SKIN DOSE EQUIVALENT MEASUREMENT FROM NEUTRON-DIFFICIENT ISOTOPES

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Skin Dose Equivalent Measurement from Neutron-Deficient Isotopes

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Abstract

Neutron-deficient-isotopes decay via positron emission and/or electron capture often followed by x-ray, gamma-ray, and 0.511 MeV photons from positron annihilation. For cases of significant area and/or personnel contamination with these isotopes, determination of skin dose equivalent (SDE) is required by 10CFR835. For assessment of SDE, we evaluated the MICROSPEC-2™ system manufactured by Bubble Technology Industries of Canada which uses three different probes for dose measurement. We used two probes: 1) the X-probe which measures lower energy (4 - 120 keV) photon energy distributions and determines deep dose equivalent, SDE and dose equivalent to eyes, and 2) the B-probe which measures electron (positron) energy distributions, and determines skin dose equivalent. Also, the measured photon and beta spectra can be used to identify radioactive isotopes in the contaminated area. Measurements with several neutron-deficient sources showed that this system provided reasonably accurate SDE rate measurements when compared with calculated benchmark SDE rates with an average percent difference of 40%. Variations were expected because of differences between the assumed geometries used by MICROSPEC-2 and the calculations when compared to the measurement conditions.

Introduction

As a contractor to the Department of Energy (DOE), Los Alamos National Laboratory (LANL) must comply with 10CFR835 (DOE 1993). Section 10CFR835.205 contains specific requirements for non-uniform exposures of the skin. The DOE Radiological Control Manual (DOE 1994a) and implementation guidance for 10CFR835 (DOE 1994b) recommend an action level of 1 mSv (100 mrem) for detailed evaluation of skin dose equivalent (SDE) resulting from a contamination incident. This action level for evaluation of the SDE has been implemented at LANL.

The literature is replete with works that address SDE assessment of beta emitting radionuclides. However, assessment of beta doses is complicated by many factors including variability in methods for volume averaging of the dose, application of fluence-to-dose equivalent conversion factors for broad parallel beam to point-isotropic geometry, and presence of air gaps and shielding provided by protective clothing. For example, the NCRP recognized the complexity of SDE assessment for point-isotropic sources as reflected in the recommendation that exposure to small area sources, less than 1 mm in diameter, be based on integrated activity of the beta source rather than tissue dose (NCRP 1989).

At LANL, assessment of skin dose equivalent resulting from radioactive contamination is normally done in a multi-step process. First, radioactive contamination is screened in the field using a hand-held survey instrument, typically a detector with a thin window GM pancake probe.
However, the usefulness of the GM detector for complete SDE assessment is limited in that: 1) it
does not measure the spectrum so it cannot identify radioactive isotopes, 2) the detector
efficiency depends on beta and photon energy, and 3) it cannot distinguish between low-energy
photons and positrons. Therefore, isotopic identification is obtained using gamma spectroscopy.
Isotopic identification is critical when there exists the potential for contamination by multiple
unknown beta and gamma emitting radionuclides. Finally, for contamination levels exceeding the
radionuclide-specific action levels based on initial screening, dose estimating is performed using
computer models such as VARSkin MOD2 (Durham 1992) or by using published fluence-to-
dose equivalent conversion factors (Chartier et al. 1996; Cross et al. 1992, ICRP 1987).

The MICROSPAC-2 (BTI 1995) manufactured by Bubble Technologies Inc. (BTI) is a portable
spectroscopy survey system capable of doing simultaneous isotopic identification and dose
equivalent measurements. The MICROSPAC-2 performs spectrum folding into a fluence-to-dose
equivalent conversion function to determine dose equivalent rates for both photon and beta
radiation at skin, eye and deep dose depths. The instrument can be operated in the field and its
unique capability could obviate the need for separate gamma spectroscopy and subsequent dose
assessment with computer codes as is currently done. Its appropriateness for both spectroscopic
and dosimetric applications has been shown for electron emitting radionuclides (Hsu et al. 1997).
Studies were initiated to assess the usefulness of the system for doing simultaneous spectroscopic
identification and dose assessment of positron emitting contaminants in the field.

The radionuclides used in this study were $^{22}$Na, $^{68}$Ga (in equilibrium with $^{68}$Ge), $^{82}$Rb (in
equilibrium with $^{82}$Sr), $^{85}$Sr, and $^{88}$Y. These are associated with medical isotope production at
LANL. Incidents of skin and personal clothing contamination with these radionuclides have
occurred as single isotopes and as mixtures with multiple radionuclides. Additionally, a nominally
pure gamma emitting radionuclide $^{73}$As was also studied.

**Methods**

Neutron-deficient sources were prepared in a nuclear chemistry laboratory. A solution of each
radionuclide was prepared and spotted on a thin mylar sheet. After the material had dried,
another layer of mylar was laid on top, sandwiching the source between the two mylar sheets.
These mylar sheets were then stretched across an 6 cm x 8 cm x 0.2 cm thick aluminum mounting
plate that had a circular hole (diameter = 4 cm) in the center of the plate. The radionuclide
sources were approximately centered in the open part of the mounting plate. This source
arrangement with the thin mylar was found to not significantly affect measurement of the positron
and photon radiations and it minimized possible backscatter. Source activities were measured
using a calibrated high-purity germanium detector.

SDE rates for each source were measured using the MICROSPAC-2 coupled to the X-probe (for
low-energy photons) and B-probe (for positrons). To simulate skin exposures, the sources were
placed close to the detector (about 0.5 cm) with the source approximately centered along the
open face of the detectors. To reduce backscatter that could have artificially elevated the dose
rate measurements, the sources were held in front of the detector and were supported from the
side by polystyrene foam. The count times ranged from 60-300s. Measurements with a GM
pancake probe were also made under similar geometry. These measurements were made to
determine the efficacy of using these instruments as a qualitative screening tool for estimating SDE. Besides SDE rate measurements, beta and gamma spectra were recorded using the MICROSPEC-2 for radioisotope identification.

To estimate the accuracy of the MICROSPEC-2 and the pancake probe measurements for estimating SDE rates, comparisons were made with calculated SDE rates. These calculated values were determined by folding positron and photon energy spectra into published fluence-to-dose equivalent conversion values (Chartier 1996, ICRP 1987). These SDE calculations did not include the contribution from the 0.511 MeV annihilation photons because they were not expected to contribute significantly to the SDE and determining the fluence of these photons is highly complex requiring Monte Carlo calculations using precise modeling of the exposure conditions.

Positron and photon fluence rates were determined based on source activities and solid-angle corrections. Source-specific corrections for the solid angle were needed to determine fluence rate from decay rate for each source. For this, measurements were made of the area containing the activity for each source. Then using Monte Carlo techniques, the fraction of radiations per decay that contributed to the skin dose equivalent was determined for each source. The assumed source/skin geometry included the following: 1) a 0.5 cm gap between the source and the skin (about the same source/detector distance for the MICROSPEC-2 measurements, 2) a circular source with a homogeneous distribution of activity, 3) and the source was centered over a circular area of irradiated tissue that had a diameter equal to the area of the source. Estimates of uncertainty in the activity per area and solid-angle correction factors were made and propagated through the calculations.

Measurement results from the MICROSPEC-2 and the pancake probe were compared to the calculated SDE rate predictions. Correlations using least-square linear regression techniques were determined and the residuals were analyzed to determine the adequacy of the linear models.

Results
Radionuclides and their respective decay modes, measured activities, source areas, and average count rates (measured by a GM pancake probe) are provided in Table 1. Table 1 also shows the results of the SDE rates from MICROSPEC-2, the calculated doses, and their uncertainties. In all cases the MICROSPEC measurements were lower than the calculated SDE rates with percent differences ranging from 9% to 80%. The uncertainties of the calculated SDE rates were mostly influenced by the estimated error in solid-angle correction factors (assumed 10%) and the error in the measurement of the source areas. Standard deviations for the MICROSPEC-2 measurements are not given because only two measurements were obtained for each source, but the averages are shown in Table 1 and the results of the two measurements were generally precise.

Correlations between the calculated SDE and the MICROSPEC-2 measurements and the count rates are presented in Fig. 1 and Fig. 2, respectively. A high degree of correlation was found between the MICROSPEC-2 measurements and the calculated SDE rates ($r^2=0.99$) with an intercept of $0.034 \pm 0.02$ and a slope of $1.13 \pm 0.03$. A weaker correlation was found between the calculated skin dose equivalent rates compared with the count rate values ($r^2=0.68$) with an
intercept of -0.04 ± 0.3 and a slope of $6 \times 10^{-6} \pm 2 \times 10^{-6}$. Further analysis showed that residuals for both correlations are scattered about the regression line and appear to be independent of the predicted values of the regression. This suggests that a linear model for these correlations is appropriate. Finally, the ratio of the calculated SDE rate to the MICROSP2-2 measured values were found to be independent of activity but is possibly related to the area of the source, suggesting a geometry dependence. This relationship was not linear, but the ratio was greatest for the smallest source area and then varied between 1.9 and 1.1 for the remaining sources areas.

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Decay modes (including progeny)</th>
<th>Activity (kBq)</th>
<th>Source area (cm²)</th>
<th>Count rate by GM Pancake (cpm x 10³)</th>
<th>MICROSP2-2 SDE Rate (mSv hr⁻¹)</th>
<th>Calculated SDE rate (mSv hr⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>⁸⁸Y</td>
<td>β⁺, γ, x-ray</td>
<td>5.1 ± 0.2</td>
<td>1.0 ± 0.4</td>
<td>5.2 ± 0.1</td>
<td>0.05</td>
<td>0.08 ± 0.03</td>
</tr>
<tr>
<td>⁷³As</td>
<td>EC, γ, x-ray</td>
<td>3.7 ± 0.1</td>
<td>0.4 ± 0.2</td>
<td>5.3 ± 0.1</td>
<td>0.01</td>
<td>0.07 ± 0.03</td>
</tr>
<tr>
<td>⁸²Sr</td>
<td>β⁺, EC, x-ray</td>
<td>4.7 ± 0.2</td>
<td>1.6 ± 0.5</td>
<td>260 ± 0.5</td>
<td>0.86</td>
<td>0.94 ± 0.28</td>
</tr>
<tr>
<td>⁵²Na</td>
<td>β⁺, γ</td>
<td>8.9 ± 0.3</td>
<td>2.7 ± 0.6</td>
<td>270 ± 0.5</td>
<td>1.96</td>
<td>2.27 ± 0.55</td>
</tr>
<tr>
<td>⁶⁸Ge</td>
<td>β⁺, EC, x-ray</td>
<td>2.4 ± 0.1</td>
<td>5.4 ± 0.9</td>
<td>61 ± 0.3</td>
<td>0.08</td>
<td>0.15 ± 0.03</td>
</tr>
</tbody>
</table>

**Discussion**

Overall, we concluded that MICROSP2-2 system provided reasonable estimates of SDE rates for the studied neutron-deficient sources, but could underestimate the SDE. This underestimation could be due to geometry considerations. Because the MICROSP2-2 measurements were highly correlated with calculated SDE rates, calibration may be possible and could provide highly accurate SDE measurements. We also found advantage in using the beta and photon pulse-height distributions for radionuclide identification. The system was easy to use, and was light and small enough to carry in the field. Finally, measurements with MICROSP2-2 can be made quickly compared with more costly laboratory measurements and is more accurate than usual health physics radiation survey instruments such as the GM pancake probe which requires a correction factor for each specific radionuclide.

Dose calculations measured by the MICROSP2-2 system assumed broad-beam geometry, and differences between calculations and measurements are expected for sources with nonuniform activity distributions and point-isotropic emissions. We found some evidence that the percent difference between calculated and measured values might be influenced by the size of the source area. Analysis of the percent difference as a function of source area showed that the largest difference was found for the pure photon emitter ⁷³As which had the smallest source area and for the ⁶⁸Ge source which had the largest source area. However, because of our small data set, the evidence of this relationship is not conclusive. Also, we found ⁷⁴As and ⁸³Sr contamination in the ⁷³As and ⁸²Sr sources, respectively. However, these contaminates were not expected to contribute significantly to the measured SDE rates because of relatively small amount of ⁷⁴As compared to ⁷³As (<2 percent by activity) and the fact that ⁸³Sr decays to stable ⁸⁵Rb with emission of high energy (0.514 MeV) x-rays only. More detailed studies looking at the response of the MICROSP2-2 system as a function of varied source geometries, beta and photon energies, and for mixtures of isotopes are needed. In addition, comparisons with predictions with other computer codes such as VARSKIN MOD2 would also be valuable.
References


Figure 1. Comparison between calculated SDE rates and those measured by MICROSPEC-2.

Function: \( y = 0.034 + 1.127x + \epsilon \)

Figure 2. SDE rate as a function of count rate. Measurements were made with a GM pancake probe.

Function: \( y = -0.041 + 6.206 \times 10^{-6}x + \epsilon \)