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LOS ALAMOS SCIENTIFIC LABORATORY
OF THE UNIVERSITY OF CALIFORNIA ○ LOS ALAMOS NEW MEXICO

EXAMINATION OF GROSS PARTICLES FROM KIWI-A3 NUCLEAR ROCKET PROPULSION REACTOR AT NEVADA TEST SITE

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(Title Unclassified)

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ABSTRACT

This report presents the data obtained from the analysis of large particles found in the effluent from Kiwi-A3 test reactor. The methods used, and their application to future test operations, are discussed. Procedures are described for the isolation of particles from static sampling systems and for paper chromatography of the fission products. A Polaroid film fallout tray for static sampling of particulates also is described.

ACKNOWLEDGMENTS

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INTRODUCTION

Objectives

This study was to gain an insight into the problems associated with the chemical and physical analysis of discrete particles, to evaluate techniques for application to future nuclear rocket engine effluents, and to accumulate sufficient data to identify the particles by simple methods. Recommendations for methods and techniques to be used in the future are based on the experiences gained in this study.

Discussion

The exact procedures used in isolating or examining particles from previous nuclear events are not fully documented. Particles associated with nuclear weapon testing are necessarily grossly contaminated with extraneous material, either from the test site or from the weapon carrier; whereas particles from nuclear test reactors of the Kiwi-A3 design are relatively pure fission products because they emanate by erosion and corrosion from inside the reactor itself. Should the reactor be tested so as to cause excessive turbulence on the ground surface, the particles then would become adsorbed on the dust surrounding the reactor, and would not necessarily be handled as described here.

The relatively large particles from the reactor test should cause little concern from a health standpoint; they represent only a means for studying radioactive decay, the distribution of masses of particles within a given area, and a general prediction of isotopic composition of the smaller fallout particles.

The techniques used in this study are essentially standard methods unless otherwise described. All the radioactive data taken at the Nevada Test Site were determined as described in LAMS-2588;[1] the area under study and the devices used in sampling ground and air contamination are described in the same document.

Location of Particles

The midpoint of the run was on October 18, 1960, at 16:43 hours. The particles were located by a ground survey team using a portable beta-gamma survey instrument. The dose rate background at 500 and 1000 feet from the reactor test cell made it impossible to find small particles or particles of low specific activity.

The particles were clustered in the three general areas indicated on Figure 1. Particles 1 to 17 were found 18 hours after the midpoint of the run; particles 20 to 25 were found one week later and numbered to indicate a second field search. Many of the particles were poorly handled and became abraded. They were isolated from the soil by means of cellophane tape, then, for optical and radiometric study, were mounted on microscope slides, which proved a poor technique. The recommended technique developed during this study, given in Appendix A, was used for the isolation of particles 20 to 25.

MICROSCOPY

Technique

Each particle was photographed with a 35 mm Contax camera mounted on a Unitron adapter and prism, through an Ortholux microscope having a 10X eyepiece, a 6X objective, with both substage and incident light. Because all the particles were opaque, polarizing or dispersion microscopy was useful only for adsorbed particulate dust. Each particle was placed on a microscope slide; as mentioned above, the early technique for isolating the particles was poor, and some particles were crushed by the coverslips used in mounting the particles. Photographs 1, 2, and 3 are representative of the three general classes of particulates found (scale: 1 cm = 1μ). Photograph 1 illustrates the friability of the particles; photograph 2 shows an adsorbed gray-white substance on black, granular material; photograph 3 illustrates the metallic character of the surface of several of the particles. Each particle is described in Table I. The size of the particles was determined with an ocular micrometer and confirmed from photographs with a stage micrometer.

Results

Table I shows the number of particles on each slide (some particles were crushed by the coverslip), the approximate combined size, location of the particles by distance from the reactor, a description of the particle, and the extrapolated beta activity at 1000 minutes.

Conclusion

The particles from within 1000 feet of the reactor are predominantly black, metallic granules; at 6000 feet the particles appear mainly brassy on a black, metallic granule. The area described as "white, yellow, or gray added residue" will be shown later to be potassium chloride and other JATO fuel products. There is no relationship between particle size and distance, but there is some correlation between appearance of the particle and distance. Particles 12, 13, and 15 show definite fractures, illustrating the friability observed in all of the particles (Photo 1).

RADIOMETRIC ANALYSIS

Beta and Gamma Counting

The beta and gamma activity was determined at the Nevada test site with the equipment described in LAMS-2588. Although each particle was followed for its decay, variations in the particle preparations and in the counters gave curves which were of little value in reconstructing the original activity.

Figure 2 illustrates the composite beta curves of all the particles and several trays which were known to have only a single particle. The solid line indicates a composite of all particles and the dotted line represents only three fallout trays and one particle. The gamma activity is not presented because the great variations within each particle preclude any interpretation.

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Gamma Spectra

The gamma spectrum of each particle was determined at various intervals and interpreted for gamma energies and half life; the spectra indicated a similarity of the particles. Those fission products which were identified positively are given in the following summary:

Particle No.	NUCLIDE					
	Ta-182	I-131	Te-I-132	I-133	Ba-La-140	La-141
1	*	*	*	*	*	*
2	*	*	*	*	*	*
3	*	*	*	*	*	*
4	*	*	*	*	*	*
5	*	*	*	*	*	*
6	*		*		*	
7	*					
9	*					
10	*					
11	*	*	*	*	*	
12	*	*	*		*	
13	*		*			
14	*					
Special Soil	*	*	*		*	*
Fallout Tray 6-24	*				*	

The gamma spectra of particles 7, 9, 10, and 14 indicated primarily Ta^{182}, in such excess that interpretation of other gamma energies was impractical.

After the particles had been returned to Los Alamos, many different radiometric procedures were carried out. The gamma spectra were repeated on a Los Alamos Model 200, 100 channel gamma analyzer with an 8 x 4 inch thallium activated sodium iodide crystal, shielded to reduce the background to a minimum. The gamma spectrum was determined

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on the 34th day after the Kiwi-A3 reactor test. The raw data were collected and processed to remove background counts and to correct for resolving time. The areas selected were reduced to relative activity by dividing by the total counts in the energy range 0 to 2 mev. Table II gives the relative activity of the primary gamma energies corresponding to 0.75 mev for $Zr\text{-}Nb^{95}$, 1.6 mev for La^{140}, and 1.1 and 1.2 for the Ta^{182} doublet.

Particles 9 and 10 are relatively pure Ta^{182} while the remaining particles are grossly contaminated with fission products. Figure 3 illustrates the curve of activity vs. channel (energy) and the energy calibration of the crystal for the two types of spectra found in this study.

Beta Absorption Curves

The beta absorption study was to determine the applicability of the technique to the identification of differences within several particles, and not to establish precise energy levels. A direct method of plotting data was used, rather than Feather's[2] more precise method for determining beta energies.

A Tracerlab E-3A beta absorber set was used, and the beta activity of the maximum absorber was subtracted from all counts; each count then was divided by the free count (no correction for air absorption) to give the normalized values. These values were plotted against the absorber values in mg/cm^2 of the aluminum absorber and the normalized count

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plotted on semi-logarithm paper as shown in Figures 4 to 17. Figures 18 and 19, for comparison, illustrate the same type of data for Cs^{137} and Ta^{182}. Table III lists the $E_{\frac{1}{2}}$ and the E_{max} determined from the graphs. The energies are based on the Glendenin beta absorption curve.[3]

The graphs themselves are perhaps more interesting because the shape of the curves is an indication of the isotopic purity of the particles, and the graphs vividly illustrate that absorbers cannot be used to reduce gross counts for beta counting when the samples contain unknown mixtures of nuclides, unless more than one absorber count is made and the reduction does not exceed 30 per cent of the original activity.

Radioautography

The particles were submitted to two types of radioautography by placing the particles directly against (a) Kodak x-ray film and (b) Polaroid film (experimentally, because the Polaroid film might prove a simple method for locating particles in similar field operations).

(a) X-ray Film

The particles were placed face down on Kodak type K x-ray film with only a coverslip separating the particles from the film. The particles remained in contact for one hour, the film was developed and dried in the usual manner, and the size of the darkened area measured. The beta activity was determined immediately before exposure. A comparison of the particle size, exposed spot, and activity

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in total disintegrations is given in Table IV. The film is sensitive also to gamma rays, but gamma activities are not included.

Skillern[4] showed that the log of the total disintegration plotted against the log of the spot diameter is a straight line. Figure 20 shows the relationship observed in the case of the particles examined here. Particles 4 and 14 are not on the line. Particle 14's spot diameter would give better agreement had the exposure time been shortened, and a more accurate beta count been possible; the method of preparation and the coverslip used to mount particle 13 caused a loss of activity. Neither spot diameter nor beta disintegrations correlate with the actual size of the particle.

If the exposures could be made immediately after isolation, the exposure time of the particles may be brief. Standard field counting can be done during the development of the films. See Appendix B for recommendations for future application of x-ray film radiography.

(b) Polaroid Film

Type 37 (ASA 270) and Type 57 (ASA 3000) Polaroid films were investigated for their possible application to particle detection and sizing in the field. Laboratory experience indicated a similar relationship, for activity and spot diameter, to that described above for x-ray film.

Particles placed on the face or paper side of the single sheet film package gave a clearly defined spot on the film and a somewhat shadow-edged clear spot on the paper, while particles placed on the

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film or back side of the package gave a clearly defined white area with sharp edges in the precise shape of the particle. It is interesting that the negative in the latter case gave better results because a reversal of the film formed a halo around the exposed area and clearly marked the particle. The central portion of the cleared area was identical to the size and shape of the particle (Photo 4).

During a special test operation, some 150 films of each of the film types were used, prepared by covering each with a mylar film and spraying with silicone grease for better particle retention, then exposing to the atmosphere for two days, during which the temperature ranged from 65° to 85°F. and 0.2 inch of rain fell. The films were not damaged by the water and those films protected from direct midday sun were satisfactory; however, because of a pinhole coding system being employed, the films were fogged. A new series of experimental trays has been prepared and tested for possible application to future fallout studies; see Appendix C.

CHEMISTRY

Introduction

The choice of chemical analytical techniques for particles emitted from nuclear reactor systems depends largely on the information desired. Standard methods of dissolution and analysis are applicable when a single given element or specific isotope is to be determined. However, the particles or fallout debris eroded during a Kiwi-type reactor test

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operation contain the fissionable material and its fractioned fission products, also neutron activated nuclides from added materials. Consequently, the chemistry becomes ultramicro in order to determine the inert elements of interest, and individual isotopic composition analysis is impractical immediately following a reactor test. When radioactive material (gas, vapor, or particulate) has become adsorbed on to normal atmospheric dust, the adsorbed must be distinguished chemically from the inert material. Techniques have been developed for handling these particles under high magnification, and for identifying eluted material in serial elutions. Most of the analyses were qualitative in nature because the study was to evaluate techniques.

Dissolution

The particles were returned to Los Alamos for chemical and physical examination, and for further study of the techniques. A particle from slide 2 and particles 1, 3, 6, 11, and 12 were transferred to platinum crucibles and ashed with nitric and perchloric acids without carriers. The residues finally were whitened with nitric acid, then evaporated several times with hydrochloric and hydrofluoric acids. One hundred microliters of a solution which was 4N in sulfuric acid and 4N in hydrofluoric acid was added to each crucible and the walls of the crucibles scrubbed down with a platinum rod. Five microliters (0.005 ml) of each solution was analyzed according to the procedure outlined in Appendix D. The results of the analyses are shown under Chromatography (below).

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Alpha Activity

Five microliters of sample were placed on a 3/4 inch diameter tantalum planchet, evaporated under infra-red heat, and finally flamed. First the planchet was alpha counted in a low background proportional counter, then the planchet was packaged in a nylon ring and disc assembly and counted for its beta activity in a low background beta counter. The results of these counts are given in Table V. The alpha activity was from uranium, which was confirmed by the standard ion exchange procedure described in LA-1858.[5] No alpha activity was found on any of the stages from cascade impactor sample 1-10 or sample 2-11.

The alpha activity should show some relationship to the location of the particles within the reactor. Particles 2A, 3, 11, and 12 are approximately the same size and have proportionately the same amount of uranium activity, indicating that they were from the undersurface of the rods, while particle 6, low in alpha activity and relatively high in tantalum activity, must have come from the surface of the rod.

Chromatography

These particles were treated according to the procedure outlined in Appendix D. The chromatograms were not read on the drum counter but were exposed to x-ray film to locate the activity associated with each particle. The fission products had decayed to such an extent that a 24 hour exposure was necessary to demonstrate the presence of nuclides other than tantalum.

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Particles 1, 3, 6, and 11 had decayed so much that identification of R_f values other than that of tantalum was impractical. Particle 2A contained a small amount of activity at R_f 0, and particle 12 had some activity between R_f 4 and 5; all had activity at R_f 1. The activity at R_f 0 was lanthanum; at 4-5, zirconium; and 1 predominantly tantalum with traces of niobium, which was confirmed by gamma spectrum analysis. The chromatographic techniques are satisfactory for particles and for pure samples. Particles found after a nuclear detonation may be eluted serially with various solvents, using microchemical techniques, and then placed on chromatographic paper strips for absolute identification.

SPECIAL ANALYSES

Spectrographic Analysis

A review of all the particles by Group CMB-1 indicated that particle 14 could be analyzed by spectrographic techniques and x-ray diffraction. The results of these analyses are given in Table VI.

The predominance of tantalum and niobium was expected because these elements were added to the fuel system to change the surface characteristics of its graphite. Titanium and silica are normal in the graphite, and the silica content may have been increased due to the absorption of dust from the ground. The presence of platinum is unexplained.

X-ray Diffraction

Particles 9 and 10 were submitted to CMB-1 for x-ray diffraction analysis. The results are consistent with the method of preparing the reactor system and confirm the spectrographic data.

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Particle 9 was primarily tantalum carbide, with traces of tantalum oxide and tantalum metal, and other patterns which were not identified.

Particle 10 consisted primarily of tantalum carbide with traces of tantalum oxide, in both the alpha and beta form, and some tantalum metal.

Particle 14 was primarily tantalum carbide and tantalum oxide, with traces of tantalum metal. The tantalum oxide was identified as Ta_2O_5 in the beta form.

High Volume Air Sample

A high volume air sampler suspended directly over the reactor was heavily laden with particulate matter and the activity was so high that gamma spectrum analysis was impractical. Two weeks after the sample was collected the activity was essentially zero. The small amount of activity that did remain was not contained in a single particle but was evenly dispersed over the entire glass filter. The large amount of residue on the filter was from the JATO smoke used to identify the cloud emitted from the reactor.

JATO fuel, 14AS1000 Type, contained a 79 pound solid propellant grain with potassium perchlorate as the oxidizer and an asphalt-oil mixture as the binder-fuel. The 14AS1000 JATO weighed 194 pounds loaded, and 115 pounds was expended. The potassium chloride resulting from the combustion of the propellant is the prime source of smoke from the unit. Combustion products obtained when using potassium perchlorate as the oxidizer are: potassium chloride (solid), water, carbon dioxide, carbon monoxide, unreacted carbon (solid), and hydrocarbons. The per cent of

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unreacted hydrocarbons and carbon is controlled by the percentage of oxidizer and by combustion catalysts.

Much of the residue fell off the filter during transportation. Microscopic examination of some of the dust from the filter, and microchemical reactions, confirmed the presence of potassium chlorate, much potassium chloride, and some carbonate. A few black granules were identified as graphite. The white residue adsorbed on some of the particles came from the JATO smoke. The residue was eluted with distilled water, and a microdrop of this wash tested by precipitation and microcrystallography techniques. The presence of chloride, potassium, carbonate, and chlorate ions was confirmed; the water washing from the particle contained only traces of beta activity.

The filter was washed with distilled water until reasonably clean and the washings were centrifuged. The supernatant was decanted from the residue and the residue again washed with water and centrifuged, and the supernatants combined. The residue was ashed with nitric and perchloric acids until it was white, transferred to a platinum crucible, and evaporated to dryness. The dried residue then was dissolved in 100 microliters of a solution which was 4N in sulfuric acid and 4N in hydrofluoric acid. Five microliters were transferred to a tantalum planchet, evaporated to dryness, and flamed to red heat. The planchet was counted for alpha and beta activity. The alpha activity in the residue was approximately 30 d/m/sample and the beta activity was 1670 d/m/sample. A 5 microliter portion was placed on a chromatographic strip and treated

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as described previously. X-ray film was exposed to the strip and the areas normally occupied by zirconium and niobium were identified. A longer exposure did not indicate any other R_f areas. The remaining residue was transferred to a plastic weighing bottle for gamma spectrum analysis, but the activity was too low for photo-peak identification.

The supernatant from the filter contained all of the water soluble compounds and essentially no activity. The presence of the ions mentioned above was confirmed by chemical tests.

SUMMARY

During the test operation of the Kiwi-A3 reactor at the Nevada Test Site on October 18, 1960, the reactor emitted many large particles along with the expected fission gases and the more volatile nuclides. The particles ranged from 35 to 2500 microns in diameter, and were extremely friable. The inert portion of the particles contained graphite, tantalum, niobium, and trace elements; the radioactivity was contributed primarily by the activation of the tantalum to form Ta^{182}. The remaining activity came from the normal distribution of fission products of uranium impregnated in the graphite.

The location of the particles in the test area was a function of reactor gas effluent velocity and time of corrosion and erosion rather than of particle size. There was no correlation between particle size and distribution along the fallout pattern. Two four-stage cascade impactor slides from the area did not contain any uranium or tantalum.

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Some of the particles were coated with the JATO smoke used to track the effluent gas cloud, indicating that the particles actually were part of the cloud. A plume sample showed only decay daughters of gaseous fission products, with no uranium or tantalum.

Radiometric procedures, such as gamma spectrum analysis, beta absorption studies, x-ray film and Polaroid film radioautography, and gross beta and gamma counting, were applied. Radioautography proved extremely useful in determining the relative activity of particles found on surfaces of static sampling units as well as those isolated from the ground; although the size of the exposed spots formed during the study did not correlate with particle size, the specific activity of a given size particle could be calculated. Beta absorption studies proved of little value except when absorbers must be used to reduce the beta activity for gross counting. The gamma spectrum analysis 34 days after the day of the run was more significant than the spectrum determined immediately after the run because of the excessive tantalum activity and the finding that the particles contained uranium and thus most of the expected long-lived fission products.

The chemical and physical methods used were spectrography, x-ray diffraction, microchemical qualitative techniques, paper chromatography, and standard quantitative procedures. They showed that chemical procedures applied immediately after a nuclear reactor test, to particles or to static sampling systems, must be rapid and can be only qualitative, unless specific isotopes are chosen for a specific decay series.

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Included in this report are those techniques which seem applicable in a future operation producing a similar type of fallout debris. A procedure is given for the isolation of particles without undue contamination or loss. X-ray film radioautography and Polaroid film fallout trays are described. The paper chromatographic procedure developed for these particles is outlined.

CONCLUSIONS

The discrete particles found in the fallout area were from the reactor fuel elements where tantalum oxide had been applied. The particles were eliminated from the reactor at different times during the run. The large particle size precluded any health hazard from inhalation, but did contribute to the integrated dose rate. The amount of uranium in the particle after hydrolysis of the carbide becomes a measurable ground contaminant. All of the tantalum carried out of the reactor in the form of Ta^{182} was contained in large particles; essentially no uranium was found in the fallout area samples, as determined from analysis of the plume and cascade impactor samples. The fission products found in the fallout area were short-lived, while those associated with the particles were primarily normal fission products because of the fission of the uranium contained in the particles before elimination of the particles from the reactor.

Those procedures which seem applicable to future reactor environmental health studies are the techniques of particle isolation and Polaroid film fallout trays. X-ray film radioautography should be an

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integral part of the standard fallout tray examination, to determine the distribution of particles based on their activity and size, as well as their specific activity where possible. The chromatographic technique has limited application for presumptive group identification because large amounts of radioactivity must be confined in a small volume. Aluminum absorbers can be used to reduce gross beta activity for counting purposes, provided that more than one absorber density count is taken.

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APPENDIX A

ISOLATION OF PARTICLES FROM SOIL AND FROM FALLOUT TRAYS

Equipment

Beta-Gamma Portable Survey Instrument

Low Power Magnifier

(A shop microscope or similar optical system having 60 to 200X magnification is preferred.)

Beta Particle Probe

(The probe used in the study, prepared by the Electronics Group, consisted of a 1/4 inch diameter by 1/8 inch thick beta phosphor coupled to a 1-1/2 inch photomultiplier tube by a light pipe. The unit was operated at maximum voltage for lowest background. The response of the tube was measured through a proportional counter to a count rate meter and recorder.)

Particle Holder

(The particle holder is essentially that described by Mackin, et al.[6] Insert a dissecting needle through a #000 cork and wipe with a thin coat of silicone grease, then store in a 2 ml glass vial. Sharpen the needle or pull a glass rod to a fine tip for finer manipulations.)

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Technique

Soil samples

When the soil sample is received in the laboratory, spread it on a glass plate and divide it into two portions. Check each portion for beta-gamma activity with a portable instrument, and discard the half containing no activity. Divide the remaining half again, check for activity, and discard the half containing no activity. Repeat the halving process until only a few grains of dust remain on the plate. Accomplish the final separations with a small brush and a lead shield. Finally, observe the particles under low power, and, with the aid of the beta probe, pick up small particles with the dissecting needle. Often the particle can be selected by its color or surface character. Usually there is only one particle in the soil, with a small amount of activity abraded from it; however, if care is taken when the soil is selected in the field, abrasion will be minimal. Store the needle and the selected particle in the glass vial until ready for analysis.

Fallout trays or other surfaces

Scan fallout trays, which are to be examined for single particle activity, with a portable beta-gamma instrument. After isolating a given area of activity scan the sector with the beta probe to narrow the area to less than 1 sq.cm. Mark the area clearly with a pen or other marking tool. Observe the area under low power to identify the particle.

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Resin or resin-coated trays

Place a small circle of filter paper over the particle and, with a fine-tipped suction tube and a micro dropper, add acetone to one side of the filter circle and aspirate the acetone from the opposite side. The acetone removes the resin or oil and the particle usually is picked up by the fibers of the filter disc. Then pick up the particle with the needle for further study, for storage, or for shipment elsewhere.

Use the needle in the field to isolate particles from surfaces which normally are free from dirt, such as stationary fallout trays, or building surfaces on which spot activity has been found.

Note:

The particles may be gamma counted directly without removal from the vial and may be studied under low magnification directly. The particle may be transferred easily to laboratory ware or to microscope slides for further examination. For microscopy, the particles may be transferred directly to a cavity microscope slide and covered with a cover slip.

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APPENDIX B

X-RAY FILM RADIOAUTOGRAPHY

Introduction

Many authors have discussed at length the use of x-ray film radioautography, but its application to field operations has been limited to weapons tests in which discrete particles were involved. This technique was not applied to the Kiwi-type reactor because the results of the study did not seem to warrant the expenditure of time and effort. Several factors which occurred with the past series of Kiwi tests, as well as the operational design of future reactors, changed this opinion. Fallout trays, poor as they are for collecting fallout data, still may be the best technique for stationary sampling of ground deposition. As in Kiwi-A3, the reactor corroded and eroded. The eroded particles fell within the test area and were counted on fallout trays as if they were representative of the general ground deposition. Soon after a run the specific activity of the individual particles is relatively high and is suitable for x-ray radioautography.

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Mylar films on fallout trays in field operations are not advisable because the mylar is difficult to handle and may become contaminated. The trays normally are sandwiched together face to face at pickup, which destroys the normal distribution of particles but gives the same total activity deposition; although larger particles are crushed, they are plainly visible and the total activity of the two trays remains unchanged.

After the trays have been returned to the laboratory, sealed in polyethylene, and counted at least once, they can be exposed to x-ray film for a known length of time.

Equipment

Kodak Type K X-ray Film

(Number each film and package each in individual, light-tight envelopes, 11 x 14 inches. Store all of the film in an area removed from possible radiation.)

Exposure

Place the trays in pairs face down on the film, always keeping the A and B trays in the same order. Allow the trays to remain in position for a given length of time as shown below:

Surface Reading (mr/hr)	Exposure Time (minutes)
5.0	15
1.0	30
0.5	60

Record the following information:

Time of Exposure .... minutes after midpoint
Length of Exposure .... minutes
Film Number
Tray Number

Expose the trays to the x-ray film, and store the films at some point removed from the general area of radiation to prevent fogging of the film. Expose only a few trays at a time, depending on the operational surface area available for the study. Develop the x-ray film in the usual manner and dry it as rapidly as possible for study.

Interpretation

1. General fogging of the film: indicates even dispersion of fallout in the area represented by the tray.

2. Many small black spots on the film: represent particulates that could be either discrete particles or adsorbed activity onto inert dust particles.

3. Large black spots on the film: represent relatively large, discrete particles which may be removed by the particle handling technique described above, if desired.

4. Combinations of these: represent the type of distribution of the particles throughout the fallout test area.

It would be possible, then, from the above data, to establish the distribution of particles throughout the test area, based on the relative size of the particles. Single particles can be removed from the general debris of the tray and followed separately, if desired.

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APPENDIX C

POLAROID FILM RADIOAUTOGRAPHY

Introduction

Polaroid film is used to isolate given particles, size them as rapidly as possible, and identify their origin. Polaroid film for field work connected with nuclear devices has not been satisfactory; however, some of the problems associated with its use have been overcome. Polaroid film does offer a rapid method for observing nuclear events occurring on the surface of the film, providing background radiation is minimal. Particles falling onto the film surface expose both the paper and the film and thus record the particles' location on the film surface. Locating the particles by low power magnification is rapid.

Equipment

Trays

(10 x 12 x 3/16 inch Masonite sheets with a 1/2 x 3/16 x 12 inch strip of Masonite glued along opposite edges and a 3/16 inch hole drilled in the center of each strip. The holes are used to stack the

[redacted]

trays for placement and collection and also are used to nail the tray to the ground to prevent the tray from blowing away or turning over.)

Film

[Type 37 (ASA 270) and Type 57 (ASA 3000) Polaroid single sheet 4 x 5 inch film.]

Holder

(Sheet film adaptor #500 for 4 x 5 inch camera, to be used for development of the film.)

Preparation of Tray for Use in Field

Place a sheet of each of the film types on the Masonite tray with the film face toward the tray, and tape along the center between the two films to prevent excessive moving of the film. Cover the film with a 10 x 12 inch sheet of black glazed paper (weighing paper) and tape the paper to the tray with masking tape along the three flat edges, then fold the overhanging edge under the tray and tape to the back of the tray. Be careful not to break the developer pod in the film pocket. The tray may be left unrefrigerated for as long as a week without demonstrable change in the films' response. Just before placing in the field, spray the glazed paper surface with a thin coat of Dow Corning Antifoam A.

Exposure

Place the trays on the ground in the field and hold in position with two large nails through the holes in the tray. Place each tray

[redacted]

so that direct radiation from the reactor is minimized. Smaller particles can be detected by this technique because their specific activity is extremely high within a few minutes after a nuclear event, so they begin to expose the film immediately upon deposition; the particles decay so rapidly that they would be missed at later times.

Development and Study

After the exposure has been completed and the trays collected, cut the paper along the folded edge and remove the films carefully to prevent separation of the film pack itself. Place the films in the holder and develop them in the usual manner. Immediately after developing examine each film for the presence of particles, then replace film and paper in the tray for future reference; discard those trays void of activity. Disregard the pinhole white areas observed at regular intervals throughout a series of films. If desired, individual particles may be isolated by microscopy and the technique described above for particle location.

Note:

When films can be attached directly to a surface, the films may be packaged in the glazed paper by folding the paper over three times, taping the double fold together on the face side of the film, and sealing each end. Then spray the back of the paper with the Antifoam and expose by clipping or taping the package to the surface.

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APPENDIX D

CHROMATOGRAPHY OF FISSION PRODUCTS

Introduction

For chromatography, samples from many sources are reduced to a pure residue and dissolved in a mixed acid. An aliquot is placed on a multiple strip CRL/1 chromatography paper and chromatographed with an acetone-HF-HCl-HOH solvent. The new technique was necessary for field use because the rapid decay of the early fission products would give a diffuse separation in the butanol solution used in other studies. The migration time on an unequilibrated paper is 12 minutes as compared with 45 minutes using butanol. Given below, in the order of their relative activity, are the nuclides in fallout at 1 hour, 1 day, and 10 days (99 per cent of the total activity is included in the isotopes listed).

[redacted]

1 HOUR	1 DAY	10 DAYS
Te (some)	I*	I*
Kr*	Xe*	Sr
Rb	Y	Te (some)
Ce	Te (some)	La
I*	Sb (some)	Ba
Ba	Ce	Y
Xe*	Kr*	Ce
Br*	Sr	Pr
La	Nb	Sb
Y	La	Nb
Sb (some)	Ba	
Se (some)	Zr	
	Pr	

*Those elements which are lost are marked with an asterisk.

In addition to these elements, activated elements also must be considered, i.e., Ta, Mo, W, and Ti. The chemical procedure used to reduce the samples to a chromatographic form causes a loss of many of the more volatile isotopes. The experimental work using this technique has been reported by several authors[7,8,9] and has been modified for application to this particular situation. Below are the elements listed above, with the reported R_f value for each. For a more complete list of elements and their R_f value, see Table I, Appendix D, page 52.

[redacted]

R_f	ELEMENT
1.0	Ta, W, Se
0.83	Mo
60.86	Sb
0.82	Nb
0.64	Ti
0.48	Zr
0.24	La
0.15	TeO_3^{--}
0.05	Rb
0.00	Y, Ba, Ce TeO_4^{-}, Sr

Reagents

Prepare the following reagents from analytical reagent grade chemicals; store each concentrated reagent in its original container:

Sulfuric Acid	96%	Hydrofluoric Acid	48%
Nitric Acid	71%	Ammonium Hydroxide	31%
Hydrochloric Acid	36%	Perchloric Acid	70%

Digestion Mixture

To 60 ml of conc. nitric acid, add 20 ml of conc. sulfuric acid and allow to cool to room temperature. Add 20 ml of conc. perchloric acid and store the mixed acids in a glass storage bottle fitted with a syringe dropper.

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[redacted]

Dissolving Solution

To 25 ml of distilled water in a graduated cylinder, add 10 ml of conc. sulfuric acid and cool. Transfer to a polyethylene storage bottle and add 10 ml of conc. hydrofluoric acid.

Chromatography Solvent

To 90 ml of acetone in a polyethylene storage bottle, add 5 ml of conc. hydrochloric acid, 4 ml of distilled water, and 1 ml of hydrofluoric acid. Prepare the solution fresh daily, always stopper it tightly, and refrigerate it when not in use. The reagent need not come to room temperature before use.

CRL/1 Chromatography Paper[10]

(Paper shape cut 4-1/2 x 8 inches, slotted to permit simultaneous running of ten ascending chromatograms.) H. Reeve Angel and Company, Inc., 52 Duane Street, New York 7, New York.

Reduction of Samples for Paper Chromatography

Single Particles

Place the particle in a platinum crucible and treat with 1 ml of digestion mixture; evaporate to copious fumes of sulfur trioxide in an aluminum block adjusted to 350°C. After the organic matter is ashed, treat the residue with several drops of conc. hydrochloric acid and evaporate to dryness. Dissolve the residue in 100 microliters (0.1 ml) of dissolving acid mixture, using a platinum stirring rod to wash down the sides of the crucible. Transfer a 5 microliter (0.005 ml) aliquot to a tantalum planchet and evaporate to dryness for alpha counting.

[redacted]

[redacted]

Place a third 5 microliter aliquot on a 7/8 inch diameter glass filter to be counted for beta and gamma and followed for decay.

Filter Papers

This technique is applicable provided no large amount of foreign material is on the filter sample. Fold carefully paper swipes, air samples, and filter paper or other cellulose samples; place in a platinum crucible; wet with conc. nitric acid; and evaporate to dryness at 150°C. Repeat the acid-evaporation until only a small amount of char remains in the crucible, then add 2 ml of digestion mixture and evaporate to copious fumes of sulfur trioxide. Treat the residue with several drops of conc. hydrochloric acid and again evaporate to dryness. Dissolve the residue in 100 microliters of dissolving solution, carefully washing down the sides of the crucible with a platinum stirring rod. Transfer a 5 microliter aliquot to a chromatographic strip. Transfer a second 5 microliter aliquot to a tantalum planchet and evaporate to dryness for alpha counting. Place a third 5 microliter aliquot on a 7/8 inch diameter glass filter to be counted for beta and gamma and followed for decay and growth.

Glass Fiber Filters

Fold the glass filter carefully, place in a platinum crucible, and hold in the crucible with a stirring rod. Slowly add conc. hydrofluoric acid until the filter is completely disintegrated. The HF must be added to the filter cautiously, because the exothermic reaction

[redacted]

[redacted]

between the HF and the filter fibers causes evolution of a large amount of free hydrogen fluoride.

After the filter is completely wet with the HF, place the crucible on a heating block at 150°C. and evaporate to dryness. Add several drops of digestion mixture and evaporate again to dryness. When dry, or when fumes of sulfur trioxide no longer are visible, add 2 to 3 ml of conc. nitric acid and several crystals of boric acid, warm, dilute to about 10 ml with water, and warm again. Transfer the solution to a Lusteroid centrifuge tube and centrifuge. Transfer the supernatant solution to a clean platinum crucible. Wash the residue out of the Lusteroid tube with several drops of conc. HF and several ml of water, and transfer it to the original platinum crucible, treat with about 1 ml of conc. sulfuric acid, heat to boiling, then transfer with water to the same centrifuge tube and centrifuge again. Transfer the supernatant solution to the second crucible and evaporate the combined washings to dryness. Treat the residue with sulfuric acid and hydrofluoric acid until all of the silicates and silica have been volatilized. After all the silica has been removed, evaporate the sulfuric acid, dissolve the residue in 100 microliters of the dissolving acid, and place a 5 microliter aliquot on a chromatograph strip. Transfer a second 5 microliter aliquot to a tantalum planchet and evaporate to dryness for alpha counting. Place a third 5 microliter aliquot on a 7/8 inch diameter glass filter to be counted for alpha and beta and followed for decay and growth.

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Chromatography

Place 5 microliters of the aliquot to be tested on CRL/1 chromatography paper. Prepare the paper by drawing a line with a ballpoint pen across all 12 strips, about 3 mm above the cut. Place the aliquot over the line; add a second 5 microliter aliquot to the strip if the activity is considered too low for the counting technique to be used. The sulfuric acid that is present will char the paper if allowed to stand more than several hours in the open. Make the chromatogram as soon as possible after adding the aliquot.

Label the strips with a pencil and chromatograph by curving the paper and placing it in a polyethylene chamber (one quart Teflon storage bottle cut flat on the top proved the best chamber for this purpose). The solvent system should be about 1/4 inch deep in the chamber. Cover the chamber with a glass plate or watch glass and allow the solvent to migrate to within 0.5 cm of the top of the cut. The front of the solvent is visible only by the migration of the blue from the ballpoint ink marking (the point of addition is marked by a black line). Air dry the paper for several seconds to remove the excess acetone and then expose to ammonia vapors in a similar chamber in which conc. ammonium hydroxide is contained in a small beaker. Fan the ammonium salt fog out of the chamber occasionally to observe when the paper is completely neutralized. Remove the paper and cut into strips; measure in mm and record the distance from the point of addition to the front. Tape the strip on a drum counter calibrated so that the front of the chromatogram

[redacted]

can be marked on the accompanying count-rate recorder. After making the activity record, cut the paper according to the distribution of the activity. The predetermined cutting points are:

R_f	Possible Element
0.00 to 0.10	Y, Ba, Ce, Sr, R, Te
0.10 to 0.50	La, Zr
0.50 to 0.90	Ti, Sb, Mo
0.90 to 1.00	Ta, W, Se

Package each of these areas in a nylon ring and disc assembly, and count for beta activity in a plastic scintillation counter. Compare the total activity to that in the direct-plated aliquot to determine the ratio of the activities, then submit the sections and the direct plate to gamma spectrometry for more precise identification of the isotopes present.

Because of the fractionation that occurs in the original sample during the ashing and dissolving of the filters, take additional aliquots at 3, 6, 12, 24, and 48 hours after plating the first sample. Follow the chromatograms for decay in order to establish the relative growth and decay of the activity with respect to the direct plate. The additional aliquots are for establishing with certainty the original constitution of the activity present. Soon after fission the migration of daughters of the elements having an R_f of 0 will cause a diffuse pattern of activity. For example, the daughters of tellurium would be distributed from R_f 0 to R_f 1 because cesium has an R_f of 1, and tellurium an R_f of 0.

[redacted]

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TABLE I. DESCRIPTION OF DISCRETE PARTICLES

Particle No.	Reconstructed Size Appx. Microns	Number of Fragments	Description	Distance from Test Cell (ft)	Beta Activity Extrapolated to 1000 Min (d/m x 10^7)
1	200	1	Black and brassy metallic with yellow fragments	6,500	0.62
2	200	1 (broken in half)	Yellow amorphous coating with black metallic particle (the coating contained no activity	6,700	2.4
3	400	1	Brassy, granular, some included silica, one edge rusty	7,000	2.4
4	200	1	Black and brassy metallic with yellow fragments (Photo 3)	6,500	0.58
5	35	1 (crushed)	Black and brassy metallic with yellow fragments	6,000	0.58
6	150	1 (cracked)	Black and brassy metallic granules	6,500	0.04
7	175	3+ (crushed)	Black and brassy metallic with yellow fragments	6,700	0.018
8	800	4 (crushed)	Gray-white added residue on black granular material (Photo 2)	1,100	----
9	500	1	Gray-white coating on black granular material, smooth surface	1,000	0.6
10	450	6	Black metallic granules	1,200	0.7

[redacted]

[redacted]

TABLE I. (Continued)

Particle No.	Reconstructed Size Appx. Microns	Number of Fragments	Description	Distance from Test Cell (ft)	Beta Activity Extrapolated to 1000 Min ($d/m \times 10^7$)
11	350	4	Black metallic granules with one rusty edge	1,400	----
12	475	1 (originally; crushed and dispersed)	Mixed black and brassy metallic granules, some with gray portions containing white amorphous coating	3,000	1.1
13	400	3	Black and brassy metallic granules, with yellow fragments and several silica crystals	7,000	0.5
14	2,500	1	Black, granular, with brassy edge	1,500	----
15	2,100	1	Black, granular, with a brassy green luster	1,000	----
16	(800)*	1	Used for gamma spectra studies at NTS	1,000	----
17	(650)	11 (Crushed and dispersed)	Black, metallic, granular, very friable	1,100	----
Special Soil	(200)	1	Black, metallic granule	6,000	----
20	100	3+	Many black granules containing activity; no single particle found (crushed in transportation)	2,000	----

*Estimated at NTS

[redacted]

[redacted]

TABLE I. (Continued)

Particle No.	Reconstructed Size Appx. Microns	Number of Fragments	Description	Distance from Test Cell (ft)	Beta Activity Extrapolated to 1000 Min ($d/m \times 10^7$)
21	50	2	Black granules	2,000	----
22	50	1	Black granule with some gray-white residue and a small, brassy area	2,000	----
23	35	1 (identified)	Black granule	2,000	----
24	150	1	Black granule	2,000	----
25	200	1	Black granule	2,000	----
Fallout Trays by Station Number					
4-24-A B	(500)	Crushed	Black granular, with some brassy fragments	4,000	0.68 0.0016
6-24-A B	(1,000)	Crushed	Black granular (difficult to determine size of the dispersed particles)	6,000	----

[redacted]

[redacted]

TABLE II. RATIO OF NUCLIDE ACTIVITY TO TOTAL GAMMA ACTIVITY

Particle No.	Ratio of Gamma Activity (34 days past 0)			Beta Activity (d/m) 11/7/60 32 days past 0
	$Zr\text{-}Nb^{95}$ Peak 0.75 mev	La^{140} Peak 1.6 mev	Ta^{182} Doublet 1.1 and 1.2	
1	0.112	0.014	0.349	69,977
2	0.111	0.025	0.413	326,457
3	0.090	0.019	0.289	276,395
4	0.099	0.023	0.321	61,980
5	0.115	0.036	0.303	76,302
6	0.109	0.017	0.346	4,587
7	0.109	0.020	0.348	2,331
9	0.0	0.0	0.3574	68,611
10	0.0	0.0	0.354	77,435
11	0.093	0.024	0.316	89,502
12	0.080	0.022	0.284	156,715
13	0.135	0.022	0.311	61,620
15	0.105	0.021	0.279	310,529
17	0.121	0.017	0.281	522,155

[redacted]

[redacted]

TABLE III. EQUIVALENT BETA ENERGY (from loss of activity by aluminum absorbers)

Particle No.	Loss of 99.0% Activity E_{max}		Loss of 50.0% Activity $E_{\frac{1}{2}}$	
	Absorber mg/cm^2	Energy mev	Absorber mg/cm^2	Energy mev
1	325	0.82	22	0.12
2	420(98%)	1.07	10	0.083
3	360	0.87	26	0.147
4	285	0.77	25	0.146
5	525	1.17	45	0.21
6	Activity too low for counting			
7	Activity too low for counting			
8	675	1.5	20	0.125
9	575	1.32	30	0.16
10	675	1.5	22	0.135
11	380	0.95	40	0.192
12	460	1.1	60	0.25
13	450	1.08	45	0.21
14	740	1.5	30	0.16
15	455	1.12	42	0.20
16	Used for chemical analysis			
17	490	1.16	44	0.21
Cs^{137}	185	0.56	15	0.106
Ta^{182}	350	0.9	17	0.125

[redacted]

[redacted]

TABLE IV. X-RAY FILM RADIOAUTOGRAPHY (1 hour exposure)

No.	D Diameter of Part. μ	d Diameter of Autograph mm	Total Disintegrations X 10^7	Disintegrations per d^2 auto. X 10^5	Disintegrations per D^2/particle X 10^4
1	200	3.	0.42	4.6	1.05
2	200	5.	1.95	7.8	4.85
3	400	4.5	1.07	5.3	0.65
4	200	3.0	0.037	4.	0.09
5	35	3.5	0.46	3.7	0.04
6	150	1.0	0.027	2.7	0.12
7	175	0.7	0.014	2.8	0.05
9	500	2.5	0.41	6.5	0.16
10	450	3.3	0.464	4.2	0.23
11	350	3.5	0.536	4.3	0.43
13	400	2.9	0.37	4.3	0.19
14	2500	22.	5.5	11.	0.88
15	650	5.	1.86	7.4	0.44

[redacted]

[redacted]

TABLE V. ALPHA AND BETA ACTIVITY OF SELECTED PARTICLES*

Particle No.	d/m alpha**	d/m beta	beta-alpha ratio
2-A	732.	39,140	53.6
11	640.	18,300	28.6
6	18.8	1,310	69.6
1	96.	7,640	79.7
12	872	24,500	28.1
3	704	41,200	58.4

*Plated directly on 3/4" tantalum planchets

**Confirmed as U by ion exchange procedure

[redacted]

TABLE VI. SPECTROGRAPHIC ANALYSIS OF PARTICLE 14

ELEMENT	CONCENTRATION RANGE
Tantalum	Greater than 10%
Niobium	1 - 10%
Platinum	0.1 - 1%
Titanium	0.1 - 1%
Silica	0.1 - 1%
Sodium	0.01 - 0.1%
Aluminum	0.01 - 0.1%
Calcium	0.01 - 0.1%
Iron	0.01 - 0.1%
Rhodium	0.01 - 0.1%
Zirconium	0.001 - 0.01%

No detectable lines observed for: Li, Be, B, P, Sc, V, Cr, Mn, Co, Ni, Cu, Zn, Sr, Y, Mo, Ru, Pd, Ag, Cd, In, Sn, Sb, Ba, La, Hf, W, Ir, Au, Pb, Bi, Th, and U.

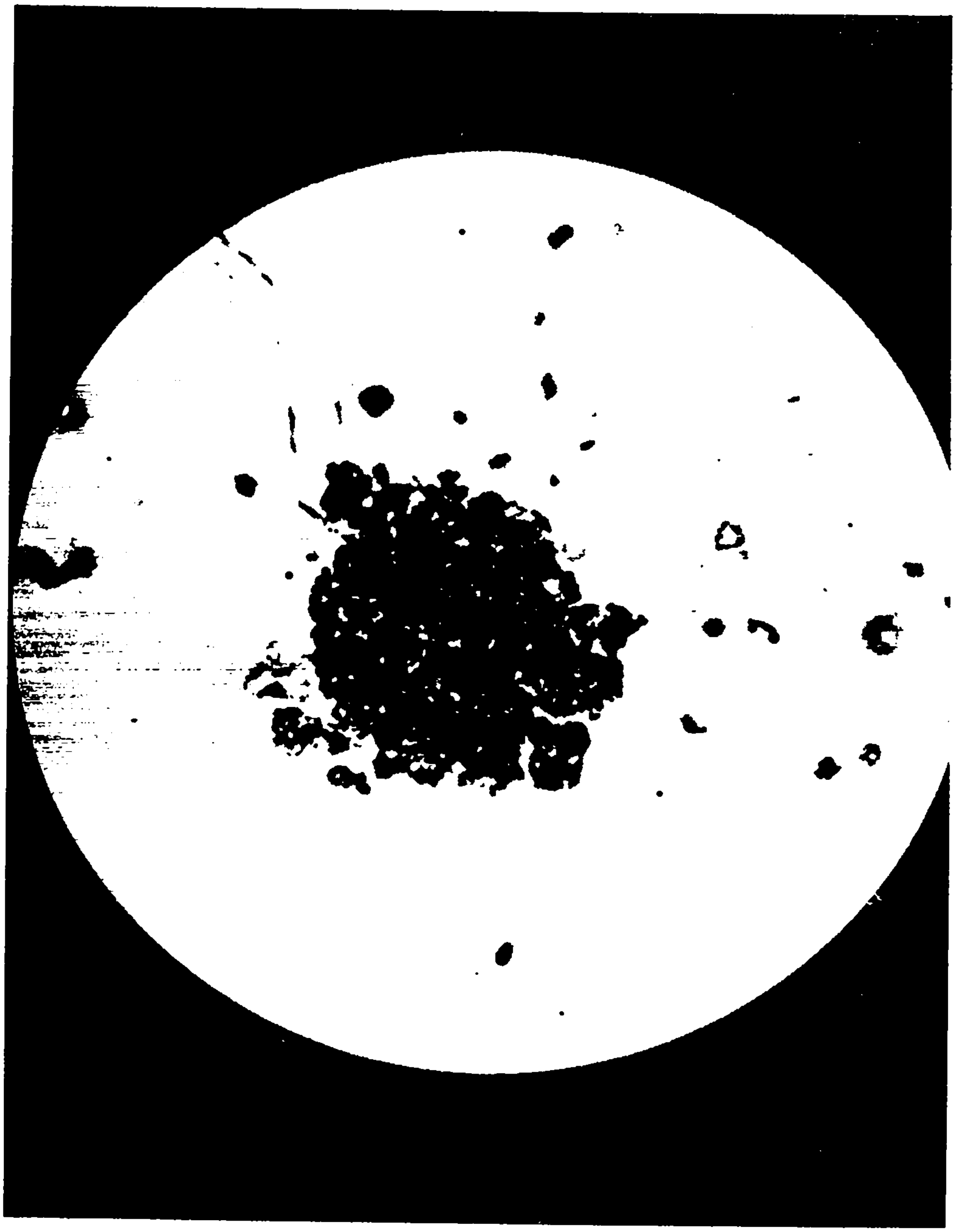
[redacted]

TABLE I. APPENDIX D. R_f VALUES OF IONS IN HCl-HF-ACETONE-HOH ON PAPER CHROMATOGRAMS

R_f Value	Ions	R_f Value	Ions
1.00	SeO_3^{-2}	0.46-0.59	Ag^{+1}
1.00	Ta^{+5}	0.48	Pb^{+2}
1.00	Hg^{+2}	0.48	Zr^{+4} Comet
0.94	Nb^{+5}	0.48	SO_4^{-2}
0.93	Pt^{-2}	0.46	Hf^{+4} Comet
0.92	Hg^{+1}	0.45	Co^{+2}
0.92	MoO_4^{-2} Comet	0.29	Ge^{+4}
0.90	Zn^{+2}	0.27	Cr^{+3}
0.86	Fe^{+3}	0.25	Mn^{+2}
0.85	Pd^{+3}	0.22	Be^{+2}
0.82	Sb^{+3}	0.16	TeO_3^{-2}
0.82	Sn^{+2}	0.05	K^{+1}
0.82	In^{+3}	0.05	Rb^{+1}
0.77	Cd^{+2}	0.04	Li^{+1}
0.68	Ga^{+3}	0.03	Ni^{+2}
0.65	As^{+3}	0.03	Mg^{+2}
0.64	UO_2^{+2}	0.03	Cs^{+1}
0.64	Ti^{+4}	0.02	Al^{+3}
0.64	V^{+5}	0.0	$Ca^{+2}Sr^{+2}Ba^{+2}$
0.62	Pa^{+5}	0.0	$Th^{+4}La^{+3}Ce^{+3}$
0.59	AsO_4^{-3}		Y^{+3} Sc^{+3}

[redacted]

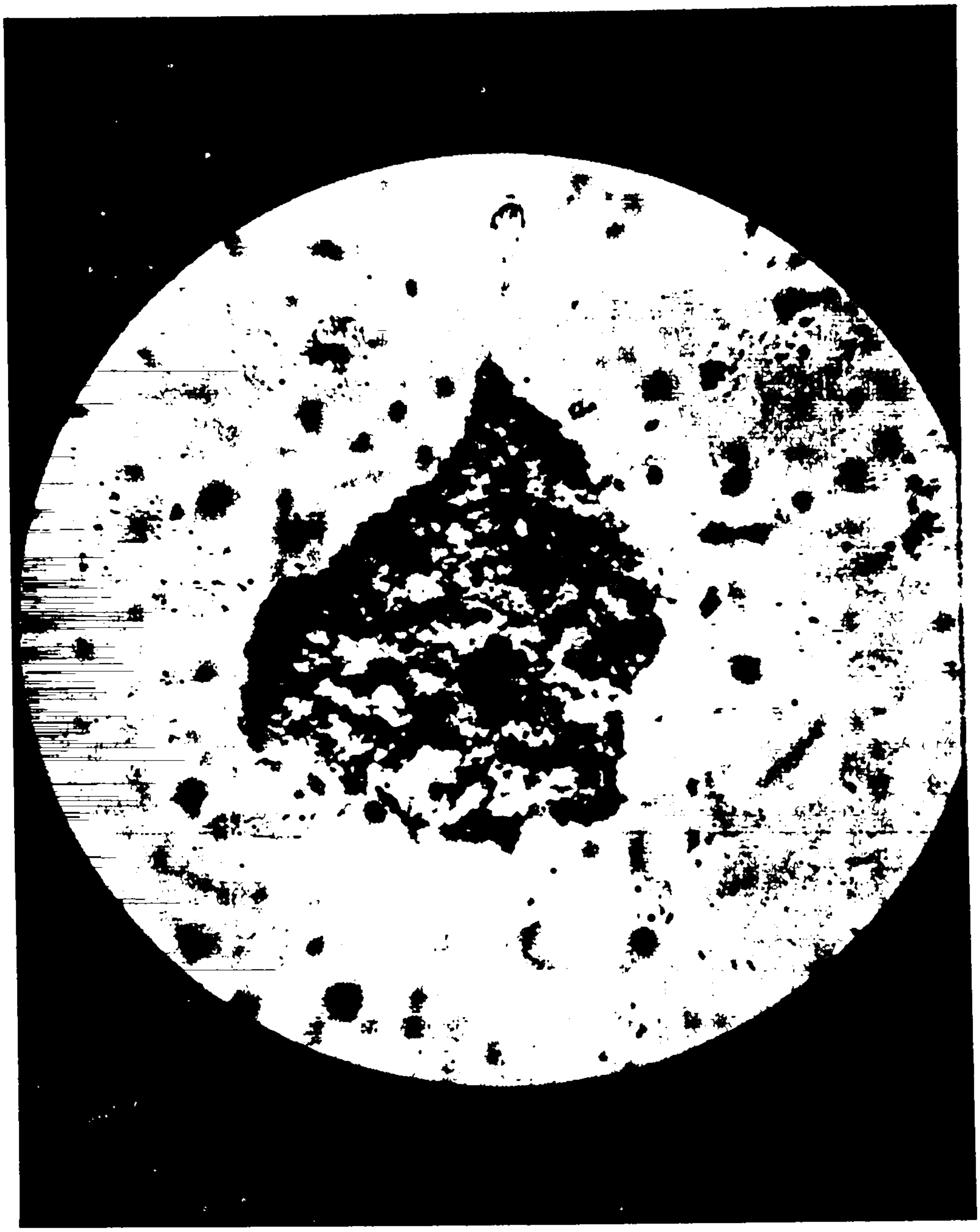
[redacted]



Photograph 1. Particle 12

Friable Black Granular Particle

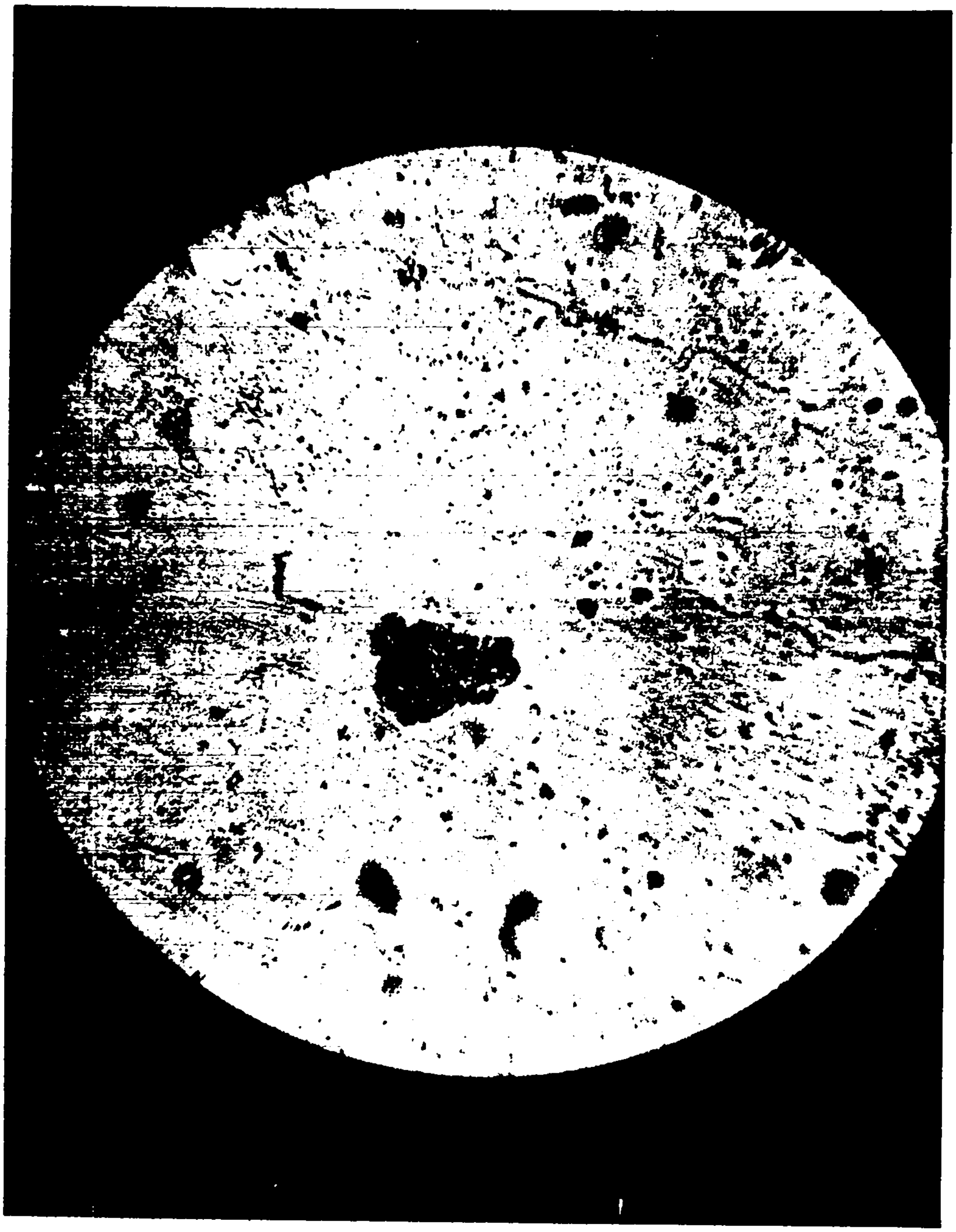
[redacted]



Photograph 2. Particle 8

Balck Granular Particle with White Added Residue

[redacted]



Photograph 3. Particle 4

Black Granular Particle with Metallic Luster

[redacted]

Photograph 4.

Polaroid Film Radioautograph

Enlargement of a radioautograph on Type 57 Polaroid negative. The solid black, irregular shape is a facsimile of the particle used. (The radioautograph would be reversed on the negative.)

[redacted]

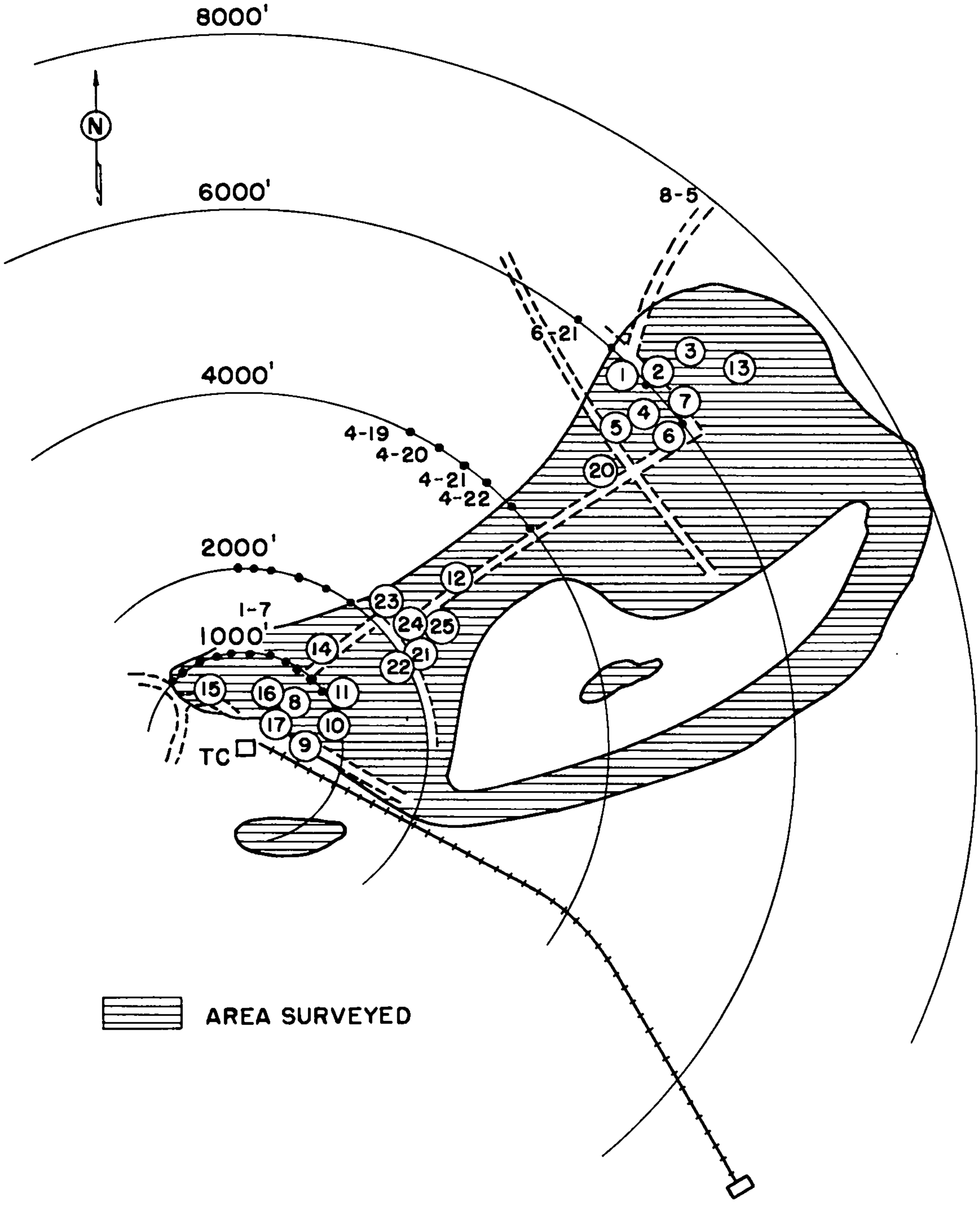


Fig. 1. Ground Survey Area and Particle Location

[redacted]

[redacted]

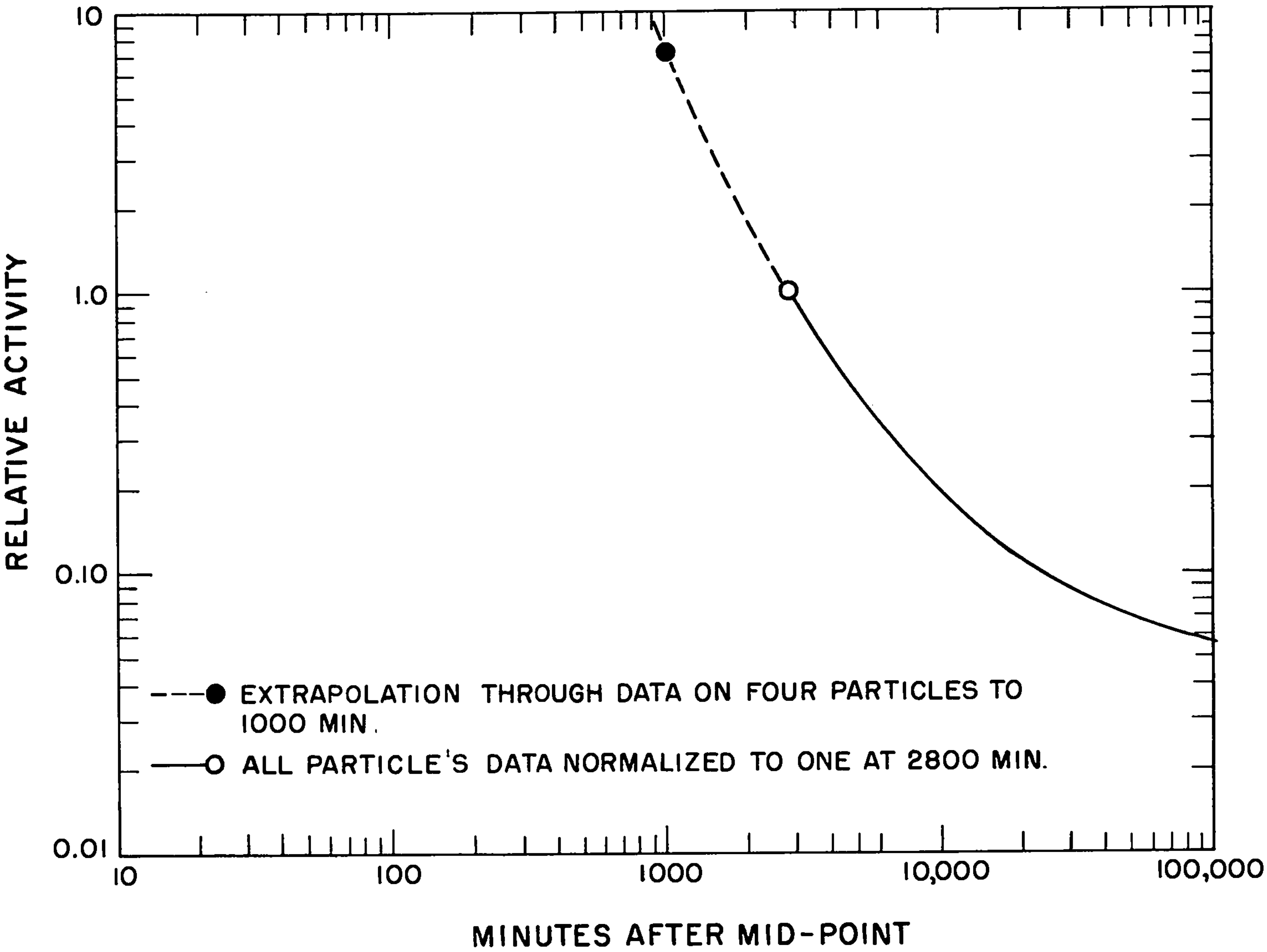


Fig. 2. Composite Beta Decay Curve

[redacted]

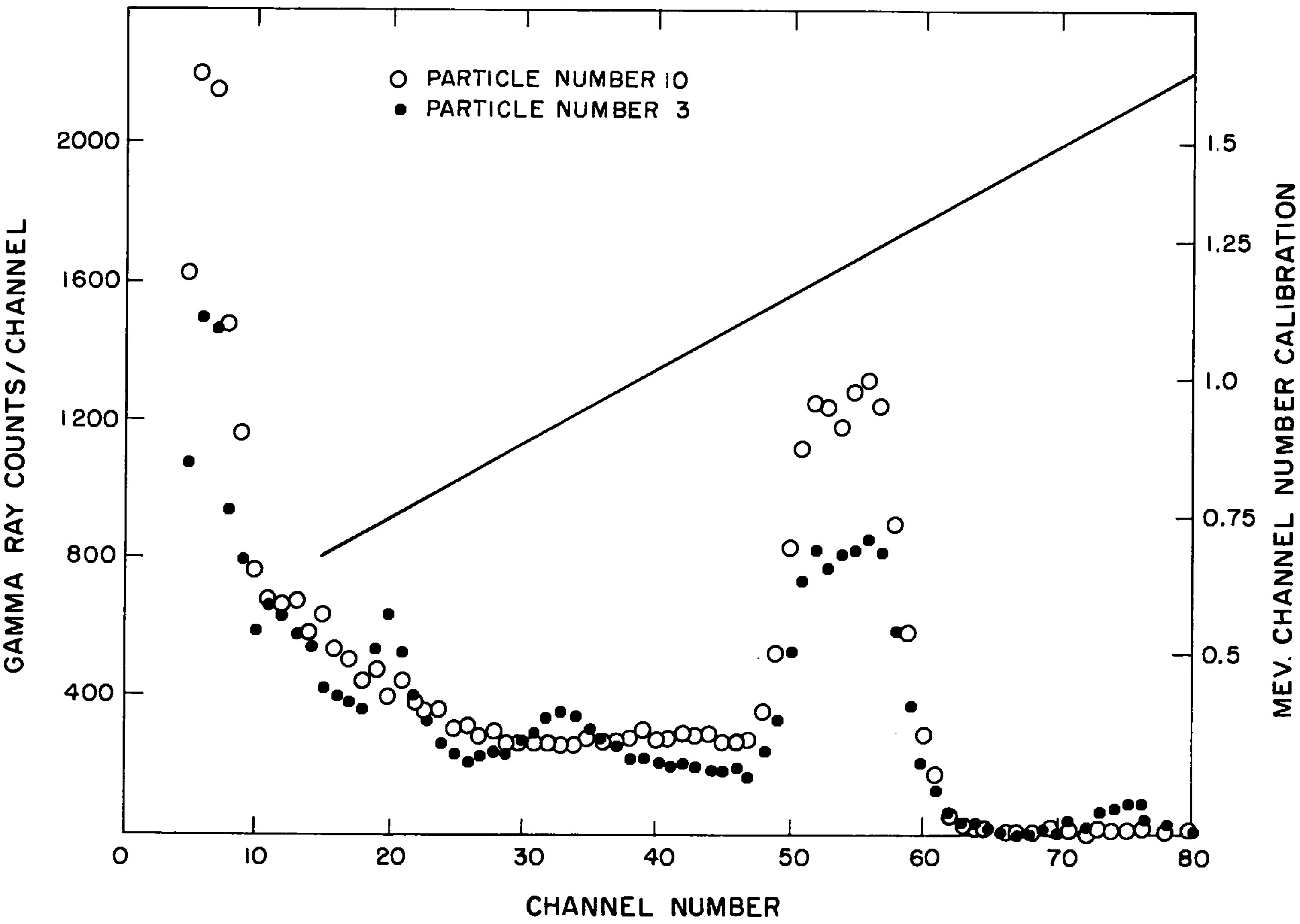


Fig. 3. Gamma Ray Spectrum

[redacted]

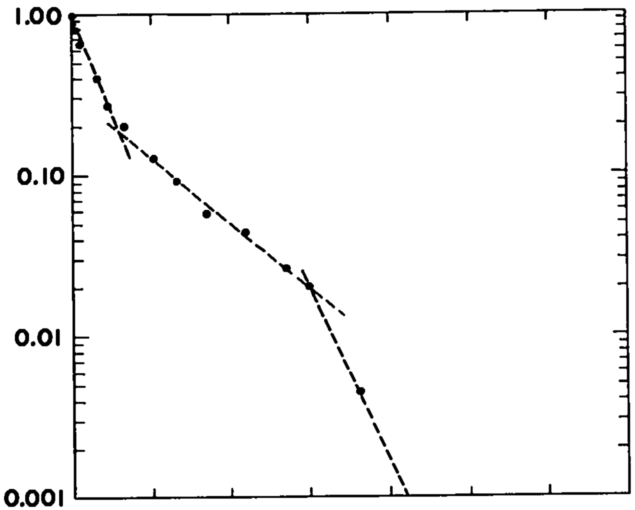


Fig. 4. Beta Absorption Curve P-1

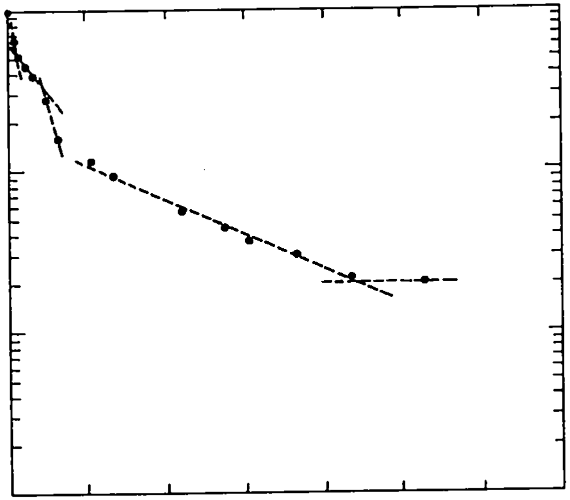


Fig. 5. Beta Absorption Curve P-2

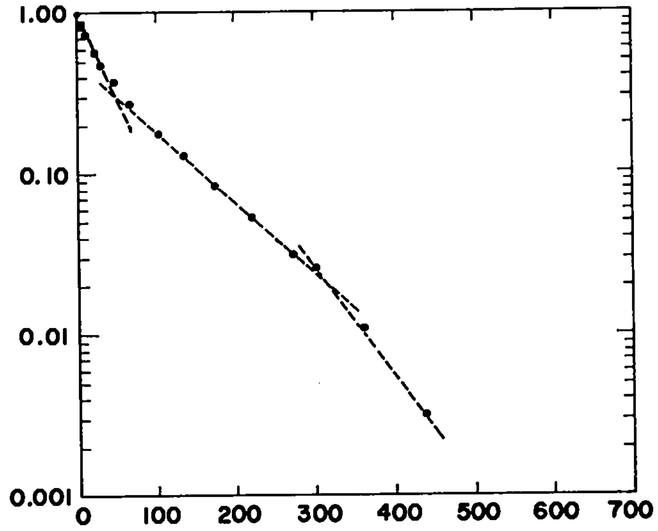


Fig. 6. Beta Absorption Curve P-3

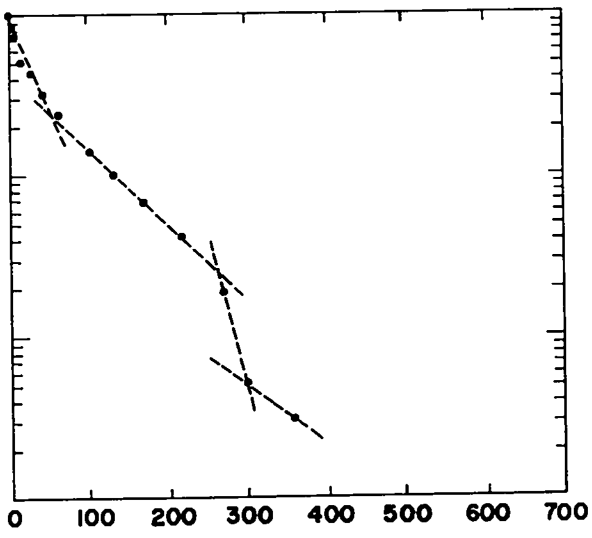


Fig. 7. Beta Absorption Curve P-4

[redacted]

[redacted]

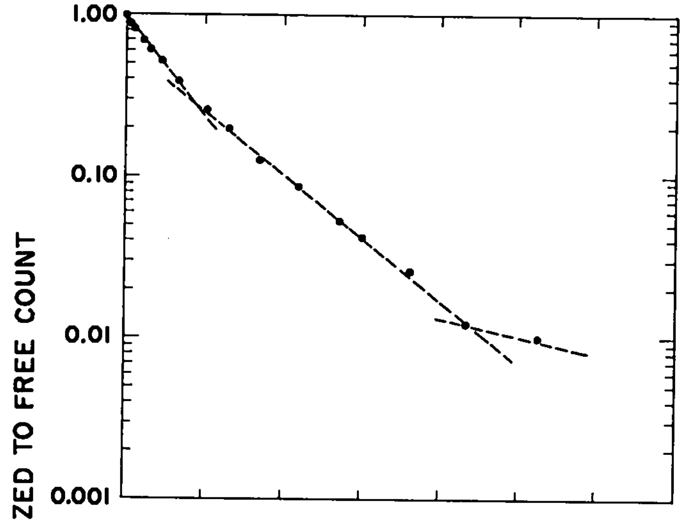


Fig. 8. Beta Absorption Curve P-5

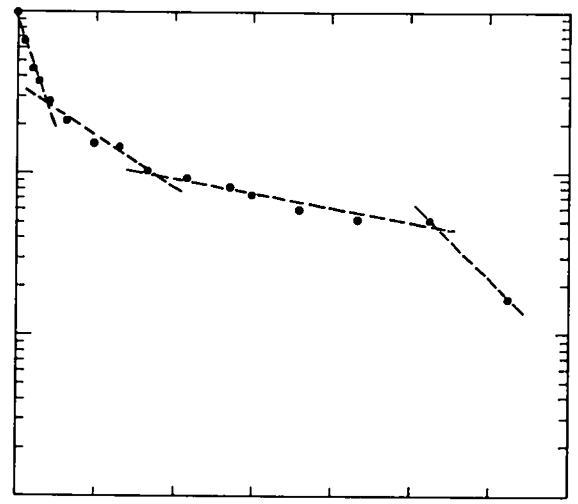


Fig. 9. Beta Absorption Curve P-8

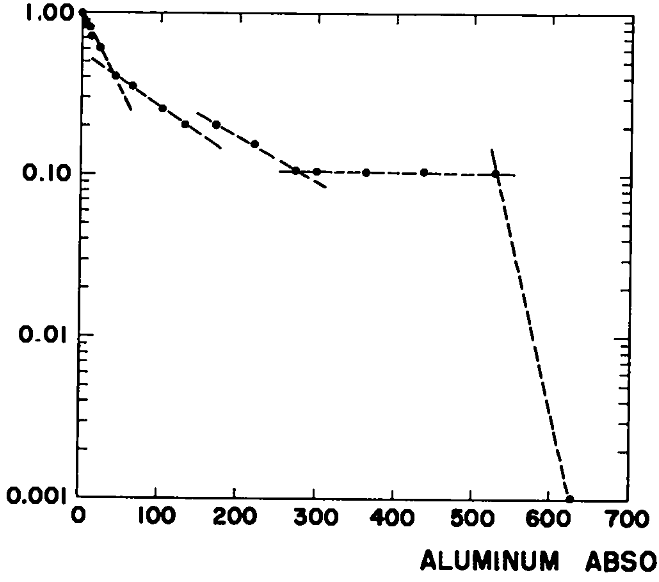


Fig. 10. Beta Absorption Curve P-9

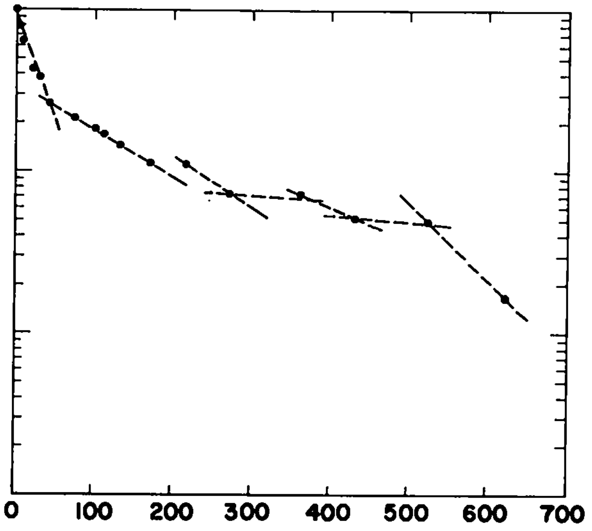


Fig. 11. Beta Absorption Curve P-10

[redacted]

[redacted]

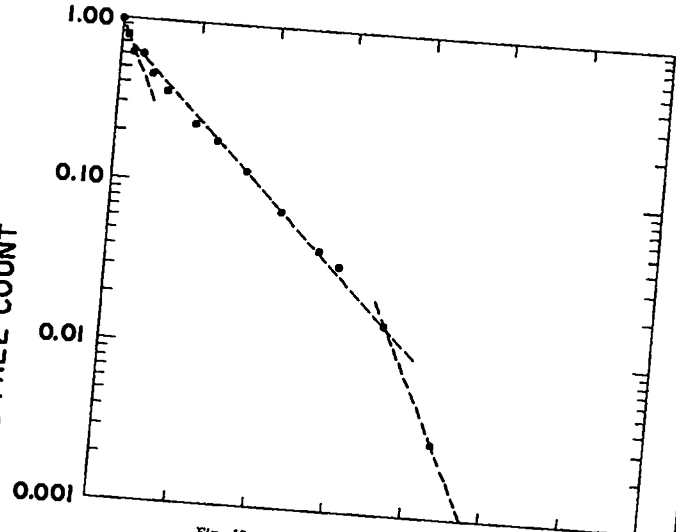


Fig. 12. Beta Absorption Curve P-11

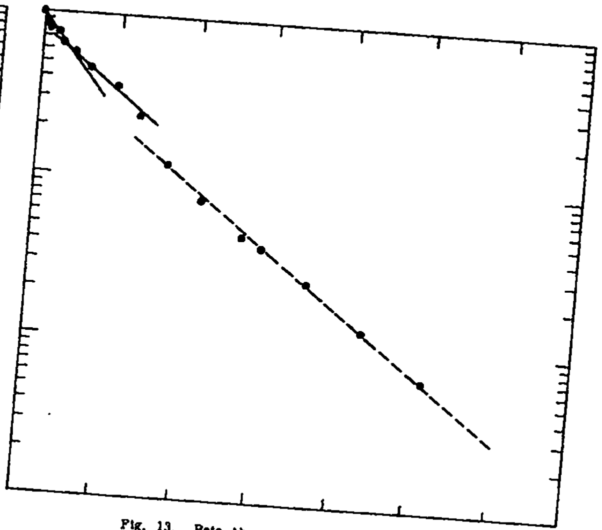
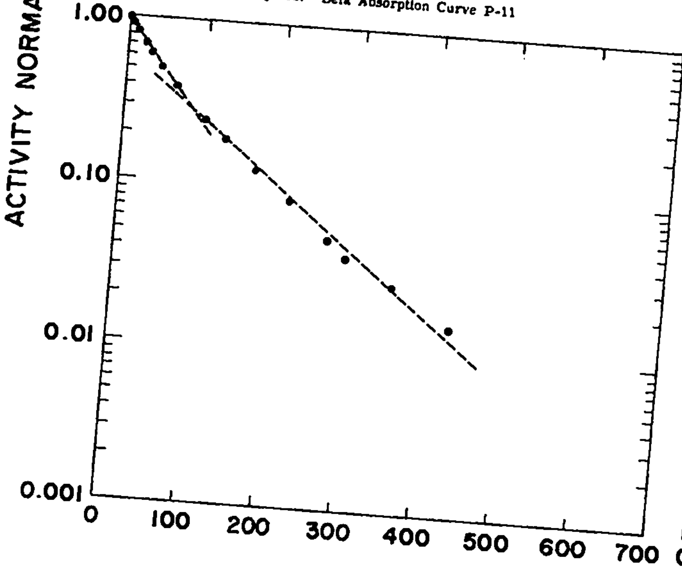
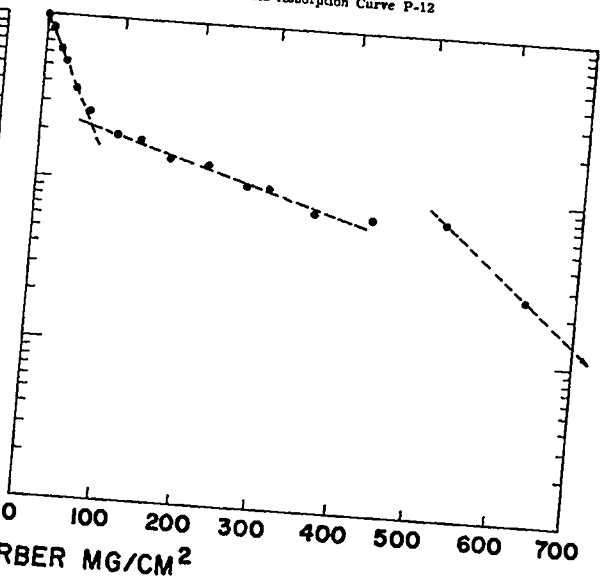


Fig. 13. Beta Absorption Curve P-12





ALUMINUM ABSORBER MG/CM^2

Fig. 14. Beta Absorption Curve P-13

Fig. 15. Beta Absorption Curve P-14

[redacted]

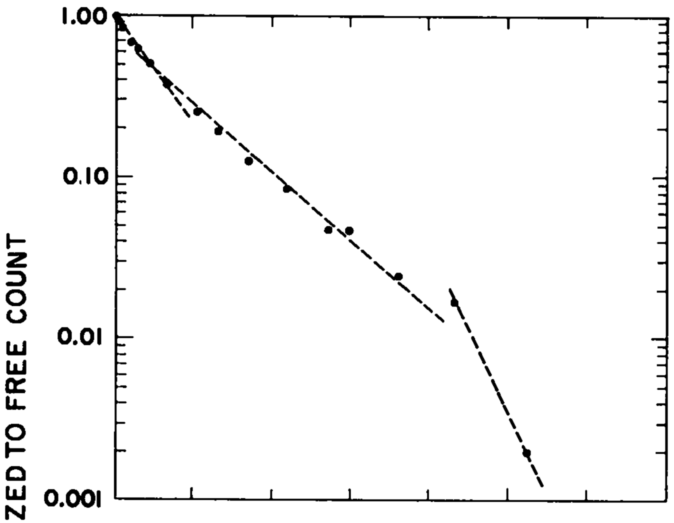


Fig. 16. Beta Absorption Curve P-15

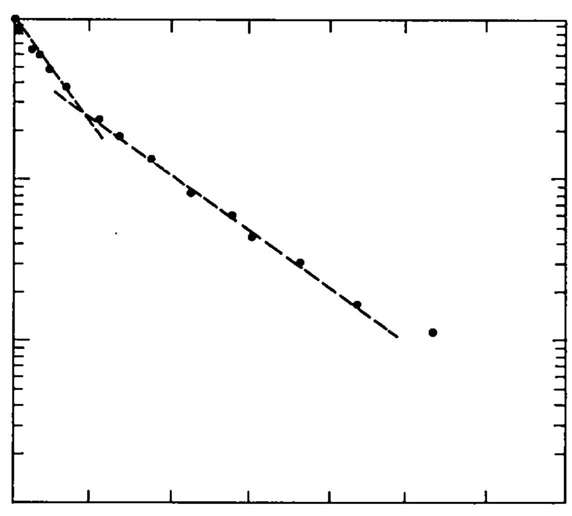


Fig. 17. Beta Absorption Curve P-17

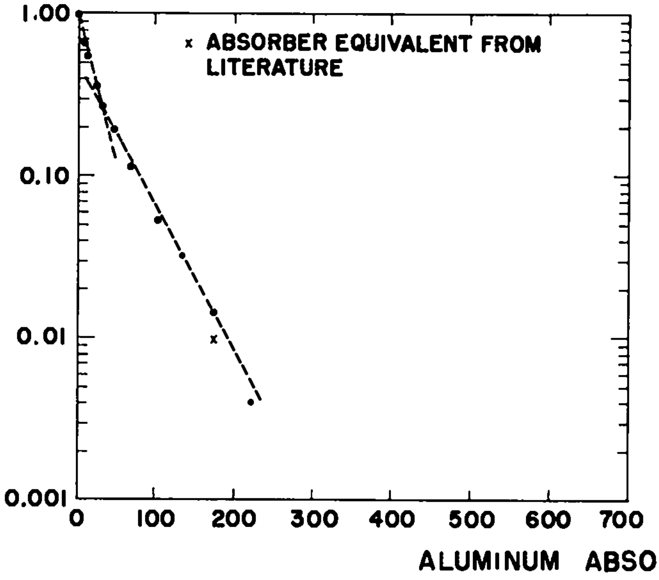


Fig. 18. Cesium-137 Control

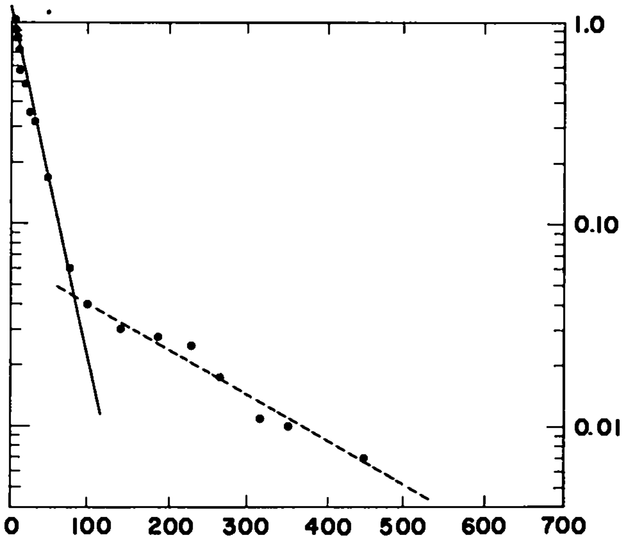


Fig. 19. Tantalum-182 Control

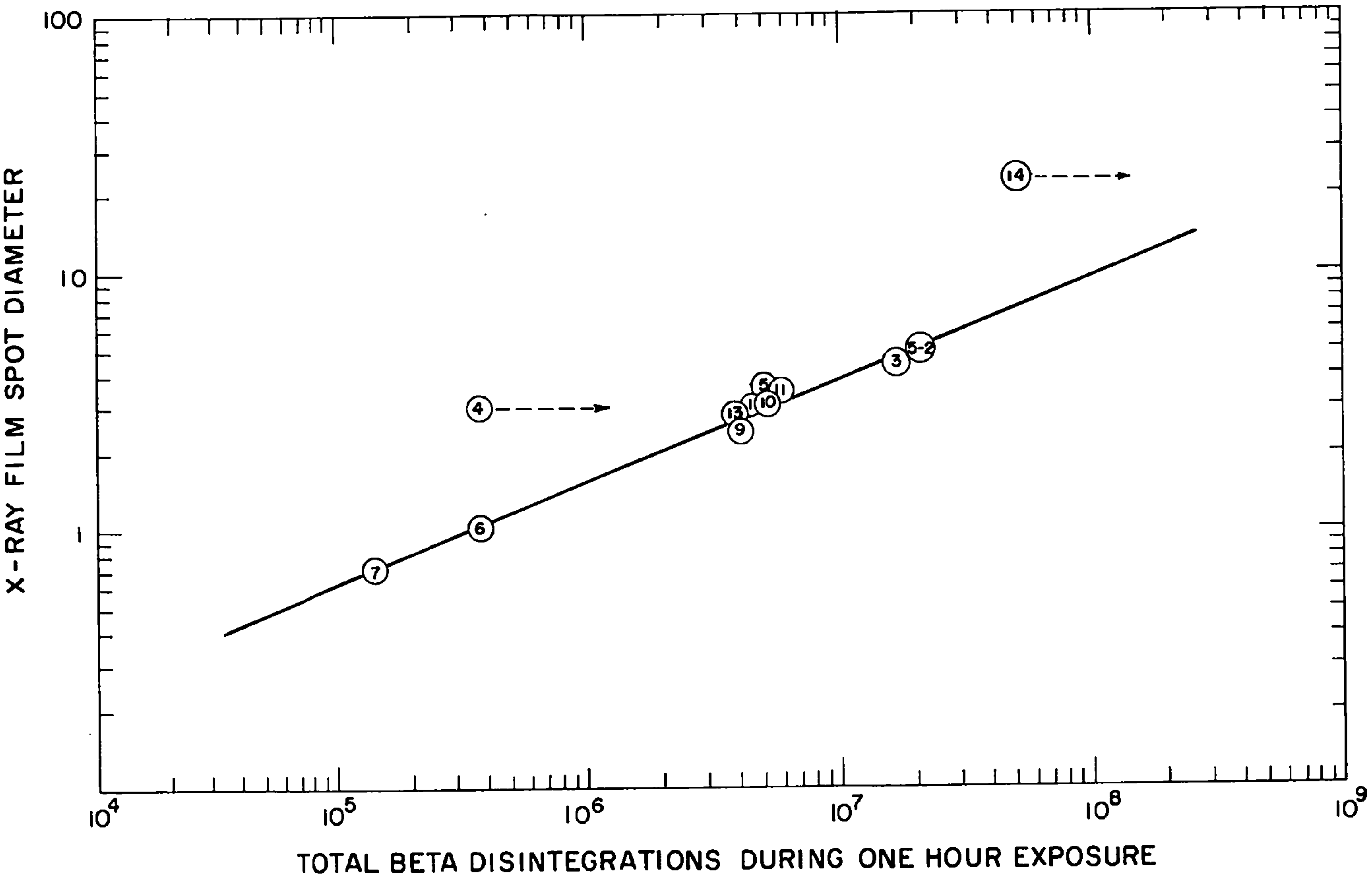


Fig. 20. X-Ray Film Radioautography (Spot Diameter vs. Activity)

[redacted] SIFIED

UNCLASSIFIED

UNCLASSIFIED

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