Room-Temperature Semiconductor Detectors for in Vivo Monitoring of Internal Contamination

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In vivo monitoring of low-energy X- and γ-ray emitters has always been a difficult task, primarily because of lack of accuracy and the high detection limits of classical techniques. Various types of PIN diodes (diodes with a large intrinsic zone) were tested in the Radiation Protection Department of the Studie Centrum voor Kernenergie, Centre d'étude de l'Energie Nucléaire (Mol, Belgium) in the measurement of radioactive body burden by direct methods. Current research is oriented toward the use of room-temperature diodes for the detection of low-energy photons escaping the body. In this paper, a new counting technique that involves a portable jacket containing the diodes is described. The system uses silicon diodes and is used out of a shielding room in order to be near the contamination. With this method rapid analysis and long counting times are possible, stress is reduced, and medical treatment can be optimized. CdZnTe detectors were also evaluated for this measurement technique but this type of detector is better adapted for counting inside a shielding room. The improvement of the accuracy of the measurement, taking into account the effect of the ribs, is described here, as well as the associated electronics necessary for this type of counting. — Environ Health Perspect 105(Suppl 6):1423–1426 (1997)

Key words: total body counting, in vivo measurements, room-temperature semiconductor detectors, internal contamination, direct methods, radioactive body burden, silicon PIN diode, CdZnTe diode, torso phantom, contaminated wounds, thyroid

Introduction

Measurement of low-energy γ-ray emitters in the body by the direct method always has been difficult because of the strong attenuation of the photon flux in different tissues. For this reason measurement is possible only in a limited number of organs and tissues e.g., in the lungs, the thyroid, and in wounds. A good assessment is not possible using standard techniques. The use of a large detector with a high efficiency such as a phoswich (a NaI[Tl] optically associated with a Cd[Tl]) is only valuable when the contamination in the lungs is homogeneously distributed and distribution is similar to the activity distribution in the phantom used for calibration. Indeed, a detector that only measures background will increase the detection limits without bringing information. For example consider two small diodes placed on a lung; only one diode sees a signal S1. The other measures a zero net signal (S2 = 0). If we consider only diode 1 for the calculation, the result will be correct. If we consider both diodes, which together have a higher efficiency and which measure the same net signal (S1 + 0), the final result will underestimate the burden. The purpose of this study is to show that several small diodes can be used to answer different questions related to internal contamination. A second goal of the research is to show that two types of available diodes are best suited for in vivo measurements: room-temperature silicon diodes, which allow certain types of measurement of 241Am in the lungs out of a shielding room by long-duration counting, and CdZnTe (CZT) diodes, which can be used in groups to reach detection limits obtained with classic hyperpure germanium (HPGe) detectors to assess 241Am contamination of the lungs. These diodes are useful for in vivo detection of 125I, 103Pd, 109Pd, 133Ba, and even 137Cs.

Materials and Methods

In a previous study (1), different types of silicon PIN diodes (diodes with a large intrinsic zone) were compared for their efficacy in whole-body counting. Research in this study was limited to two types of diodes: a 500-μm thick Hamamatsu S3590-05 silicon PIN diode (Hamamatsu, Japan) and a 1-mm thick CZT detector (model ev 180-9-8-4-SD from eV Products, Saxonburg, PA). The objective was to determine the parameters of the counting devices (e.g., volume of the diode, thickness of the depleted zone, bias voltage, diode containment) that would optimize the counting capabilities of these two diodes in specific contamination cases. The experimental work was aided by a home-made humanoid torso phantom containing a true thoracic cage with two foam volumes to simulate the lungs. A photograph of the torso, with the front Lucite plate removed to show the thoracic cage, is presented in Figure 1. Although manipulation of this device is difficult, especially when radioactive charges must be changed, its advantages are important: It has a true (corrected) skeleton; the cartilage is represented (with leather); it contains 40K homogeneously distributed; and it is possible to simulate all types of radioactive charge distribution. The absorption parameters of this phantom were controlled by loading it with different charges of 241Am homogeneously distributed and by measuring the phantom

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Figure 1. Homemade phantom made of Lucite with a true human skeleton. The ribs of the skeleton have been adapted with 0.2 mm tin foil to give the correct absorption properties at 60 keV.
with a germanium measuring device that is currently used for routine measurements (2). Correction of this phantom was required because the skeleton portion does not contain blood or bone marrow and does not absorb a 60-keV photon in the same way that a living one does. For this reason, the ribs have been covered with 0.2 mm tin foil to give them the correct absorption properties. Figure 2 displays a vertical scan of the torso measured with the CZT detector after the placement of tin foil on each rib. This graphic, compared to the calculated scan (1), indicates that the phantom is correctly adapted to simulate an actual thoracic cage. The abscissa of this graph represent the vertical displacement in meters with respect to an arbitrarily chosen level. The peaks on each curve represent an interrib position where absorption is minimal. Comparison of the various curves corresponding to different energy zones indicates the importance of diffusion of the photons in tissues of the body. This is why it is necessary to consider a broad range of energy and why the resolution of the diode is of secondary importance.

The bias voltages were chosen to reach full depletion of the junctions (125 and 100 V, respectively, for silicon and CZT instead of the recommended 100 and 60 V). This technique allows a higher efficiency and a stable response and does not affect the resolution or the lifetime of the semiconductor even if an increase of the leakage current intensity is observed.

Results

Wound and Thyroid Measurements

Table 1 presents detection limits for two cases: 125I in the thyroid and 241Am in the wound. Measurements were performed with three different silicon diodes: a Hamamatsu S3590-05 diode with a 0.81-cm² active area and a 500-µm depletion layer; a passivated implanted planar silicon diode PD-16-300-AB with a 4.6-cm² area and a 300-µm depletion area, kindly lent by Canberra Semiconductor, Olen, Belgium; and a SFH 217 PIN diode (Siemens, Nixdorf, Germany) with a 0.0095-cm² area and a 200-µm depletion thickness. Even when they are used as a single diode both detectors are suitable for special types of counting, e.g., wound and thyroid, thus meeting the recommendations of the International Committee on Radiological Protection (3) and various regulatory requirements. The CZT diode was also tested for measurement of contaminated wounds, as described later in "Applications."

Lung Measurement

As mentioned previously (1), the counting of lungs contaminated with 241Am must be conducted with an array of diodes to improve efficiency and keep the counting duration within an acceptable limit for human controls (maximum 1 hr) when counting in a counting room. The results of this study can be summarized in two points. First, the counting of lungs with CZT diodes can bring the detection limits equivalent to germanium detectors in a 1-hr counting time if the measurement is performed inside a shielding room and if the number of CZT diodes is increased to 48 units. Second, when silicon PIN crystals are used, measurements could be achieved outside of a shielding room. Thus the counting time can be raised to 24 hr or more (not unreasonable if we consider the use of an electrocardiogram in real time). In such cases the number of diodes could be reduced to about 50 U (Table 2) to reach the detection limits required by law in certain types of contamination (i.e., pure or high concentration of 241Am).

Use of the Detector Jacket with Small Diodes

The accuracy and efficiency of the measurement can be improved by using an

Table 1. Critical levels and detection limits Lc/Ld (Bq) for different detectors and for two types of measurements: 125I in the thyroid and 241Am in a superficial wound.

| Radionuclide | Diode 1, S3590-05 | Diode 2, PD-16-300-AB | Diode 3, SFH-217 |
|--------------|------------------|-----------------------|------------------|
| Active area  | 0.81 cm²         | 4.6 cm²               | 0.01 cm²         |
| Thickness    | 500 µm           | 300 µm                | 200 µm           |
| Thyroid, 2000 sec | 20/50         | 40/80                 | 720/1500         |
| Wound, 2000 sec | 241Am         | 9/20                  | 7/15             |

Table 2. Critical levels and detection limits Lc/Ld (Bq) for three type of detectors in different numbers and for different counting times.

| Detector | 1 hr | 24 hr | 24 hr |
|----------|------|-------|-------|
| Two LEGe, total active area=5775 mm² | 4/10 | 1250 | 1250 |
| Silicon inside a shielding room, 500 µm | 1 diode, 300/950 | 1 diode, 60/130 | 48 diodes, 40/90 |
| Silicon outside of a shielding room, 500 µm | 1 diode, 1500/3000 | 1 diode, 280/580 | 48 diodes, 45/95 |
| CZT inside a shielding room, 1 mm | 48 diodes, 4–10 Bq | — | — |

LeGe, low energy germanium. Values indicated are extrapolated from measurements with one diode.
Associated Electronics

To manage the signals coming randomly from several detectors and send them to a unique spectrometric device, special electronic circuits must be used. Currently there is no circuit available that is used in conditions adequate for X- and γ-ray spectrometry; however, different laboratories are developing such electronics in integrated circuit or in discrete form (4–6). Two other circuits were tested with positive results.

The first circuit (made by Velleman, Brussels, Belgium, catalog no. K8000) is based on 10C interfaces (PCF8591 and PCF8574 from Philips, Eindhoven, Holland). It allows the selection of diodes one at a time among a whole series using a computer program named Select, which we wrote to follow the requirements of the measurement. This device, which has been positively tested, cannot be used for the counting of randomly appearing events (spare readout), but it can be used to study metabolism by measuring the quantities of radioactive burdens in different organs.

The second circuit uses the mixer routers frequently operational in nuclear spectrometry (model 8222B from Canberra, Olen, Belgium). It contains four analog inputs able to receive signals from four silicon diodes. An example of spectra collected with this system is presented in Figure 3 (counting time = 24 hr), where three S3590-05 silicon PIN diodes have been connected. Figure 3, where the respective backgrounds are also displayed, indicates the different shapes of the collected spectra due to the different positions of the diodes on the phantom. Figure 3 also indicates that diffusion in the tissues produces an important deterioration of the spectra so that the resolution of these diodes is not of primary importance. The detection limits of this mockup are $L_C = 380$ Bq and $L_D = 760$ Bq for a collection time of 24 hr. [Critical level $L_C$ and detection limit $L_D$ are defined according to Currie's concepts: $L_C = 2.33\sqrt{N_B}$ and $L_D = 2.71 + 4.65\sqrt{N_B}$ where $N_B$ is the total count measured in the investigated zone of the background spectrum.]

Detection of Contamination Hot Spots

After the acquisition time, the sum spectrum (summation of the different spectra) can indicate a light contamination or a null result whereas examination of each spectrum separately might reveal a high concentrated contamination. For this reason, the detector jacket also can be used for detection and localization of hot spots, which is not possible with a large phonswich crystal. Experimental measurements indicate that detection limits are $L_C/L_D = 44/88$ Bq for a collection time of 24 hr with silicon diodes and 15/30 Bq for the same counting time with CZT (for measurements outside of a shielding room). An example of spectrum of a hot spot with CZT is shown in Figure 4. In this case, the use of CZT outside of a shielding room is appropriate.

Applications

The devices described here can be used in industry workers to detect eventual high lung contamination by $^{241}$Am before using classic techniques in shielding rooms. The devices are also useful for the detection of $^{125}$I, $^{133}$Ba, $^{137}$Cs in wounds or in organs when the organs are not too deep in the body (thorax, lungs, liver, and bladder). The best application of these devices is the measurement of skin contamination when the contaminant is a low-energy photon emitter, which is the case in many applications of nuclear medicine e.g., diagnostics and therapy. The tracer can be labeled with one of the radionuclides mentioned, a valuable application in metabolism followup. A single silicon PIN diode is well adapted for counting $^{252}$Cf in thyroid burden or metabolism studies.

In our laboratory we assessed the capabilities of the CZT diode measuring $^{241}$Am in a wound using a finger model with high-density polystyrene plates of (C$_8$H$_8$)$_n$—a material equivalent to muscle for absorption and diffusion of X-rays. A 2.5-mm deep incision was made in this plastic finger, in which 15 (± 0.5) Bq $^{241}$Am was placed. Measurement with an HPGe crystal resulted in a count of 12 Bq in 10 min, whereas the CZT diode detected 13.7 Bq $^{241}$Am in 6 min. This count assumes that no other γ-ray was present from $^{241}$Am in the 60 keV energy zone. This technique can be used advantageously instead of one described previously (7). Finally, because they do not require liquid nitrogen, silicon diodes and CZT can be used

![Figure 3. Three spectra of $^{241}$Am homogeneously distributed in the lungs and measured with three silicon (S3590-05) diodes working in a random mode. Counting time = 86,400 sec. Total activity = 102,300 Bq $^{241}$Am. The sum of background is also presented (bold line).](image-url)
in environmental studies by using the first electronic circuit described in this paper.

**Discussion and Conclusions**

Room-temperature silicon diodes and CZT can be used to replace HPGe detectors in many cases for assessment of actinides in the lungs or other radioactive burdens in organs. The number of diodes should be chosen according to the type of measurement. Because of their size and weight the diodes can be attached to a detector jacket worn by the user.

An important advantage of the detector jacket is the elimination of systematic error in the counting because the diodes are fixed on the torso. The close proximity of the diodes with respect to the measured organ increases the counting efficiency. The shielding room is not required with silicon diodes in the energy range of 20 to 100 keV. Long counting times are possible with the jacket; because the diodes are not sensitive to radon variations there are no fluctuations in the background rate in the considered energy range. The counting can start immediately after a suspected contamination, which is an important psychological aspect. Discrete square diodes are preferred to long strip detectors because they can be selected after counting to reduce the background and the detection limits.

The concept of a portable detection jacket can be applied to different types of room-temperature detectors, suggesting applications in radiation protection and nuclear medicine; but electronics for the random readout of an array of diodes has yet to be developed. An example of a design for a detector jacket is shown in Figure 5. Silicon and CZT as single detection elements provide interesting applications in the control of $^{125}$I in the thyroid and $^{241}$Am (from mixed oxide fuel particles) in wounds.

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