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July 1968 through June 1969

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August 1969

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REPORT OF THE BIOASSAY PROGRAM*

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Anne de G. Low-Beer, Thornton W. Sargent,
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ABSTRACT

This report of the Bioassay Program updates the information contained in UCRL-18588 with respect to results of radiochemical analysis of biological samples and whole-body counts on selected employees of LRL, and to continuing studies of in vivo counting of alpha activity by means of a solid-state semiconductor. It presents new developments, including continuing research with the Si(Li) wound counter, and acquisition and operation of a system for alpha pulse-height analysis, and it describes a survey of alpha contamination in Bay Area residents who have no occupational exposure to radiation.

I. Introduction

The ongoing concerns of the Bioassay Program are (1) monitoring on a routine basis of the group of LRL employees selected by the Safety Services Division, and (2) investigation of possible exposure of employees who have been involved in known incidents such as spills or releases from accelerators. In addition to the foregoing, developmental activities have been carried on to extend and improve the Bioassay Program.

II. Routine Radiochemical and Whole-Body Counting Program

Results of the program are summarized in Table I. One hundred thirty-nine persons participated in the program of routine radiochemical analysis, 14 of whom also had whole-body counts. Compliance was 84.2%.

The overall incidence of positive alpha determinations of urine, including both gross alpha and specific actinide determinations, is 11.4%, as compared with 8% for the preceding year.[1] With one exception the highest level of activity in any 24-hour urine specimen was 0.99 pCi. In the case in which an initial value of 45 pCi was found, subsequent determinations showed values that were either below 1 pCi or were negative. It was concluded that the original high value was due to contamination of the bottles. It will be noted that a number of fecal samples were processed for alpha activity during the current year. Fecal samples were requested from all individuals whose urine indicated alpha contamination. In all but one instance fecal analysis confirmed the result of analysis of urine, and the values in feces were slightly higher.

The incidence of beta contamination found in the routine program is 74% for the

*Work done under the auspices of the U. S. Atomic Energy Commission.

Table I. Routine bioassay program.

Type of determination	Number of determinations	Determinations found positive (Number)	(%)	Range of values found
I. Radiochemical				
A. Gross alpha				
1. urine	137	14	10.3	0.1 – 45.00 pCi
2. feces	5	4	80.0	0.2 – 2.00 pCi
B. Specific actinides				
1. urine	11	3[a]	27.2	0.15 – 0.99 pCi
2. feces	3	3[b]	100.0	0.15 – 1.00 pCi
C. Radium	3	0		
D. Gross beta	127	95	74.0	1.2 – 111.00 pCi
E. Phosphorus-32	10	2	20.0	2.5 – 135.00 pCi
F. Carbon-14	16	2	12.0	7.0 – 15.00 nCi
G. Tritium[d]	5	0		
H. Gamma spectroscopy				
1. urine	134	2[c]	1.49	
2. feces	2	0		
3. blood	1	0		
I. Beryllium[d]	1	0		
II. Whole-body count	14	2	14.3	^{125}I trace (1)[e] ^{59}Fe 20 nCi (1)

a. Isotopes identified: ^{237}Np (4); ^{244,245}Cm (1, 1); ^{240,244}Pu (2, 1) ^{230,232}Th (1, 2); U (2).

b. Isotopes identified: ^{237}Np (1); ^{244}Cm (1); U (2).

c. Isotopes identified: Two peaks, at 66 and 75 keV (1); ^{57}Co ≈ 1n Ci (1).

d. Processed at LRL-Livermore.

e. The number of individuals in whom each isotope was found is shown in parentheses.

current year, compared with 84.5% in the preceding year. The levels of beta activity are somewhat higher than in the preceding year.[1] Ninety percent of the positive cases are within the range 2 to 25 pCi per 24-hour sample. Five cases (5%) are between 25 and 50 pCi and four are more than 50 pCi. The range for all positive cases in the preceding year was 1 to 20 pCi.

The routine whole-body counting program included 14 individuals. Of these, two were found to have low-level comtamination from ^{59}Fe and ^{125}I (Table I).

III. Incidents

Seventeen persons were involved in incidents involving potential exposure to a variety of radionuclides. Table II presents results of investigation of these exposures. Seven possible exposure cases involved actinide elements only, two involved both actinides and gamma emitters; one person (case 10, Table II) was exposed to ^{160}Tb, and ^{65}Zn was found to be present as a contaminant; seven persons were exposed to gamma

Table II. Results of investigation of exposure of personnel to radionuclides.

Case No.[a]	Urine			Feces			Isotopes identified	Whole-body count		
	No. determinations Total	No. determinations Positive	Range of values per 24 hr	No. determinations Total	No. determinations Positive	Range of values		Isotope or energy (MeV)	Activity	Comment
I. α-contamination										
1	1	0		1	1	0.5 pCi				negative
2	1	0								
3	5	0		2	2	0.5 pCi	^{244}Cm			negative
				1(blood)	0					
4	8	7	0.39-5.5 pCi	5	5	0.9-16 pCi	^{244}Cm	^{244}Cm	trace	negative in 24 hours
				1(blood)	0					
				1(sputum)	1	100 pCi	^{244}Cm			
5	1	0		2	1	1.22 pCi				
6	1	1								
7				1	1	0.27 pCi				negative
8	1	0								
9	1	0								
II. β-contamination										
10	5	3	30-120 pCi	4	4	30 pCi-160 nCi	^{160}Tb	^{160}Tb	231 nCi 14 nCi	First count 77 days later
III. γ-contamination										
11	1	0		1	1	1.9×10^4 γpm	$^{57,58}Co$[b]	^{57}Co ^{58}Co, ^{8}Be ^{58}Co, ^{54}Mn 0.060	17 nCi 13 nCi 27 nCi 27 nCi	one count only
12	1	0		1	0			^{57}Co ^{58}Co, ^{54}Mn 0.060	5 nCi 8 nCi 9 nCi	
13	1	0						0.060 ^{57}Co	2 nCi 1 nCi	
14	1	0								
15	1	0		1	1	3.8×10^4 γpm	$^{57,58}Co$	0.060 ^{57}Co ^{58}Co ^{58}Co, ^{54}Mn	270 nCi 170 nCi 190 nCi 330 nCi	10 nCi 14 nCi 2 nCi 3 nCi 12 days later
16	1	0						0.060 ^{57}Co	trace trace	
17	1	0		1	1	1.8×10^3 γpm	$^{57,58}Co$	0.060 ^{57}Co ^{58}Co, ^{54}Mn	50 nCi 33 nCi 18 nCi	
18	1	0								
19	1	0		1	1	3.2×10^3 γpm	^{160}Tb	^{65}Zn	27 nCi	6 nCi 77 days later
20	2	0		1	0			0.060 ^{57}Co ^{58}Co, ^{8}Be ^{58}Co, ^{54}Mn	20 nCi 13 nCi 12 nCi 17 nCi	one count only

[a] Three individuals were involved in two types of contamination, i.e., in cases 1 and 11, 7 and 15, and 10 and 19.

[b] Many small unidentified peaks.

emitters only. Fecal examinations were made on six of the persons exposed to alpha irradiation and on five of the persons exposed to gamma emitters. Blood and sputum were analyzed for one person who had had exposure to actinide elements. The sputum had the highest level of activity (100 pCi) of any sample examined. Even this value, however, was well below 0.01 MPBB. It is noteworthy that all fecal analyses were positive, whereas only two persons with actinide exposures and one with beta-gamma exposure had positive urines.

In nonroutine whole-body counting several incidents occurred which warrant comment (see Table II).

At the 88-inch cyclotron there have been two releases within 6 months. In one of these many of the personnel in the building were contaminated, and in the second one individual received about 200 nCi of ^{57}Co. The isotopes involved were primarily ^{57}Co, ^{58}Co, ^{54}Mn, and ^{8}Be; the doses are not high enough to cause concern, as the MPBB for those isotopes is high and the burden is rapidly lost, apparently being largely on the surface of the body or not absorbed from the gut. However, an isotope is present which has not been identified. It has a 60-keV γ ray and persists in the body longer than all the others. Of the seven subjects in whom this gamma peak was observed, two also had detectable alpha activity in the feces, but at a level far lower than that which would correspond to the whole-body 60-keV gamma activity. It is doubtful, therefore, that this activity can be attributed to ^{241}Am. Primarily because its identity is unknown and because it is apparently retained and biologically utilized, and also because of its frequent release at the 88-inch cyclotron, identification of this isotope is a matter of continuing concern in the bioassay program, and also in the operation of the cyclotron.

A spill of ^{244}Cm involving two persons (cases 3 and 4, Table II) demonstrated the relatively poor ability of the whole-body counter to detect this isotope. Because of the very low abundance of γ rays (0.02%), the minimum detectable, as determined with a standard ^{244}Cm source in a phantom, is about 1 μCi for the whole body and 0.3 μCi in the chest. On the day of the spill, a ^{244}Cm peak could be observed visually in the spectrum of one individual, but was not statistically significant. Alpha counts were measurable on this person, and the activity was thus thought to be on the surface. The following day the peak was no longer present. Because of the low sensitivity, the whole-body counter will apparently be useful only in determining whether a subject has more than 3 to 10 times the MPBB of 0.1 μCi of ^{244}Cm.

An investigator (case 10, Table II) working with a source believed to be ^{169}Dy, but which he found to be actually ^{160}Tb, was contaminated from the source. His apparent whole-body burden of 230 nCi of ^{160}Tb fell to 26 nCi within 4 days, indicating that the activity was external or was not retained after ingestion or inhalation. However, after 77 days, 14 nCi of ^{160}Tb still remained, as well as about 6 nCi of ^{65}Zn, which apparently was a contaminant of the original source. This subject is being followed to determine the long-term excretion rate.

IV. Research and Development

1. Si(Li) Semiconductor

As was indicated in an earlier report,[1] a prime concern of the Laboratory is accurate detection of internally deposited actinides. We have undertaken a comprehensive study of the performance and operating characteristics of a Si(Li) semiconductor wound counter.[2] Proceeding along lines described by Bistline and Tyree,[3] we have developed a program for estimating the average depth and activity of an

actinide wound in soft tissue. This has been accomplished by comparing the ratios of attenuated low-energy photons transmitted through various thicknesses of fiberboard, Lucite, water, and a tissue-equivalent liquid.[4] Figures 1 and 2 show typical plots obtained from these attenuation studies. The counter efficiency and response to source geometry were determined with ^{241}Am and ^{243}Am electrodeposited standards (Figs. 3 and 4). The minimum detectable activity (MDA) was calculated to be 14 photons/min for a 10-min count. (This is based on the equation

$$MDA = \frac{K}{E} \left(\frac{b}{t}\right)^{1/2} ,$$

where K = 3.3 for a 95% confidence level, E is the efficiency, b is the background count rate, and t is the counting time.) Samples are detectable at this level, but not identifiable; consequently, longer counting times are frequently necessary.

Reference collections of actinide spectra analyzed with semiconductors are difficult to obtain--if available at all. We have established our own actinide library, which will be continuously enlarged as nuclides are made available to us. To date our studies have included ^{238}Pu, ^{239}Pu, ^{241}Am, ^{243}Am, ^{244}Cm, ^{252}Cf, and ^{253}Es.

The capability for monitoring these nuclides in wounds is ordinarily limited by the type of equipment available. Our present equipment, in addition to the 300-mm^2 Si(Li) semiconductor, includes a 3-mm-thick by 2-in.-diam thin-window NaI probe, a 1/4-in.-thick by 2-in.-diam probe, a 4-by 7-in. NaI whole-body counter crystal, and an Anger scintillation camera.

To evaluate these systems and the data available for wound analysis, a simulated hand wound study has been made. Two Lucite hand phantoms were fabricated, and a pure point source implanted in each one. The phantoms were then distributed to technicians who were unaware of the source's location, activity, identity, and depth. The technicians then proceeded to follow an outlined program for wound analysis. The phantoms were checked with typical survey meters, and only normal background activity was observed. The phantoms were then analyzed by using the 1/4-in. NaI probe to locate the wound, and the Si(Li) semiconductor for identity, depth, and activity determinations. The sources were located and identified as 44 nCi of ^{239}Pu at a depth of 6 mm, and 11 nCi of ^{241}Am at a depth of 14 mm. The true values were 44 nCi at 5 mm for ^{239}Pu and 13 nCi at 15 mm for ^{241}Am. Both phantoms were then checked with the Anger camera, and the ^{241}Am source was readily observed in a 1-minute count (Fig. 5). The ^{239}Pu source was not detectable after 30 minutes of count time. Although the conditions of this test case might not be considered optimum, we gained valuable experience from the entire procedure. In an actual case of contamination, analysis of a wound could be very complicated. Errors can be introduced by (a) self-absorption in the source, (b) errors in tissue absorption measurements, (c) fragmentation of the source, and (d) unknown isotopic composition. In view of these limitations, we feel the counter may best be used to identify sources and determine an approximate average depth. The determination of a source depth to within 1 mm is still quite questionable.

Data processing is partially handled by our computer, and we are currently working on a program that will be able to do the entire analysis. Future work will be directed towards refinement of our current data and techniques.

2. Alpha Pulse-Height Analysis

Experience over past years has indicated a need for a high-resolution alpha system to determine isotopic composition of actinides

in order to avoid dependence on other departments of LRL for such determinations. We have now acquired a premium-grade silicon alpha detector, which is currently in use. This detector system enables us to obtain 14 keV FWHM for ^{241}Am sources. Figure 6 indicates the relative ease with which sources can be identified, and also the effect of the method of source preparation on alpha resolution.

As will be seen in Fig. 6, electrodeposition performed with our present equipment does not provide samples that give highest possible resolution. Moreover, electrodeposited activity has been found to rub off with handling of the sample, causing possible contamination of equipment and loss of activity. For these reasons we have been experimenting with preparation of monomolecular layers according to the method of Deal and Chanda.[5,6] The method involves extracting the alpha emitter from an aqueous solution with 0.2 M TTA (thenoyltrifluoroacetone) dissolved in benzene. The organic phase, freed of any aqueous contamination by centrifugation, is plated in small aliquots (10 to 25 λ) on a platinum planchet. When dry and flamed, the planchet bears only the elements extracted from the aqueous phase. Alpha pulse-height analysis has revealed that the preparation is suitably thin and uniform for high-resolution alpha spectroscopy.

To date we have applied this method only to the preparation of chemically pure sources. We will determine next the effectiveness of the method for isolation of alpha-emitting nuclides detected in biological material.

3. Survey of Alpha and Beta Contamination in Bay Area Residents

Continuing low levels of beta activity found in a high percentage of LRL personnel included in the routine bioassay program, and the occasional incidence of very-low-level alpha activity in these individuals,[1,7] have prompted a survey of a sample of residents of the Bay Area who have no occupational exposure to radiation. Urine samples have been collected from 59 volunteers, and have been analyzed for alpha and beta activity. We propose to continue this sampling to include at least 100 persons. A report of this survey will be issued when the sample is complete.

V. Conclusion

The record presented in this report is evidence of the high degree of safety from radiological contamination at LRL-Berkeley. The incidence of exposure in the sampling of workers selected for bioassay is 12% for alpha activity, 74% for beta activity, and 14% for gamma activity. In no case has the activity exceeded 0.01 MPBB, even following incidents in which a high potential for contamination of individuals exists. Our 1968 report[1] established that low-level beta contamination is prevalent among the general population and therefore should probably not be regarded as due to industrial exposure when found in the routine bioassay program. The current study on alpha contamination in the general population will assist us to evaluate the significance of those positive alpha determinations that are found in routine analyses on LRL workers.

Continuing development of the x-ray counting system provides a new and increasingly efficient means of assessing the contamination of wounds with radioactive nuclides.

New methods for preparation of alpha sources, and the introduction of an alpha spectrometry system, insure the accurate identification of all nuclides in the actinide series.

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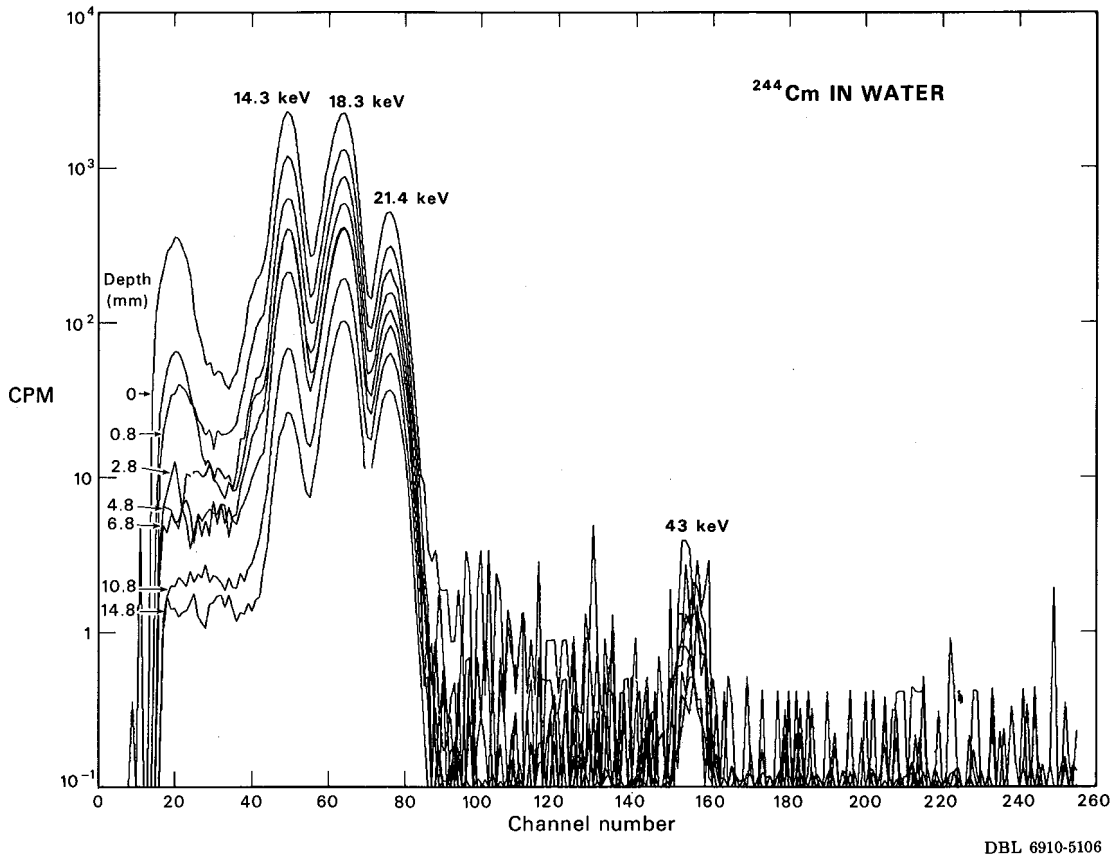

Fig. 1. Spectrum of ^{294}Cm, showing attenuating effect of various depths of water.

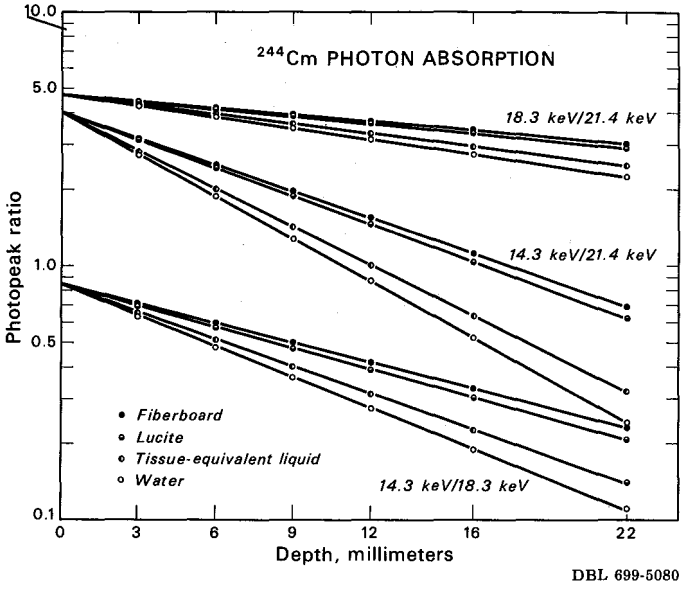

Fig. 2. ^{244}Cm L x-ray photopeak ratios as a function of depth in various absorbers.

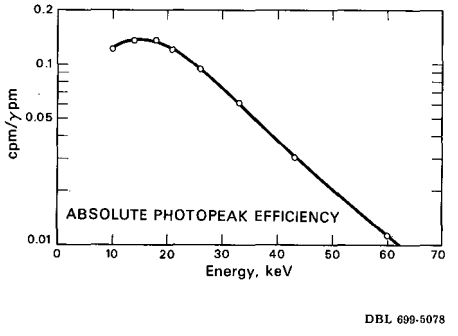

Fig. 3. Full peak efficiency based on the ratio of counts per minute to photons per minute for a 4 mm diameter electrodeposited ^{241}Am source.

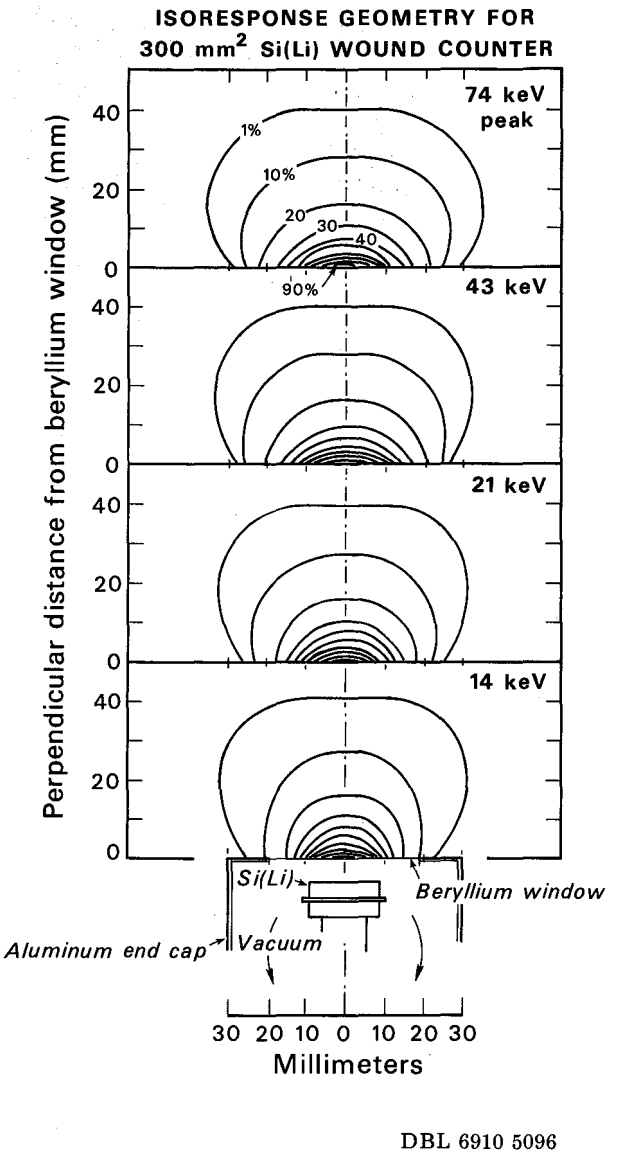

Fig. 4. Isoresponse curves for various energies, showing the attenuation due to source position relative to the detector. The source was 500 nCi of ^{243}Am electrodeposited on stainless steel.

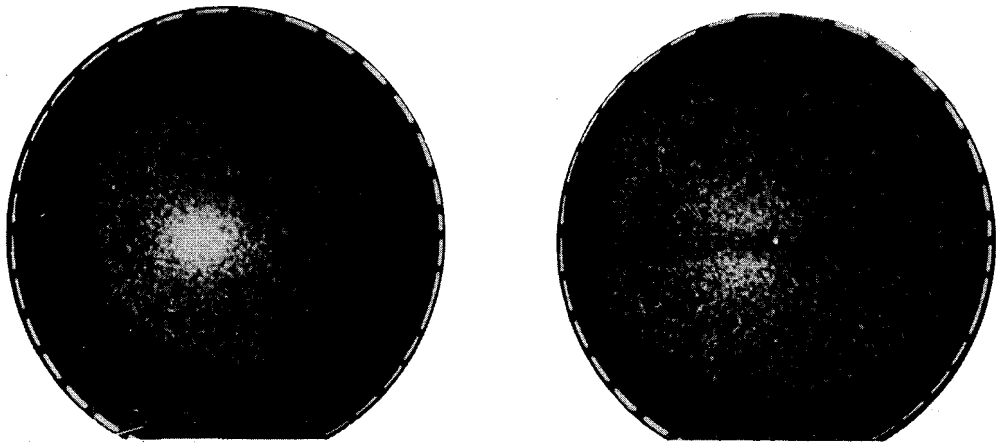

Fig. 5. One-minute pictures by the Anger scintillation camera of a simulated wound containing ^{241}Am in a Lucite phantom. The location of the activity in the phantom was determined by incremental movements of a lead rod. The pictures were taken with different field sizes. Left: no mask; right: with with Pb mask.

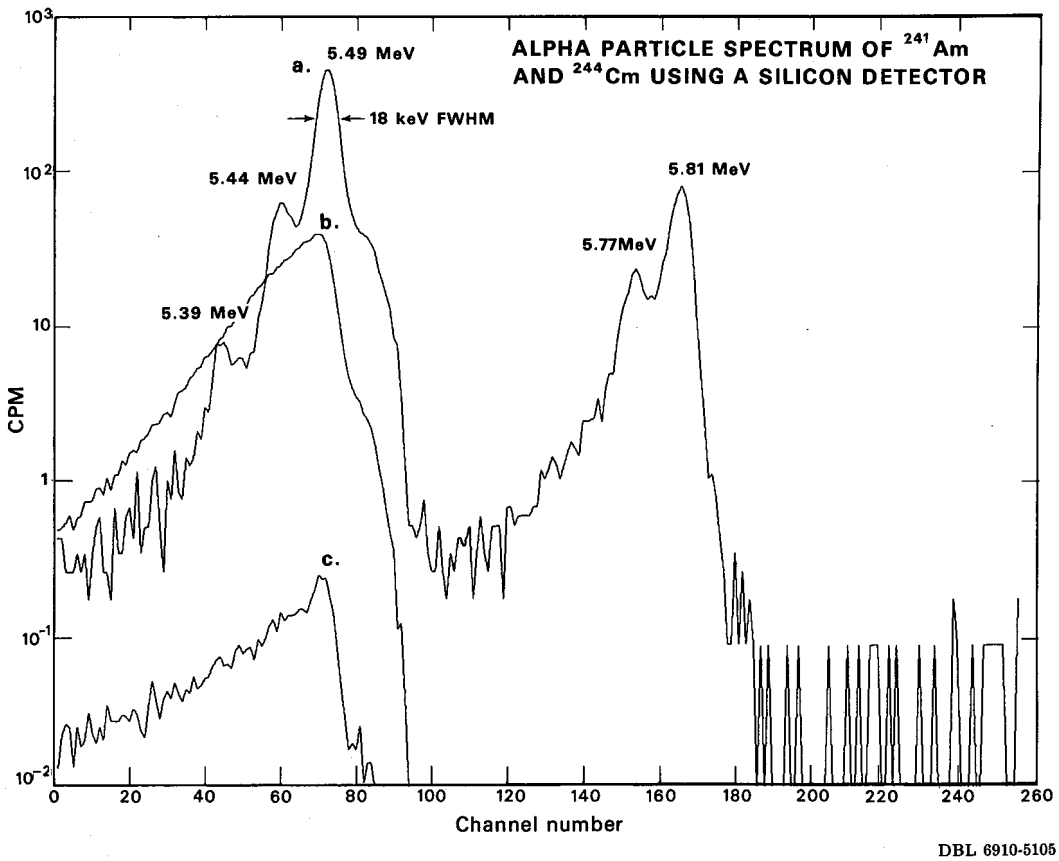

Fig. 6. Comparison of ^{241}Am resolution as a function of source preparation. Curve a is a standard provided by the Safety Services Department; b is a sample electrodeposited on stainless steel; c is a monomolecular layer plated on platinum after TTA solvent extraction.