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Lawrence Radiation Laboratory
University of California
Berkeley, California

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ABSTRACT

Ninety three accelerator workers were assayed for radioactivity at the Lawrence Radiation Laboratory–Berkeley. The techniques used were whole-body counting and radiochemical analysis of urine. Of the 93, three had measurable but very small quantities of alpha activity in the urine, six had detectable gamma activity by whole-body counting, and almost all had beta activity in the urine. We believe the beta activity is from fallout in food, water, and air.

I. INTRODUCTION

This report is concerned with a bioassay program including radiochemical analysis of biological material, usually urine, and whole-body counting on 93 accelerator workers.

Several cases of internal deposition of ^{65}Zn in accelerator workers have been reported previously, from Massachusetts Institute of Technology,[1] Los Alamos Scientific Laboratory,[2] and Lawrence Radiation Laboratory–Livermore.[3] In addition Los Alamos Scientific Laboratory reported a single case of suspected ^{7}Be contamination, which may have been external, and LRL-Livermore reported the occurrence of internal ^{54}Mn, ^{22}Na, and ^{48}V. In 1962, 10 workers on the Berkeley 60-inch cyclotron were shown to have internal contamination from ^{65}Zn.[4]

The investigation reported here was made in spring 1968 on accelerator workers at the Lawrence Radiation Laboratory–Berkeley. This study is supplemental to the routine Laboratory bioassay and personnel monitoring program.

Characteristics of the four accelerators presently in operation at Berkeley are given in Table 1. The accelerators are of conventional design and use stainless steel, steel, copper, duraluminum, and various hydrocarbons in their construction and function. Table 2 lists their materials and gives the isotopes one might expect to find in workers who have handled radioactive accelerator parts or have drilled, sawed, or soldered them.

In considering the question of whether these or other isotopes are actually ingested by workers and whether they can subsequently be detected the following aspects of the problem must be kept in mind.

First, the specific activity is very low. Except for actual accelerator targets and beam dumps, accelerated beam losses are small, as are the activities induced by charged particles. Neutrons, which are formed when the beam strikes a target or other loss point, diffuse widely and may induce a nuclear reaction anywhere within a large mass of accelerator material or shielding.

Second, the induced activity is not generally concentrated or confined. Again, except for targets which are accorded special handling, the bulk of the activity induced in accelerator structures is almost never so intense as to require the techniques for handling which are used for ordinary radioisotopes. No shielded enclosures, gloved boxes, or remote-handling equipment are used or are really necessary. Even in high radiation areas (> 100 mrem in an hour) control of working time is almost the only method used to control radiation exposure.

These two aspects of the problem--widespread low specific activity and ordinary handling of this material--convinced us that anyone associated with the operation of an accelerator <u>might</u> ingest <u>some</u> activity but that the quantity would probably be small.

Accordingly, we selected for each accelerator a group of people who were primarily responsible for the operation and maintenance of the machine. Included were accelerator operators, electrical and mechanical technicians, physicists and chemists, and engineers. In

meetings it was explained that the program we proposed should be considered as supplemental to the ordinary personnel monitoring program. The proposed program included both a whole-body count (for gamma activity) and radiochemical analysis of urine (for beta, and alpha activity). Two overnight urine specimens were required. In spite of the extra time and inconvenience, cooperation was good. Ninety-nine persons had been selected for the study; 93 individuals reported for a whole-body count, and of these 84 submitted a 24-hour urine sample.

II. TECHNIQUES

Bioassay

Urine specimens are treated by a survey method which involves preparation of the sample by alkaline phosphate precipitation, in which the monovalent cations are eliminated. The ash from the alkaline phosphate precipitation is suspended in 10 ml of 2N HNO_3. The whole sample is counted by gamma spectroscopy using a 4-inch NaI crystal and a 400-channel pulse-height analyzer. One fifth of the sample is then plated on aluminum for counting of gross beta activity in a Nuclear-Chicago proportional gas-flow counter. The remaining four fifths of the sample is processed by bismuth phosphate and lanthanum fluoride co-precipitations and counted for gross alpha activity in the Nuclear-Chicago proportional gas-flow counter.

Whole-Body Counting

The Donner whole-body counter is of the Argonne type, with a 6-inch steel shield. The subject sits in an inclined chair and is counted by a stationary 9-3/8 by 4-in. NaI(Tl) crystal, positioned with its center 40 cm from the back and seat of the chair. The gamma pulses are analyzed by a Packard 400-channel pulse-height analyzer, calibrated at 5 keV/PHU, yielding a γ-ray spectrum from 0 to 2.0 MeV. Data are recorded on printed paper tape, which is then keypunched for computer analysis. Calibration of the area under the photopeaks to yield microcuries body burden is obtained from the spectra of medical patients injected with known quantities of short-lived radionuclides with various gamma-ray energies. Counting times for this series of subjects was 15 minutes. The limit of sensitivity for detecting the isotopes in Table 1 is between 1 and 10 nanocuries.

III. RESULTS

Results of radiochemical analysis of urine and of whole-body counts are shown in Table 3. Three of the positive alpha determinations shown in the table must be regarded as questionable until they are confirmed by analysis of a second specimen. Such confirmation is now pending. What appeared to be clearly a ^{237}Np peak, energy 4.77 MeV, was present in each of the positive samples examined by alpha pulse-height analysis. In addition, a number of smaller peaks were seen in all these samples. These had energies of 4.10, 4.35, 4.56, 4.67, 5.18 (^{238}U), 5.30 (^{241}Am), and 5.80 (^{241}Cm) MeV. The

activities are not significant from a health protection standpoint, but their source is the subject of a general alpha contamination study.

The high incidence of beta contamination is consistent with observations of the Bioassay Laboratory that during the winter and spring of 1967-68 residents of the San Francisco Bay Area, irrespective of locality or occupation, had levels of beta activity ranging from 5 to 20 pCi per 24-hour urine sample.[5] Studies of the rate of radioactive decay on some of these samples indicate a half-life in the neighborhood of 50 to 60 days. It seems unlikely, therefore, that a significant amount of this activity is due to ^{90}Sr.

The fact that no gamma activity was found in any of the urine samples illustrates the high detection limit of the method in comparison with whole-body counting; gamma activity was found in some individuals by the whole-body counting technique although not detected by gamma counting of urine.

Individuals were whole-body counted preliminarily while fully dressed. In all cases of apparent contamination, a second count was made after the individual had showered and donned a hospital gown. Values shown in Table 3 represent internal contamination confirmed in this manner. In general, however, it is difficult to distinguish between surface and internal contamination with a single whole-body count. Computer analysis of gamma spectra generated in the whole-body counting program is now under way. It is not expected that this will alter significantly the findings presented here.

Among the 88-inch cyclotron workers, in addition to the three subjects listed in Table 3, eight subjects had measurable activity of ^{58}Co, ^{57}Co, and ^{185}W on their clothing. This was at a time shortly after the cyclotron had been shut down for extensive work and clean-up of the acceleration chamber, and these activities were thought to be present during this operation. The activity attributed to ^{185}W was tentatively identified by a 0.06-MeV γ ray and an 80- to 90-day half life. The isotope tables do not show agreement as to the presence of this gamma energy for ^{185}W, however. This isotope, still not positively identified, is noted here because of the possibility of confusing it with ^{241}Am, which has a γ ray of the same energy; the activity seen here, however, had no associated alpha activity.

A survey was conducted in 1962 of ten members of the crew of the 60-inch cyclotron before it was dismantled. Seven of these men had burdens of ^{65}Zn, ranging up to 9 nCi, comparable to results of others.[1-3] One route of ^{65}Zn into the body was found to be due to soldering copper parts which had been activated; apparently the zinc vaporizes and is inhaled by the man doing the work. A ^{65}Zn burden was found in a man who had been soldering a copper part while wearing a respirator. Activity was found on the respirator filter as well.

The much lower incidence of internal burdens of gamma-emitting nuclides in the crews of more modern accelerators attests to the efficacy of present safety standards.

IV. CONCLUSIONS

Accelerator workers who have been studied in this survey show the following incidence of radioactive contamination: alpha (confirmed) 3.5%, gamma (by whole-body count) 6.4%, beta 94%. It is believed that the high incidence of beta activity represents environmental contamination. The levels of activity found do not exceed 1 pCi per 24-hour urine for alpha emitters, and 10 nCi per whole-body count for gamma activity. Accordingly, no changes in existing procedures are contemplated as a result of this survey, since it seems evident that normal habits of cleanliness and occasionally the use of protective clothing and gloves are adequate to insure that the ordinary accelerator worker at LRL-Berkeley will not receive radiation exposures of any consequence from internal sources. Nevertheless, it is believed that periodic surveys of the type reported herein have value in helping to maintain safe working conditions and in bringing to light any unsuspected instances of possibly significant contamination of workers.

FOOTNOTES AND REFERENCES

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

1. Annual progress report, Massachusetts Institute of Technology, contract AT(30-1)-952 (May 1959).

2. M. A. Van Dilla and M. J. Engelke, Zinc-65 in cyclotron workers, Science 131, 830-832 (1960).

3. A. L. Anderson and C. T. Schmidt, Zn^{65} content in 90-inch cyclotron workers at Livermore, Lawrence Radiation Laboratory Hazards Control Progress Report No. 25 (April-August, 1966), UCRL-50007-66-1, 1966.

4. T. W. Sargent, Donner Laboratory internal report.

5. Anne de G. Low-Beer and Thornton W. Sargent, Report of the Bioassay Program, July 1, 1967-June 30, 1968, UCRL-18588, Nov. 1968.

Table 1. Characteristics of accelerators at Lawrence Radiation Laboratory–Berkeley.

184-Inch synchrocyclotron (modified in 1957)			
Particles accelerated	protons	deuterons	alphas
Maximum energy (MeV)	730	460	910
Average beam current (μA)	0.75	0.75	0.25
Maximum energy before modification (MeV)	350	190	380
Bevatron (proton synchrotron)			
Particles accelerated	protons		
Maximum energy (MeV)	6200		
Maximum intensity	5×10^{12} protons/pulse, internal 2×10^{12} protons/pulse, external		
Heavy-ion linear accelerator			
Particles accelerated	deuterons, ^{12}C, ^{14}N, ^{16}O, ^{20}Ne, ^{40}A, and others		
Maximum energy (MeV)	10/nucleon		
Average beam current (μA)	0.01 to 4.0		
88-Inch cyclotron			
Particles accelerated	protons	deuterons	alphas
Maximum energy (MeV)	60	60	130
Average beam current (μA) (presently limited by deflector cooling).	30	40	20

Table 2. Isotopes to be expected from irradiation of some selected materials.

Material	Isotope
Cu	^{63}Ni, ^{65}Zn
Stainless steel	^{57}Ni, ^{57}Co, ^{58}Co, ^{60}Co
Steel	^{55}Fe, ^{59}Fe, ^{54}Mn
Duraluminum	^{24}Na, ^{22}Na
Plastics and oils	^{7}Be

Table 3. Results of survey of accelerator workers. (Numbers in parentheses indicate number of workers in whom isotopes were found.)

Location and type of determination	Number: Individuals	Number: Determinations	Number: Individuals positive	Isotope or energy	Values (or range) (μCi)
184-Inch cyclotron					
A. Radiochemical					
1. alpha	11	16[a]	3[b]	^{237}Np[c] (2)	0.15 to 1.0 × 10^{-6}[d]
2. beta	11	14	12	---	4.2 to 42 × 10^{-6}[d]
3. gamma	11	11	0	---	---
B. Whole-body count	14	14	2	0.825 MeV(1)	trace
				^{65}Zn (2)	3 × 10^{-3} (1); 7 × 10^{-3} (1)
Bevatron					
A. Radiochemical					
1. alpha	42	42	1[b]	---	0.3 × 10^{-6}[d]
2. beta	42	42	38	---	1.7 to 120 × 10^{-6}[d]
3. gamma	42	42	0	---	---
B. Whole-body count	44	44	1	^{65}Zn	1 × 10^{-3}
Heavy-ion linear accelerator					
A. Radiochemical					
1. alpha	10	11	1	^{237}Np (1)	1 × 10^{-6}[d]
2. beta	10	10	9	---	2.7 to 6.7 × 10^{-6}[d]
3. gamma	10	10	0	---	---
B. Whole-body count	13	13	0	---	---
88-Inch cyclotron					
A. Radiochemical					
1. alpha	21	21	1[b]		0.7 × 10^{-6}[d]
2. beta	21	21	20	---	1 to 10 × 10^{-6}
3. gamma	21		0	---	---
B. Whole-body count	22	22	3	^{185}W? (2)	→ 7 × 10^{-3} (2)
				$^{58,57}Co$ (1)	→ 3 × 10^{-3} (1)

a. Multiple determinations on cases thought to be positive.

b. One pending confirmation.

c. What appeared to be clearly a ^{237}Np peak, energy 4.77 MeV, was present in each of the samples examined by alpha pulse-height analysis. In addition, a number of smaller peaks were seen in all the samples. These had energies of 4.10, 4.35, 4.56, 4.67, 5.18, 5.30, and 5.80 MeV. It is thought that peaks with lower energies may be isotopes of uranium, but because of the very low counting rate of all the samples, positive identification is not attempted.

d. Per 24-hr urine sample.

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LAWRENCE RADIATION LABORATORY
UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720