

REPORT FOR AD HOC COMMITTEE ON STATUS
AND TRENDS IN BIOASSAY OF INHALED
HEAVY ELEMENTS BY PHOTON COUNTING

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COUNTING HARDWARE I-A

Currently, phoswich detectors are the most prevalent type of detectors in use for assessing lung depositions of heavy element low-energy photon emitters. These detectors, along with their associated instrumentation, have proven their reliability for routine monitoring. Most phoswich detector systems are capable of detecting about one-half lung burden of ^{238}Pu or about one and one-fourth lung burdens of ^{239}Pu on an average individual who has a uniform distribution of plutonium in the lungs.¹

Most all the large area NaI detection arrays have been replaced by the phoswich detectors. The NaI detectors are similar in basic characteristic to the phoswich, however, the large area NaI detectors exhibit higher background and sensitivities at the lower energies (< 70 keV) used for assessing inhalation exposures of heavy elements.

Some lung counter facilities use proportional counters for assessments^{2,3,4}. Those in current use have anti-coincidence chambers to reduce background counts and improve the sensitivity. The inherent, absolute counting efficiency of proportional counters is lower than that for most other types of detectors and decreases as photon energy increases. Their detectable limit is generally higher than the phoswich detectors. They are at a disadvantage at higher energies when ^{241}Am is used to assess the lung deposition of ^{239}Pu . However, one rather significant advantage of proportional counters is their high resolution capability which can be used to distinguish between external and internal contamination.

Semiconductor detectors are only practical when used in arrays and, therefore, are the most expensive detection system available. Resolution from these detectors is excellent. There is currently only one such system that was constructed specifically for bioassay of inhaled heavy elements⁵. This system was installed in 1976 and consists of 2 arrays of 4 intrinsic germanium detectors, where each of the 8 has an area of 10 cm^2 . Preliminary results indicate that

backgrounds per unit area are excellent and resolution (< 700 eV at 60 keV) is superior to other types of detectors. Absolute detection efficiency should be essentially 100% for photons below 60 keV.

A comprehensive report of the semiconductor detector system should be forthcoming in 1977. It should be compared to the phoswich detector system which is currently the best type of system available.

REFERENCES I-A

1. Proceedings of the Workshop on Measurement of Heavy Elements In Vivo, held at Battelle Seattle Research Center, Seattle, Washington (June 24-25, 1976).
2. Argonne National Laboratory, Radiological and Environmental Research, 9700 South Cass Avenue; Argonne, Illinois 60439.
3. Atomic Energy Research Establishment, Harwell (AERE), Environmental and Medical Sciences Division, AERE Harwell, Didcot, Berkshire, OX11 0RA, Great Britain.
4. Atomic Energy Establishment, Wirtfrith, Dorset, Great Britain.
5. Rocky Flats Plant, Rockwell International, Post Office Box 888, Golden, Colorado 80402.

BACKGROUND PREDICTION I-C

Whenever a radioactive worker is counted and the gross results are significantly above those from persons who have never worked with radioactivity, an inhalation of radioactivity is highly possible. The amount of background, which is part of the gross count, is handled in a variety of ways when background data are not available on the particular subject in question.

Some laboratories determine the background by averaging background counts from 2 or more individuals of similar physical build who have never worked with radioactive materials. This background is then subtracted from the gross count to give a net count which is the result of the inhaled heavy element. The accuracy in determining the correct background with this method is estimated at $\pm 15\%$ (2σ). This method of background prediction does not account for variations from day to day or month to month such as room background or a person's diet. Therefore, it is used most successfully by laboratories where room backgrounds are not significantly affected by atmospheric changes, nearby reactors, plant effluents, or diet uptakes of ^{137}Cs .

Other laboratories arrive at a person's background by correlation of the subject's count in the 75-100 keV energy band to that in lower energy bands of interest. The subject of interest is, therefore, used for his own background and accuracy is believed to be as good as $\pm 10-12\%$ (2σ).

The effect of any error in estimating a subject's background is a function of the amount of the inhaled heavy elements. When the lung deposition approaches the detectable limit, an error in background can affect the final assessment by as much as $\pm 50\%$ (2σ). In cases where the deposition is well above the detectable limit, (gross count > 2 times background) the error in an assessment resulting from an incorrect background ($\pm 10\%$) is only about $\pm 10\%$ (2σ). However, when the levels approach the detectable limit, this error is quite significant.

This error could conceivably be reduced by a factor of 2 with a concentrated effort in the area of predicting low energy person backgrounds from various higher energy photon spectra.

PRECISION AND ACCURACY I-G

The possibility of error in making an assessment of inhaled heavy elements is rather large when the uptake measured approaches the detectable limit of the lung counter being used. Most facilities find it necessary to qualify their results by stating various hypotheses that were used in developing a method of making the lung assessment. Some of the hypotheses that appear most frequently are: the lungs contain a uniform distribution of activity; ultrasonic chestwall measurements are accurate; the subjects physical characteristics are closely approximated by the phantom used for calibration; the subjects background can be predicted with some precision; and the isotopes(s) being counted is the only one(s) yielding the data.

Since there are as many specific methods of calibration as there are laboratories, error analysis of the known factors must be handled differently at each facility. The errors involved in calibration were a significant part of a recent in vivo conference¹. The final session was followed by a general discussion concerning the effect of errors on the assessment of a lung burden (16 nCi) of ²³⁹Pu. Five major items are listed below with the estimated effect assuming a typical phoswich detector system.

- | | |
|----------------------------------|-------------------------|
| 1. Background Person | - ±50% (2 σ) |
| 2. Phantom Lung | - ± 5% (2 σ) |
| 3. Chestwall Thickness | - ±40% (2 σ) |
| 4. Activity Distribution in Lung | - ±70% Systematic Error |
| 5. Counting Statistics | - ±50% (2 σ) |

Another indicator of precision in the assessment of inhaled plutonium is suggested by the results of interlaboratory measurements on an individual in 1974². Here, the subject of an accidental exposure was counted at 7 different facilities within 2 weeks and results varied from 10 to 23 nCi of ²³⁸Pu (one laboratory's data of 54.8 nCi expected). An average of the 6 results is 17.7 nCi or about 2 times the average

laboratory's detectable limit. This is only an indication of precision and not necessarily accuracy.

Precision is expected to improve significantly by 1979 as a result of a single intercalibration phantom that will be used by most U.S. lung counter facilities during the next two years. This should improve precision to the point where true accuracy will be the challenge. True accuracy will probably develop slowly because of unknown factors of distribution for lung depositions of heavy elements.

REFERENCES · I-G

1. Proceedings of the Workshop on Measurement of Heavy Elements In Vivo, held at Battelle Seattle Research Center, Seattle, Washington (June 24-25, 1976).
2. Unpublished Data, Mound Laboratory, Miamisburg, Ohio, May, 1974.

NEW COUNTERS II-A

A significant amount of recent research has centered about the use of semiconductor detectors for lung deposition assessments. In order to approach a sensitivity level comparable to existing phoswich detectors, it has been necessary to use arrays of semiconductor detectors.

The only arrayed detectors actually being investigated for lung counting are intrinsic germanium and 2 arrays of 4 detectors, each having 10 cm^2 of active area, costing about \$100,000.¹ Because of the high cost, other laboratories have not been able to secure sufficient funding for research with similar counting systems. Consequently, the research has been limited to only a few persons at one installation and, therefore, is narrowly based.

Others^{2,3} have investigated germanium detectors on a preliminary basis by projecting measurements from a single detector to that of an array so they could be compared to a phoswich detector. Results look somewhat promising as they have with the arrayed detectors, but a conclusive report has yet to show actual improvements in detectable limits compared to a phoswich counting system.

Little consideration has been given to the use of arrayed silicon detectors. However, there is a possibility that they could be used with fewer problems than intrinsic germanium. Silicon surface barrier or silicon lithium drift detectors should work almost as well as germanium and could possibly be operated without liquid nitrogen on a cascaded Peltier device. Silicon detectors (1-2 cm depleted depth) should be comparable to germanium for lung counting photons approaching 60 keV in energy.

Other types of semiconductor detectors that have yet to be fully developed are cadmium telluride, gallium arsenide and mercury iodide

All these have potential for lung counting, but currently lack large active volumes.

Recent advances have been made in the use of proportional scintillation counters⁵. Preliminary results indicate volumes are not critical, resolutions are superior to normal proportional counters, and backgrounds are somewhat lower than phoswich detectors⁶.

Both the solid state and proportional scintillation detectors deserve a concentrated effort in their development if they are to be used to make routine lung assessments. An improvement in the detectable limit is conceivable with such detectors while improving resolution. Improved resolution would significantly help in distinguishing internal from external contamination.

REFERENCES II-A

1. Rocky Flats Plant, Rockwell International, P. O. Box 888, Golden, Colorado 80402.
2. Bemis, C. E., et al, Detection of Internally Deposited Actinides Part IV; Preliminary Considerations in the Use of Large, Planar Intrinsic Ge Detectors, Proceedings of the Ninth Midyear Topical Symposium of the Health Physics Society, February 1-2, 1976, pp. 578-587.
3. Umbarger, C. J. Los Alamos Scientific Laboratory of the University of California, Los Alamos, California 87545, Private Communication, 1975.
4. IEE Transactions on Nuclear Science, Vol. NS-22, February 1975.
5. Palmer, H. E., Recent Advances in Gas Scintillation Proportional Counters, IEE Transactions on Nuclear Science, Vol. NS-22, February 1975, pp. 100-103.
6. Ibid.