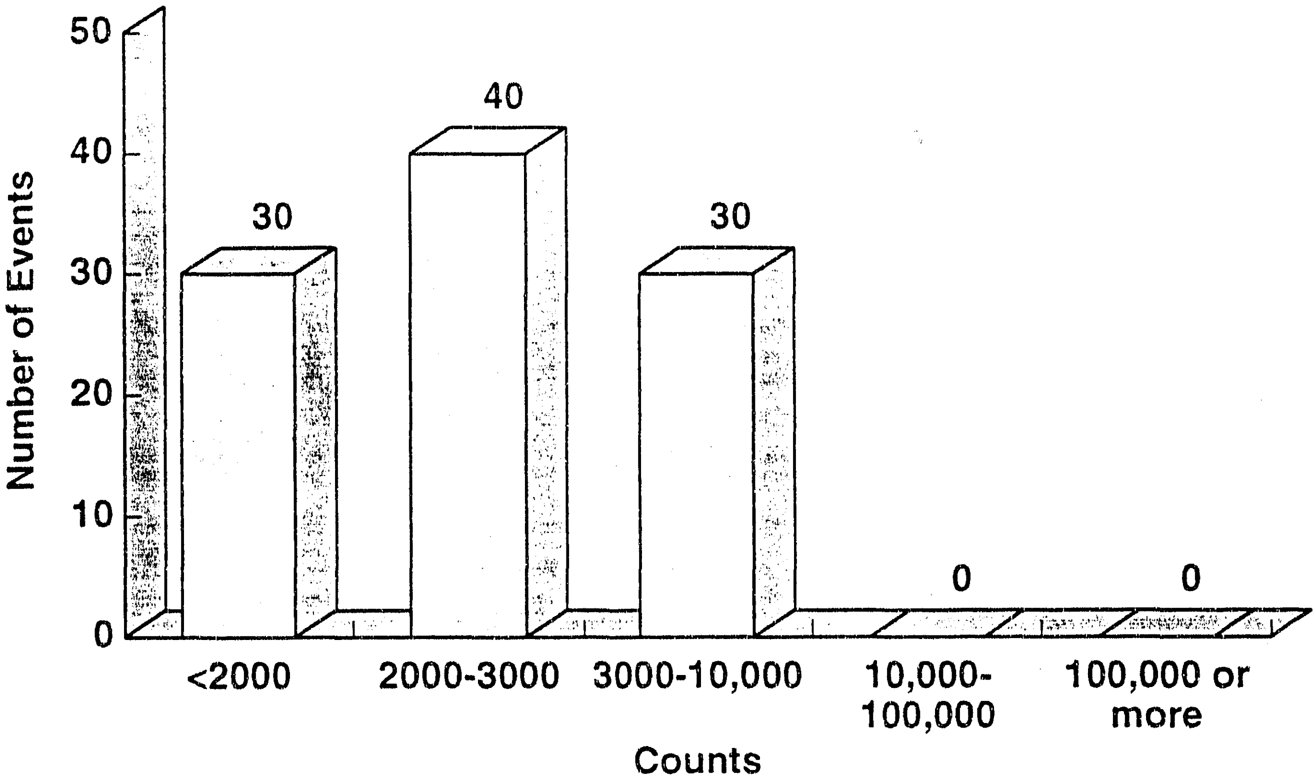
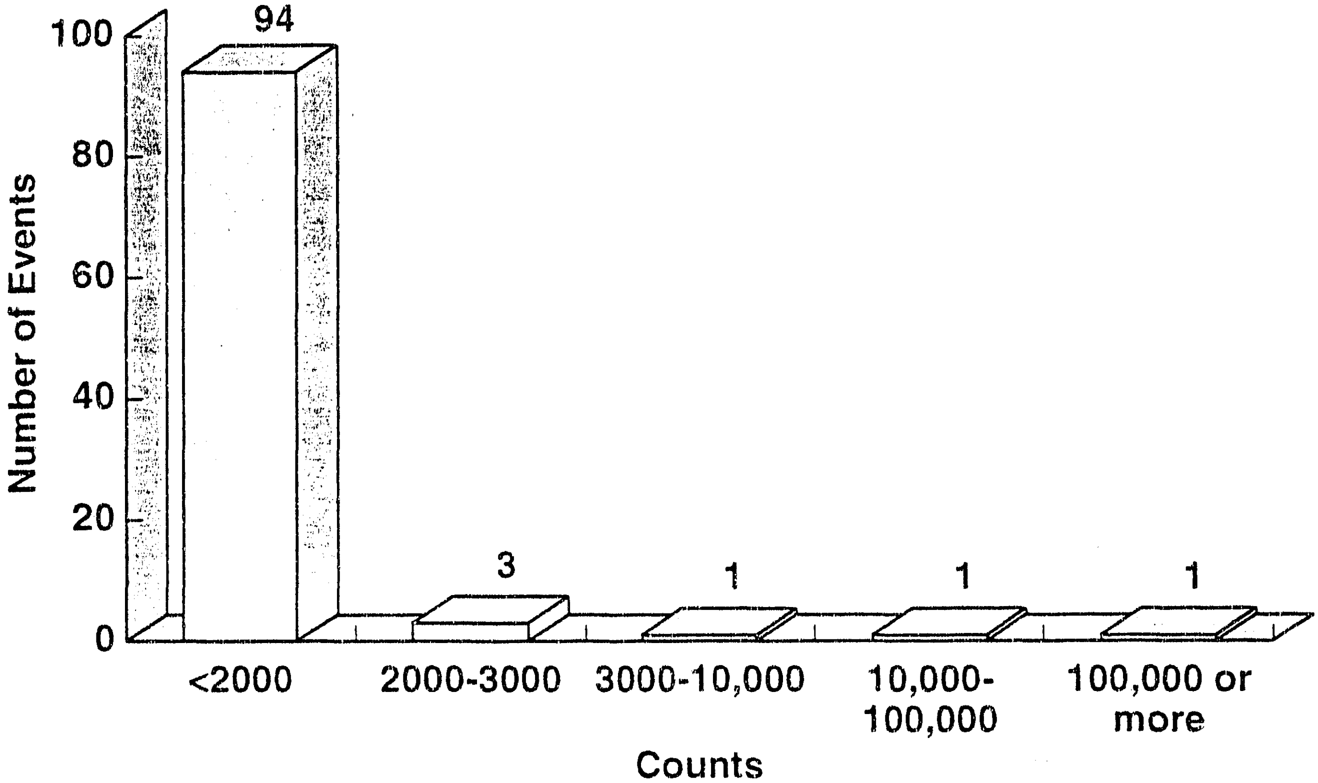
Pulse Height Distribution from 100 cSv (100 Rem) Dose Equivalent of Fast Neutrons



Pulse Height Distribution from an Unexposed Dosemeter (control)



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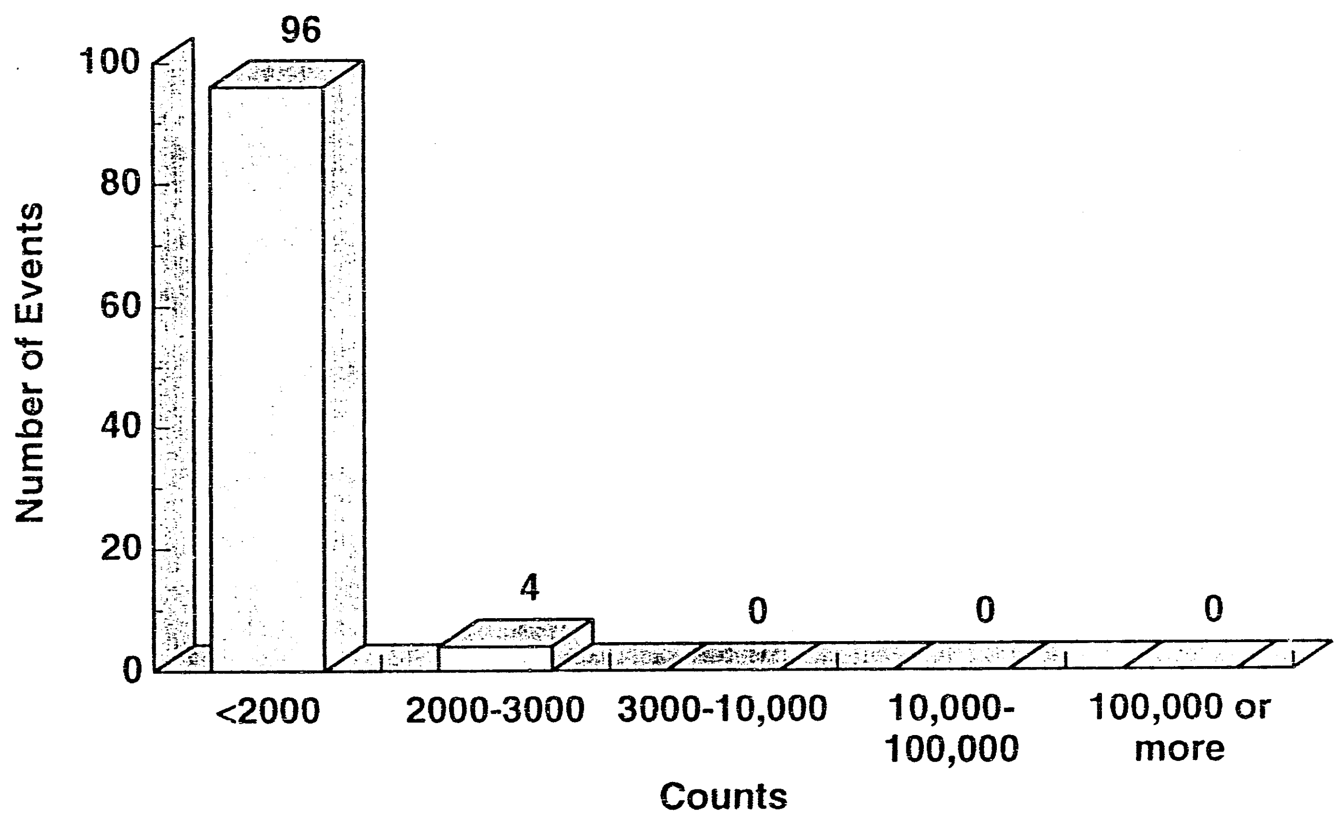
Response of CaF_2:Mn/Polyethylene Dosemeter to 6 cSv (6 Rem)



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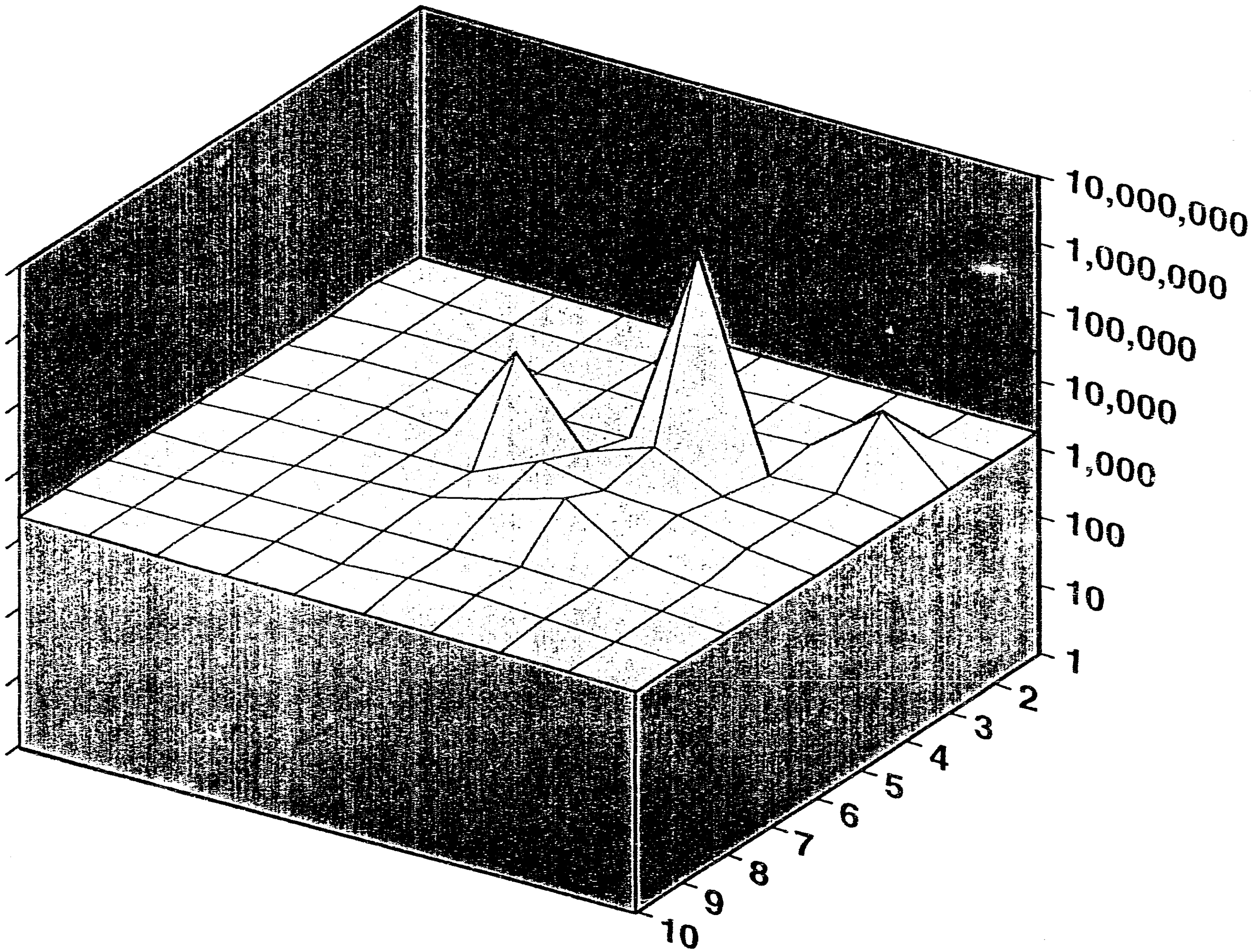
COSL emission from 100 sites on a polyethylene/ CaF_2:Mn dosemeter exposed to a 5.5μCurie ^{60}Co particle. The particle had a major axis diameter of 50 microns.



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