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FINAL REPORT OF STUDIES OF ^{238}Pu DEBRIS PARTICLES FROM THE SNAP-9A SATELLITE FAILURE OF 1964

Contract No. AT(30-1) 3803

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For

Health and Safety Laboratory
United States Atomic Energy Commission
New York Operations Office

Tracerlab / WEST
A DIVISION OF LABORATORY FOR ELECTRONICS, INC.
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DISCLAIMER

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Health and Safety Laboratory
Environmental Studies Division
New York Operations Office
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376 Hudson Street
New York, New York 10014

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ABSTRACT

A study was conducted on ablated ^{238}Pu particles from the SNAP-9A satellite failure of April, 1964, to provide data for the evaluation of the satellite re-entry burnup hazard. During this study, two new techniques were developed: the thick-nuclear emulsion method for viewing the alpha tracks produced by alpha-emitting particles in LTA-ashed IPC paper; and the photoreversal technique for electron microscopy of submicron radioactive particles.

The size distribution of the ^{238}Pu particles after a one-month exposure in nuclear emulsion appears lognormal with a mode at ~13 mμ (particle diameter) and a size range of ~ 10 to 40 mμ. Scanning the sample slides after a one-year exposure confirmed the presence of many ^{238}Pu particles less than 10 mμ in diameter which were not seen in the one-month exposures. The contribution of Pu particles in the two- to five-track star region of the one-year-exposed samples is still unknown.

Scanning slides with ^{238}Pu particles covered by nuclear emulsion also revealed that partial leaching (or dissolving) of these particles was occurring during the exposure period. A study of this effect indicated that as the equivalent $^{238}PuO_2$ particle diameter decreases, the percentage of the original particle leached increases.

Since photoreversed particles were remaining on the microscope slides during float-off, three more efficient techniques for preparing these particles for electron microscopy were developed. When a suspension of ashed IPC, including ^{238}Pu particles in PVC (1%) was pipetted onto blank microscope slides; 65% of the particles remained with the PVC film segments during float-off. No calculations were made to determine possible loss to individual particles. Very thin PVC films were successfully fabricated; particles could then be sandwiched between two thin films to ensure complete encapsulation. The scanning electron microscope was used to view photoreversed weapon debris particles as small as 1 μ in diameter directly on the processed slides.

All remaining portions of IPC were ashed; the ashed residue was covered with collodion (4%) and exposed to the scintillation-screen/x-ray film arrangement for 12 days. Two SNAP-9A particles were found, both with equivalent $^{238}PuO_2$ diameters greater than 50 mμ. An experiment with control slides to evaluate the scintillation-screen/thin-film nuclear emulsion arrangement in photoreversal studies showed no photoreversed areas indicating stars with more than six tracks. A

second experiment with SNAP-9A particles was inconclusive because portions of the scintillation screen separated from the thin-film nuclear emulsion during exposure.

Silicone oil on impactor slides was successfully diluted by a series of centrifugation-dilution steps. Electron microscopy of an aliquot from the fourth dilution showed no interference from silicone oil.

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TABLES

FIGURES

1.0 INTRODUCTION

In April, 1964, a SNAP-9A satellite containing ~ 17 kilocuries of ^{238}Pu with a mass of ~ one kilogram failed to attain orbit and re-entered the atmosphere over the Indian Ocean.[1] Before August, 1964, attempts to collect the SNAP-9A debris on aircraft samplers at 70,000 feet and on balloon samplers at ~ 100,000 feet were unsuccessful. Samples collected by balloon at 105,000 feet 34° S during August, 1964, and analyzed radiochemically contained amounts of ^{238}Pu significantly above the usual levels.[2] However, radiochemical analysis yields no data on the size distribution of the particles or on their individual morphology and composition.

These early collections were made at extreme altitudes and probably represent the debris material in its initial condensed form. It was expected that, as the ^{238}Pu-containing particles entered the troposphere, which contains a relatively high concentration of background particulate material, the ^{238}Pu would agglomerate with these larger airborne particles prior to sedimentation or washout by rain.[3]

In order to understand the nature of the debris in its primary form and to study both the eventual fallout to earth, resulting from possible accretion by larger particles, and the consequent implications to the world-wide fallout hazard, the Health and Safety Laboratory (HASL) of the Atomic Energy Commission contracted with Tracerlab to characterize these ^{238}Pu particles (Part 2.0 of this report) and the changes they undergo as they descend through the atmosphere (Part 3.0).

2.0 SIZE-DISTRIBUTION STUDIES

2.1 SAMPLE DESCRIPTION

Analyses were performed on filter papers from high-altitude samplers with ^{238}Pu from the SNAP-9A abort and filter papers exposed before this event.

Sample slides are prepared by adding liquid nuclear emulsion to microscope slides holding ashed portions of flown IPC filter paper known by radiochemical analysis to have ^{238}Pu activity above background. The slides prepared by adding liquid nuclear emulsion to clean microscope slides are called blank slides. Slides with LTA-ashed (see Appendix Section A-1), blank IPC (4 cm^2) covered with nuclear emulsion are designated as blank IPC slides. Flown control slides are prepared by adding liquid nuclear emulsion to microscope slides containing ashed portions of flown IPC filter paper known to have no ^{238}Pu activity above background. Blank, blank IPC, and flown control slides are collectively called background slides.

Table I lists the filters received from the Health and Safety Laboratory for analysis, together with pertinent collection data. A ratio of ^{238}Pu particles to ^{239}Pu particles greater than 0.05 is considered a significant indication of SNAP-9A abort particles. Table II lists the techniques used to analyze each sample.

2.2 PRELIMINARY INVESTIGATION

Filter papers from high-altitude samplers with ^{238}Pu particles from the SNAP-9A abort and filter papers exposed before this event were divided into 4-cm^2 pieces, ashed on microscope slides in a Tracerlab LTA-600 Low-Temperature Asher, and alpha-counted in a methane end-window proportional gas-flow counter* to determine if the ^{238}Pu activity was concentrated in a few or in many particles. A wide difference in count from slide to slide would indicate that the alpha activity is concentrated in a few particles. If the ^{238}Pu activity were concentrated in many particles, all 4-cm^2 pieces from the same filter paper would give approximately the same counts.

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*The Tracerlab methane end-window proportional gas-flow counter, normally a beta counter, was used for alpha-counting after the proper voltage adjustments and corrections for loss of efficiency had been made. The alpha counts on these slides could not be determined in a standard alpha-counter because the static on the glass slides caused erratic readings.

Table I. Flight Collection and Radiochemical Data of Filters

HASL No.	Collection Date	Latitude	Altitude (10^3 ft.)	^{238}Pu dpm in Quadrant	^{239}Pu dpm in Quadrant	$\frac{^{238}Pu}{^{239}Pu}$
A-1404C	11/12/64	34°S	106	4.5	0.08	56.2
A-1498D	1/27/65	34°S	89	3.0	0.3	10.0
A-1155C	5/20/64	34°S	88	0.03	3.8	0.08
A-1305H*	8/12/64	32°N	65	3.8	70	0.05
A-1436	(Blank IPC)					
A-1499C	1/27/65	34°S	89	3.0	0.3	10.0
A-1501E	1/28/65	34°S	93	2.8	0.12	23.3
A-1583C	2/25/65	34°S	96	2.5	0.63	3.69
A-1613E	3/11/65	34°S	93	5.2	0.09	57.7
A-1709E	4/8/65	34°S	82.2	7.1	0.1	71.0
A-1809	5/19/65	31°N	94.2	0.81	0.068	11.90
A-1839	6/4/65	65°N	93	1.21	0.082	14.75
A-1916	7/17/65	65°N	81.3	0.90	0.14	6.43
A-1955	8/17/65	65°N	105	3.36	0.13	25.8
A-2006	9/28/65	9°N	80	0.19	5.6	0.04

*This collection was made by aircraft, while other collections were by balloon.

Table II. Techniques Used to Analyze the Various Types of Samples Received for ^{238}Pu Particle Analysis

Samples Examined	Techniques			
	Thick-Nuclear Emulsion*	Hollow Star*	Photoreversal*	Additional Radiochemical Analysis**
1. Filters (IPC) HASL No.				
A-1404C	X			
A-1498D	X			
A-1155C	X			
A-1305H	X			
A-1436	X			
A-1499C	X			
A-1501E	X			X
A-1583C	X			X
A-1613E	X	X	X	X
A-1709E	X	X	X	
A-1809	X			
A-1839	X			
A-1916	X			
A-1955	X			
A-2006	X			
2. Impactor Probe		X		
3. Silicone Oil Impactors		X		
4. Litton Grids		X		

*These three techniques are described in the Appendix.

**The ^{238}Pu activities obtained by performing radiochemical analyses on several sample slides with ashed segments from these three filters were compared to corresponding ^{238}Pu activities obtained by scanning these same sample slides.

The results indicated no unusual concentrations of activity on any of the slides. For the sample with the highest count, it was calculated that, if this count were due to only one particle, this particle could not have a disintegration rate greater than 0.6 dpm (within a 95% confidence interval of ± 0.6 dpm). Since an activity of 1.2 dpm is the alpha activity of a 0.18 μ -diameter $^{238}PuO_2$ particle, this indicated that the ablated SNAP-9A particles were less than 0.18μ in diameter (assuming $^{238}PuO_2$ spheres). Therefore, to obtain size distributions of the ^{238}Pu particles, it was decided to expose these and future slides with ashed debris from the SNAP-9A abort to a thick-nuclear emulsion, type NTA (manufactured by Kodak), since this is a more sensitive technique for measuring radioactive aerosol particles (see Appendix Section A-2).

2.3 SIZE DISTRIBUTIONS

Of prime importance was the quantitative determination of ^{238}Pu content by counting the individual alpha tracks emanating from the debris after a known exposure period. On sample slides, single alpha tracks were observed as well as alpha stars consisting of a few tracks to many hundreds of tracks (see Appendix Section A-3). Exposure of blank nuclear-emulsion slides and slides with ashed blank IPC covered by nuclear emulsion revealed only stars with one to five tracks. Therefore, stars with six or more tracks were assumed to be caused by SNAP-9A debris. Errors in the size distribution for these stars were statistical errors relating to the number of stars observed and possible errors in the determination of ^{238}Pu content (see Appendix Section A-4).

Typical results of the one-month exposures are shown by the numerical size distribution of the ^{238}Pu particles from Filter A-1916, shown in Figure 1. The smooth curve shown with this histogram was prepared by graphical differentation (see Appendix Section A-5). As Figure 1 illustrates, the equivalent $^{238}PuO_2$ diameters range from 10 to 40 mμ with an arithmetic mean diameter of 15.4 mμ. From the cumulative log probability plot shown in Figure 2, it appears that the distribution is lognormal. The geometric mean diameter is 13.5 mμ and the geometric standard deviation is 1.4.

In conjunction with this phase of the SNAP-9A abort particle analysis program, statistical tests were conducted to determine the significance of the one- to five-track stars on the sample slides. In these "Z-Tests" (see Appendix Section A-6), sample slide values were compared with corresponding values from blank and blank IPC slides. Although the results did not permit a quantitative evaluation, a small contribution of activity by ^{238}Pu particles in the one- to five-alpha-track-star region was inferred from the predominance of significant differences between sample and background slides.

Several sample slides with ^{238}Pu particles from Filters A-1404C, A-1498D, and A-1499C and several background slides with portions of Filters A-1155C, A-1305H, and A-1436 were prepared and exposed to thick-nuclear emulsion for one year to determine the number of SNAP-9A particles with equivalent $^{238}PuO_2$

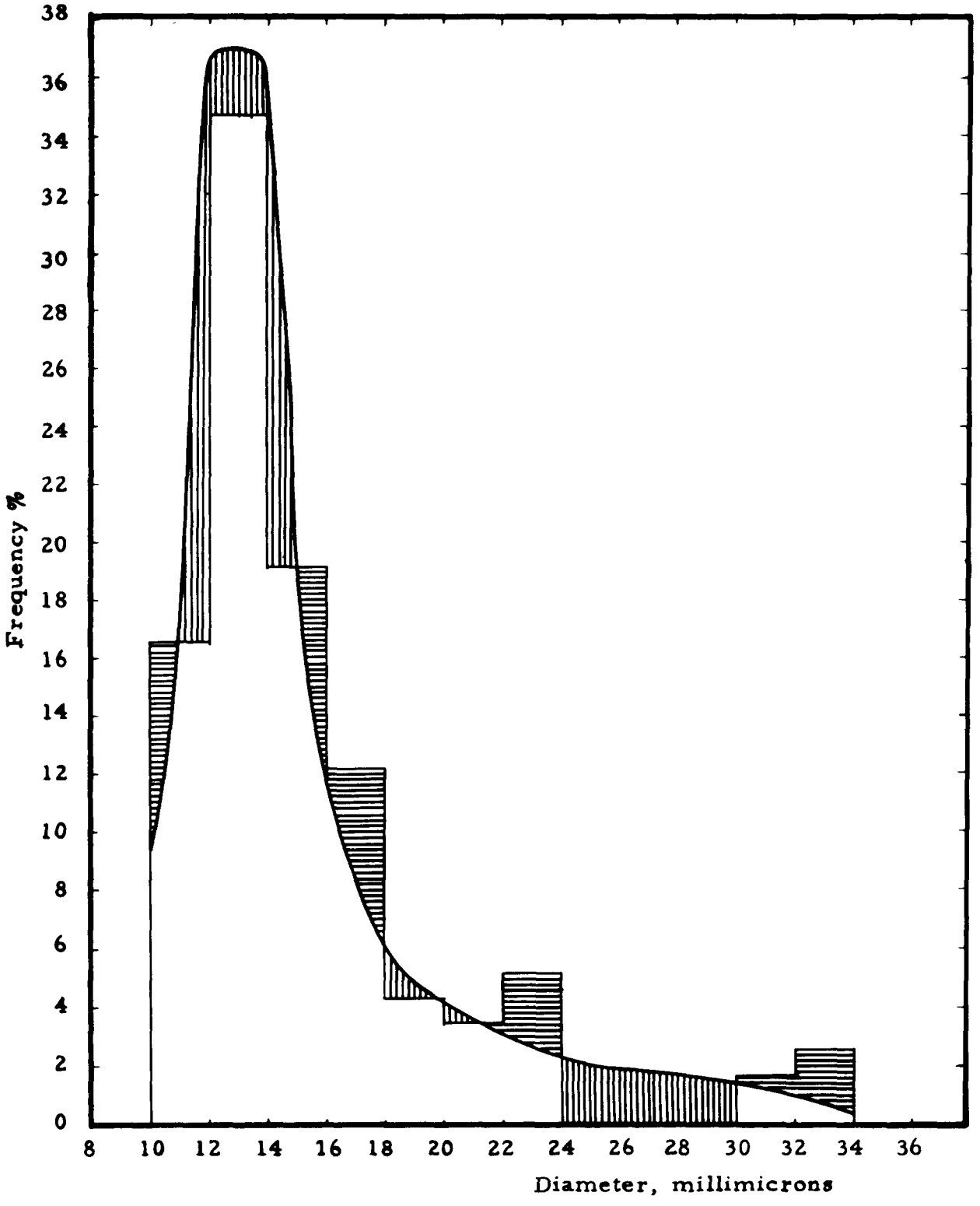
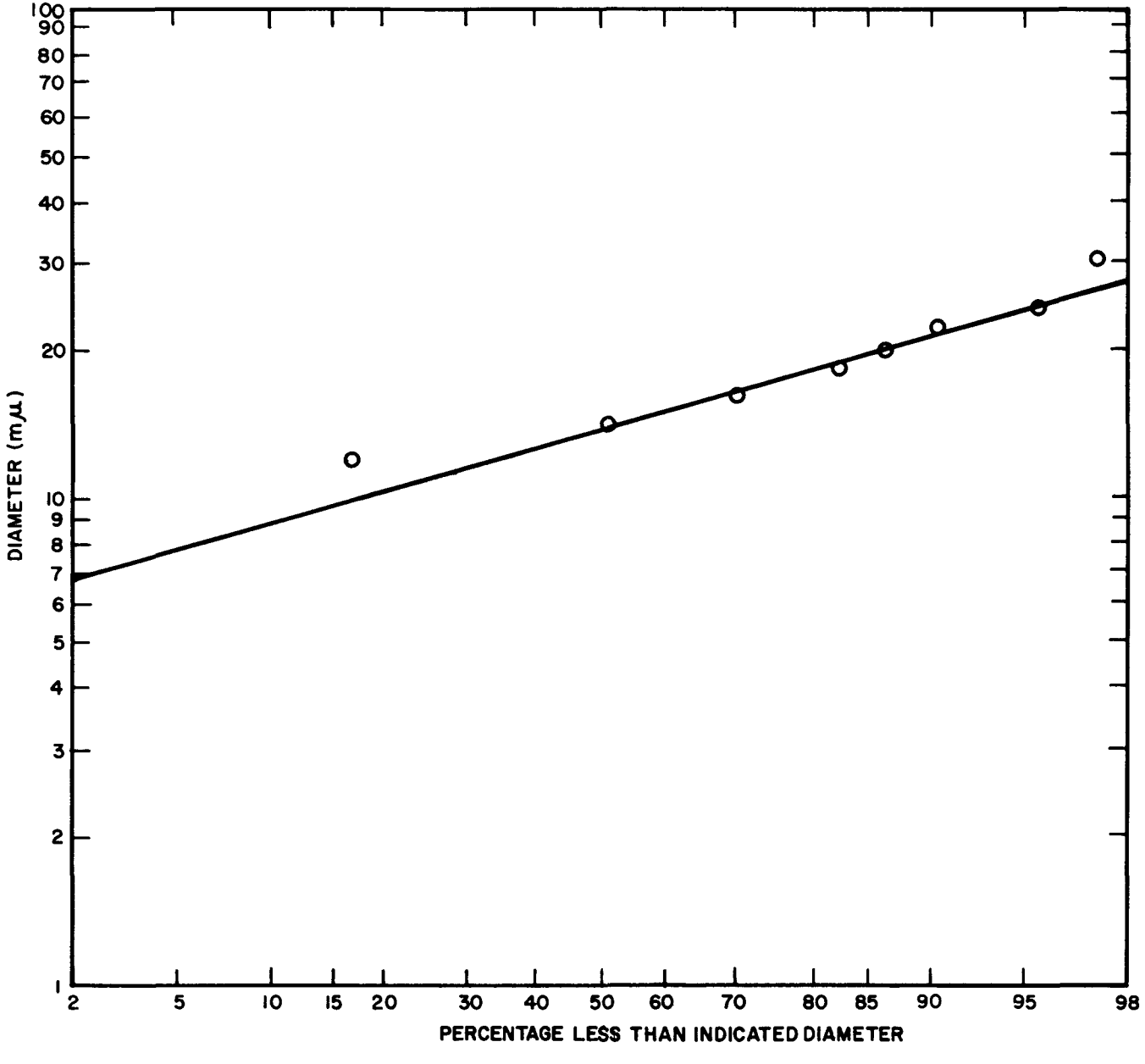


Figure 1. Numerical size distribution of SNAP-9A debris particles from Filter A-1916, based on alpha stars of six or more tracks. A total of 115 such particles was found. The arithmetic mean diameter is 15.4 mμ. The mode lies between 12 and 14 mμ.

Figure 2.
Cumulative distribution curve of Figure 1 results as a logarithmic probability plot. The geometric mean diameter is 13.5 millimicrons and the geometric standard deviation is 1.4.



diameters smaller than 10 mμ which are masked by the one- to five-track background stars after the one-month exposure.

The results of scanning the background slides exposed to thick-nuclear emulsion for one year (Table III) were significant in that only two stars with more than five tracks were found, indicating that the stars of six or more tracks on the one-year sample slides were indeed due to ^{238}Pu. A significant number of stars with more than five tracks on the one-year flown control slides would have suggested the presence of radioactive species other than ^{238}Pu.

The results of optical microscopy of the one-year exposures, represented in Figure 3, confirmed the presence of many ^{238}Pu particles less than 10 mμ in diameter. This one year histogram also revealed two points of particular interest.

The first is the peak in the 11.8 to 14 mμ-diameter range, which corresponds to the 12- to 14-mμ peaks often observed in size-distribution plots of data from one-month sample exposures. However, possibly contributing to the formation of this peak is the inaccuracy of track-counting in stars with more than 90 tracks. This error, which is particularly significant for the 150-, 200-, 300-, etc., track stars because of their extreme grouping (Figure 3) is due to the difficulty of counting the tracks in such stars and the tendency to round off track counts to multiples of 50.

To estimate the magnitude of this track-counting error, the size-distribution histogram of Figure 3 was compared with one obtained by plotting the results from scanning slides of the same filters exposed for 61 days. After normalizing the results of the 61-day exposure that corresponded to the 9.2- to 18.0-mμ portion of the one-year histogram, the results were plotted in Figure 3 as the dotted-line curve. The peak in the 12- to 18-mμ region based on the 61-day data is smaller than the peak obtained from the one-year results.

The second point of interest occurs in a region (4.9 to 10.2 mμ) of the histogram which is not affected by the track-counting error described above. To determine if the solid curve in Figure 3 continues to rise or if it falls as it passes 4.9 mμ toward the smaller particle sizes, an effort was made to estimate the number of particles in the next smaller size range (two to five tracks). It was computed that, if the curve were to continue upward smoothly, there would be 200 SNAP-9A particles in the two- to five-track range. Such a large number (641) of four- and five-track stars produced by background sources was counted that it was impossible to determine the actual number of SNAP-9A particles producing two- to five-track stars; hence, it was impossible to determine whether the curve rises or falls.

To permit a different interpretation of the one-year data, corresponding mass values were calculated and plotted (Figures 4 and 5). Figure 4 relates particle diameters to the percentage-less-than-total mass and number. Figure 5 illustrates how the mass-frequency percentage is related to particle diameter.

The mass-distribution graphs plotted from the one-year data disclose such mass and numerical information as:

Table III. Alpha Stars from Sample Slides Exposed for One Year*

Alpha Tracks per Star	Sample Designations																			
	A-1404C Slide No.				A-1498D Slide No.				A-1155C Slide No		A-1305H Slide No			A-1436 Slide No			A-1499C Slide No.			
	25	26	27	29	26	27	28	29	26	27	26	27	28	6	9	11	26	27	28	29
4	--	--	31	40	71	93	62	53	66	39	--	--	--	58	74	26	33	39	35	50
5	--	--	14	10	23	19	19	16	13	8	--	--	--	6	14	8	6	5	8	14
6	4	5	4	1	4	6		1							1	2	1	1		1
7		2	2	5	3	7	1	6									1	1		1
8	5	6	4	1	4	6	3	2											3	1
9		7	2	1	3	2	1	1					1				1	2		1
10	3	5	5	1	2	6	4	6									1	1	1	3
11	2	4	1	1	2	1														
12	2	6	4	4	4	5	4	3									1		1	4
13	2		1																	
14	1	3	1		3	2	2													
15	2	2	11	3	3	5	1	10									2		2	
16	1	3	2	2	3		3										2			
17	4	5	1		1	1	1	2												
18	4	4	4	4	4	3		2											1	1
19	4	1																		
20	1	2	6	5	7	3	4	4									2	2		4
21		3					1											1		1
22	2	1	2		2	1														
23			2		1	1	1													
24		2																		
25	4	2	13	3	11	3	4	3										4	1	1
26		1				1														
27	2		1																	
28		1	1			1														
29	1																			
30	6	5	12	2	5	5	1	7									4	2	3	
31		1																		
32		2			1	1														
33	1	3	1			1		1												1
34	1																			

*All slides were scanned for stars of 4 or more tracks except #25 and #26 of A-1404C and #26, #27, and #28 of A-1305, which were scanned for stars of 6 or more tracks.

Table III (Cont.).

Alpha Tracks per Star	Sample Designations																			
	A-1404C Slide No.				A-1498D Slide No.				A-1155C Slide No		A-1305H Slide No			A-1436 Slide No.			A-1499C Slide No.			
	25	26	27	29	26	27	28	29	26	27	26	27	28	6	9	11	26	27	28	29
35	1	1	6	1	4	1	2	5												2
36		1																1		
37	2																			
39	1																			
40	3		2	3	8	3	1	5						1			4	2		1
43	1					1														
44	1																			
45	4	3						2												1
50	8	7	17	3	14	5	7	11									5	3		1
55	1	1			1	1														
58	1																			
60	5	10	2	1	1	1	4	3									5	3		3
70	2																			
75	6	5	5	2	1	4		4										5		7
76																				
77																				
78																				
79																				
80		7	2			4	1										2		1	
85		1																		
90					1	1	1	1										1		
100	10	13	32	8	5	13	3	15									4	5	1	5
110		1																		
120		1																		1
125	2	2				1														1
150	1	6	10	1		4	6	2									3	3	2	4
175	1					1														
200	2	4	7	1		3		2										2		1
250		2			1	2													1	
300	2	3	5	2				2												1
500		3	3			1	1	1									1			1
1000	1	6			3	3		1												
2000	1																			
5000		1	1																	
10000		1	1																	1

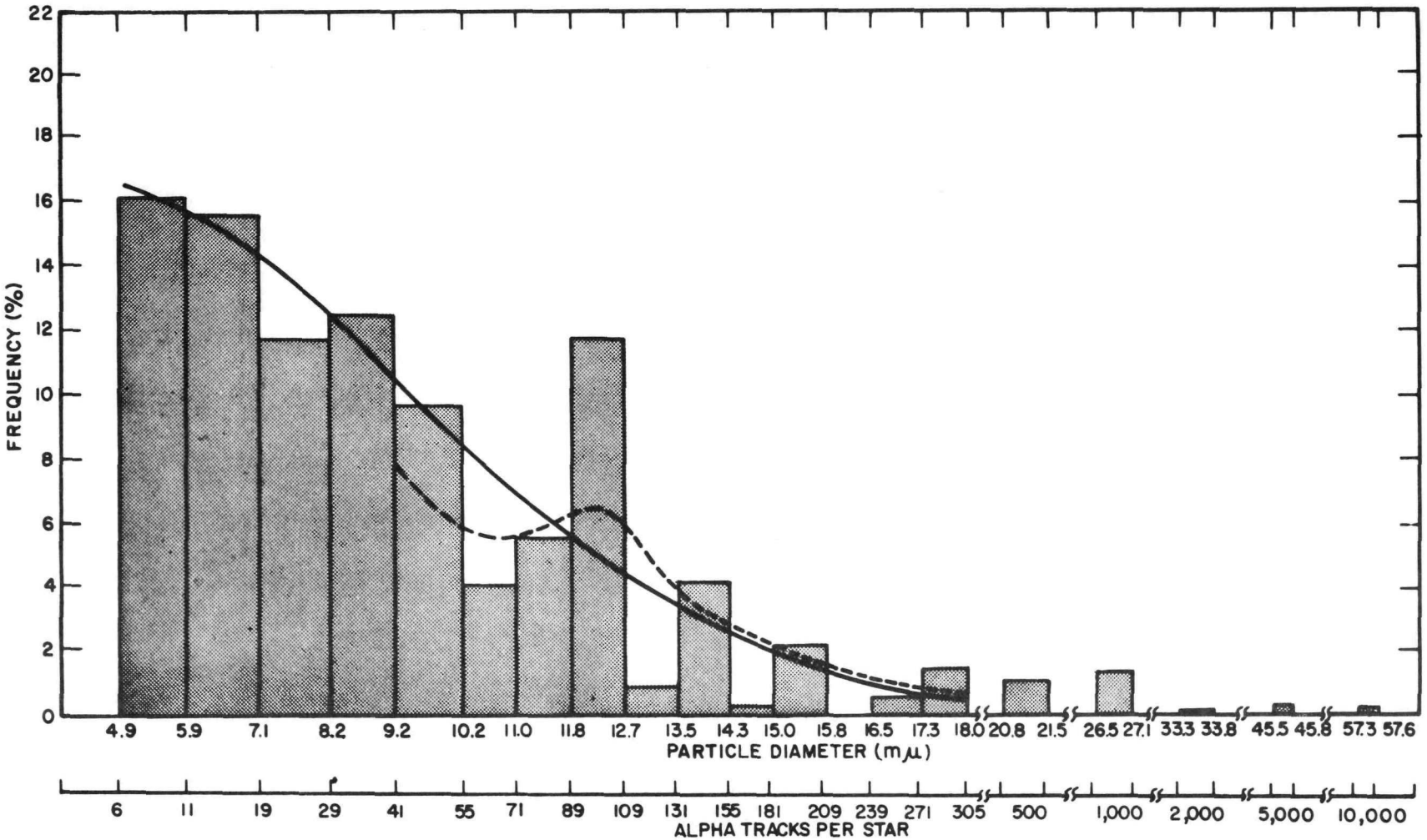


Figure 3: Numerical size distribution of SNAP-9A particles from sample slides of Filters A-1404C, A-1498D, and A-1499C, based on alpha stars of six or more tracks. The exposure time was one year. Size intervals which represent ± 1 σ Poisson errors are used. The arithmetic mean diameter is 9.7 mμ. A total of 1008 particles are plotted. The solid-line curve was obtained from this histogram by graphical differentation. The dotted-line curve was obtained from normalized results from two-month exposures of sample slides from the same filter.

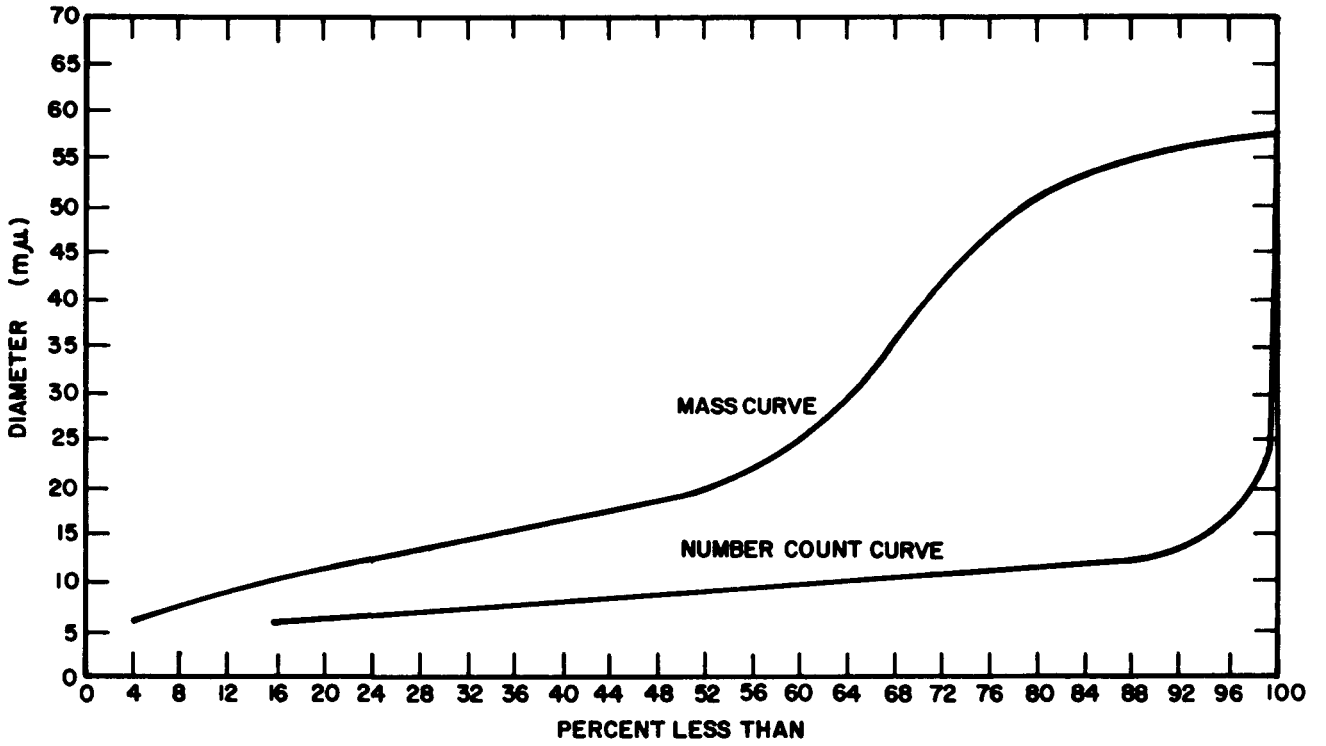


Figure 4. Plot of particle diameter versus percent less than number count and mass for one-year sample slides with SNAP-9A debris from Filters A-1404C, A-1498D, and A-1499C. The total calculated mass for this 1008-particle distribution is 1.57×10^{-13} grams.

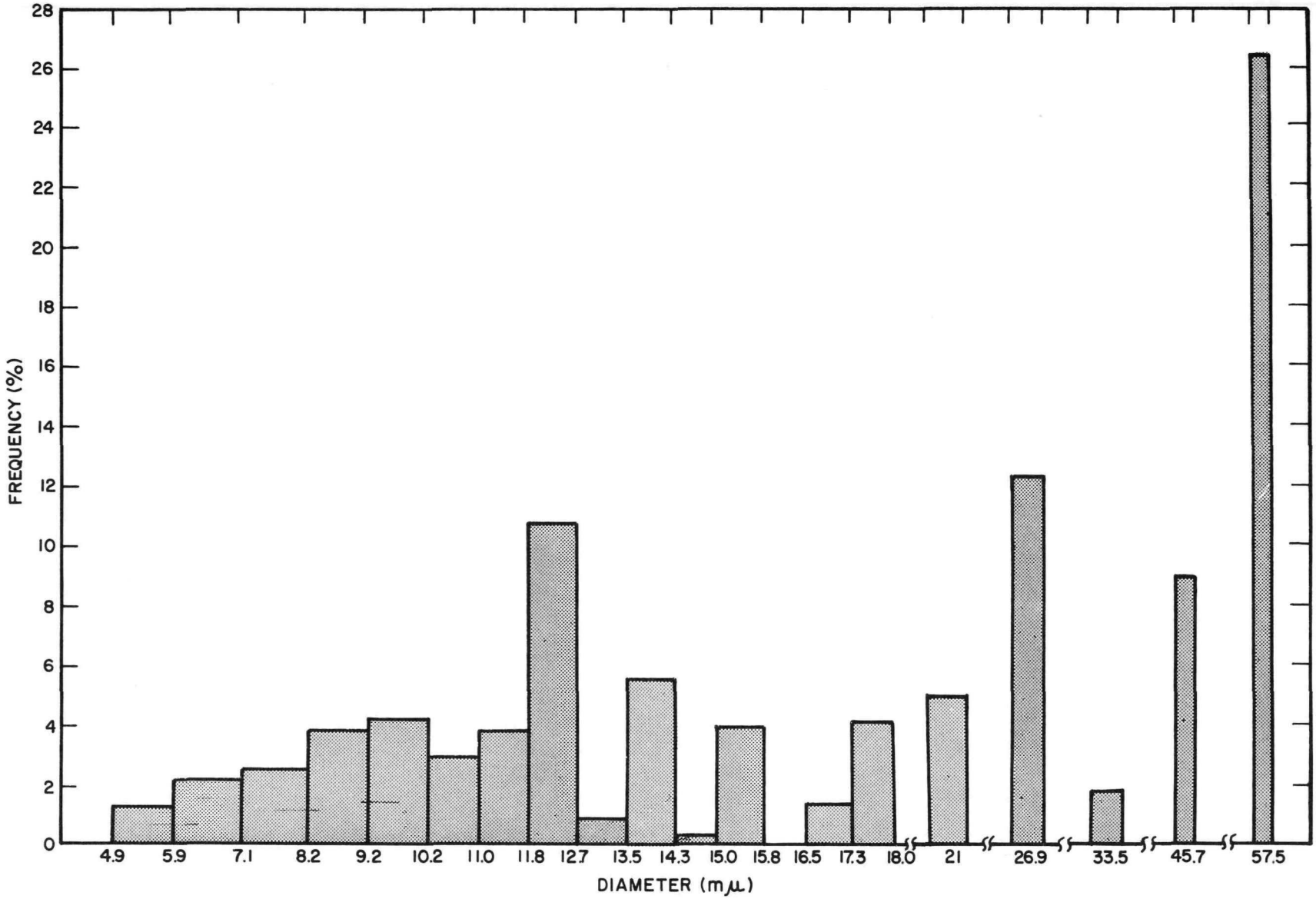


Figure 5. Mass frequency percent versus particle diameter for one-year sample slides with SNAP-9A particles from Filters A-1404C, A-1498D, and A-1499C. The total calculated mass for the 1008 particles in this distribution is 1.57×10^{-13} grams.

- Particles smaller than about 8.6 mμ in diameter compose 50% of the total number but only 11% of the total mass of the SNAP-9A particles collected, while particles greater than 8.6 mμ in diameter compose 50% of the number and 89% of the mass.

- About 65% (numerically) of the particles which compose 13.6% of the total mass are in the 4.9- to 10.2-mμ diameter range. This is the region of SNAP-9A particles producing one- to five-track stars which had been hidden by the background one- to five-track stars in the one-month exposure to thick-nuclear emulsion.

These statements are valid only for the samples collected and cannot be considered more than remotely related to the size distribution formed initially (before the largest particles were removed by sedimentation).

2.4 CONCLUSIONS BASED ON SIZE-DISTRIBUTION STUDIES

Data to assist in the evaluation of the health hazard associated with the SNAP-9A debris has been collected.

The thick-nuclear emulsion technique was developed to obtain equivalent $^{238}PuO_2$ spherical measurements of SNAP-9A debris particles in LTA-ashed IPC filter paper. Size distribution plots based on these measurements after nuclear-emulsion exposure for one month appear lognormal with a mode at 13 mμ (particle diameter) and a size range of 10 to 40 mμ.

Several sample and flown control slides were exposed for one year to determine the number of SNAP-9A particles with equivalent $^{238}PuO_2$ diameters smaller than 10 mμ, which is the lower limit of detection of ^{238}Pu particles exposed to thick-nuclear emulsion for one month. The results of scanning these slides after a one-year exposure confirmed the presence of many ^{238}Pu particles in the 4.9 to 10 mμ range. That the stars in this range were produced by SNAP-9A debris was established by the fact that stars with six or more tracks were found only on the slides with filters known to contain SNAP-9A debris.

The one-year mass distribution plot for the ablated ^{238}Pu particles collected in the upper atmosphere is well defined even though the actual number of SNAP-9A particles in the one- to five-track region is not known. To determine the significance of the ^{238}Pu particle contribution in the three- to five-track star region of the one-year study, several sample slides from the same filters have been prepared and are being exposed to thick-nuclear emulsion for 24 months.

3.0 WORK PERFORMED IN CONJUNCTION WITH SIZE-DISTRIBUTION STUDIES

3.1 LEACHING STUDY

The extremely small size of some of the SNAP-9A particles revealed by the one-year exposure suggested the possibility that they were artifacts produced by leaching in the nuclear emulsion rather than actual ablation products. In many instances, there were a greater number of single tracks in the vicinity of a large star than in other areas of the slides. This indicates that some ^{238}Pu may be leaching into the nuclear emulsion during the exposure period or during application of the emulsion. Consequently, a leaching study was conducted by optically scanning four of the one-year sample slides. Although this study revealed evidence of leached stars, the leaching was entirely in the form of single tracks, not six- to 55-track stars as found on the one-year sample slides.

The leaching curve in Figure 6, plotted with the data from Table IV, suggests that as the equivalent $^{238}PuO_2$ diameter decreases, the percentage of the original particle leached increases. Therefore, for any ^{238}Pu particle, the ratio of surface area to volume is the factor governing degree of leaching. This conclusion, however, conflicts with Figure 7, plotted with the data from Table V, which indicates that the numerical percentage of particles leached for each size range increases with increasing equivalent $^{238}PuO_2$ diameters. This contradiction may be partially explained as follows:

All single tracks in a number of fields of view were counted to establish an average number of single background tracks per field of view. This average (~6) was subsequently subtracted as background from the total number of single tracks surrounding leached stars, giving the total number of leached tracks associated with each star. It was observed that the number of single tracks per field of view varied from two to ten. Although this variation is not important for the larger leached stars, it is quite significant for the smaller leached ^{238}Pu particles; it is much easier to detect leaching in large stars than in small stars. Assuming that about 10 leached tracks must be produced in the field of view surrounding a particle for this effect to be recognized, 1% leaching can be detected in a particle producing 1000 tracks, but no less than 10% leaching can be detected in a particle producing 100 tracks.

3.2 PHOTOREVERSAL STUDIES (Appendix Section A-7)

The objective of photoreversal work is to locate and view submicron SNAP-9A abort particles by electron microscopy to determine if the ^{238}Pu particles are in

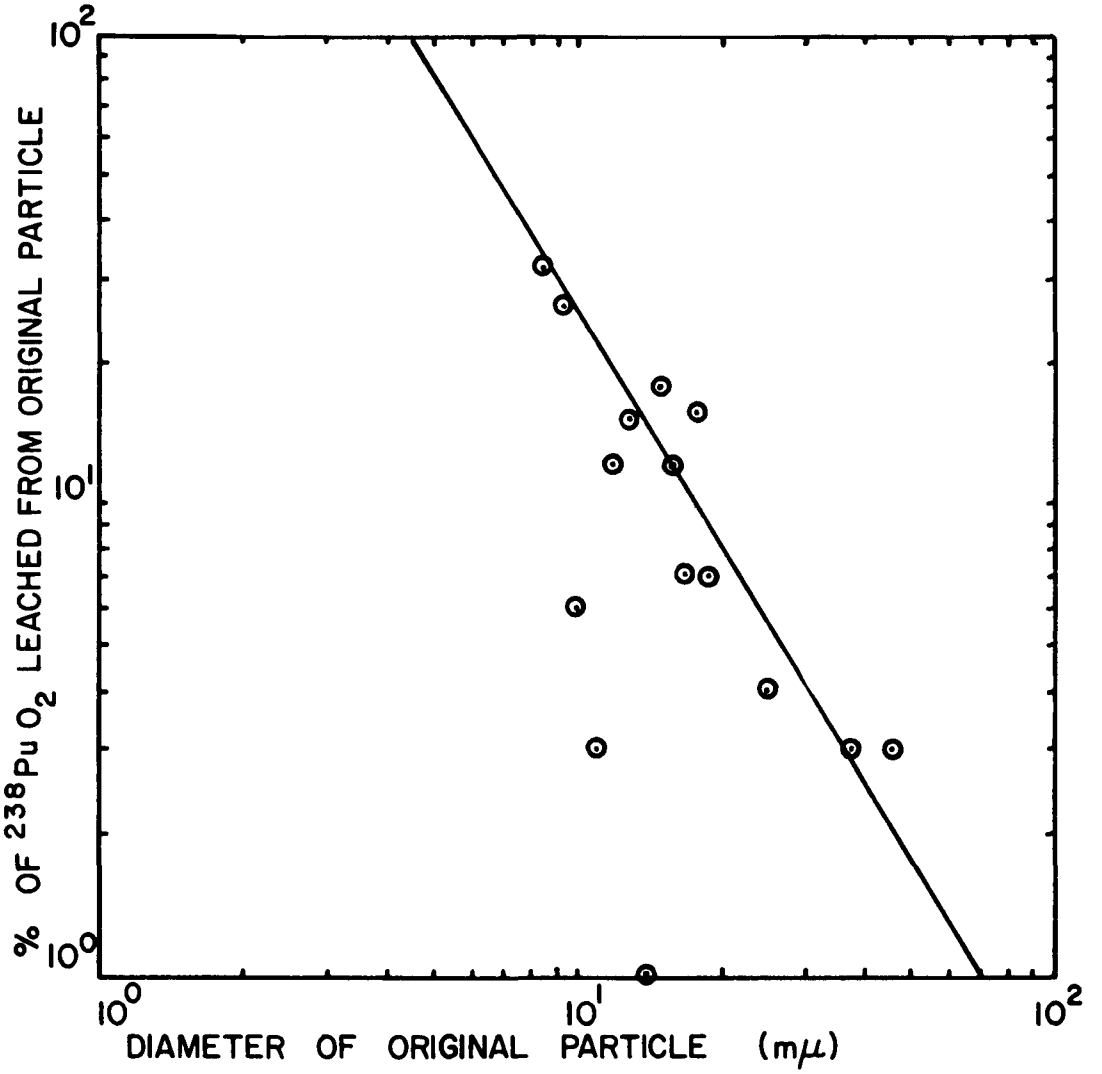


Figure 6. Plot of average mass percentage leached from $^{238}PuO_2$ particles per size range versus particle diameter.

Table IV. Average Mass Percentage Leached from ^{238}Pu Particles per Size Range

Size range of Equivalent $^{238}PuO_2$ Diameters of Unleached Particles (mμ)	Average Equivalent Mass % Leached per Size Range*	Ratio: Surface Area to Volume
8.0 - 8.9	32	1 : 1.4
9.0 - 9.9	25	1 : 1.6
10.0 - 10.9	6	1 : 1.7
11.0 - 11.9	3	1 : 1.8
12.0 - 12.9	12	1 : 2.0
13.0 - 13.9	16	1 : 2.2
14.0 - 14.9	1	1 : 2.3
15.0 - 15.9	18	1 : 2.5
16.0 - 16.9	12	1 : 2.7
17.0 - 17.9	7	1 : 2.8
18.0 - 18.9	17	1 : 3.0
19.0 - 19.9	8	1 : 3.2
22.0 - 28.0	4	1 : 4.2
39.0 - 39.9	3	1 : 2.7
46.0 - 46.9	4	1 : 7.2

*These values are obtained by averaging for each size range the values from the right column of Table V. For example, in the size range 8.0 - 8.9 mμ, the three values 29, 25, and 42, which represent the percent of equivalent $^{238}PuO_2$ in the original particles removed by leaching, are averaged to obtain 32 percent.

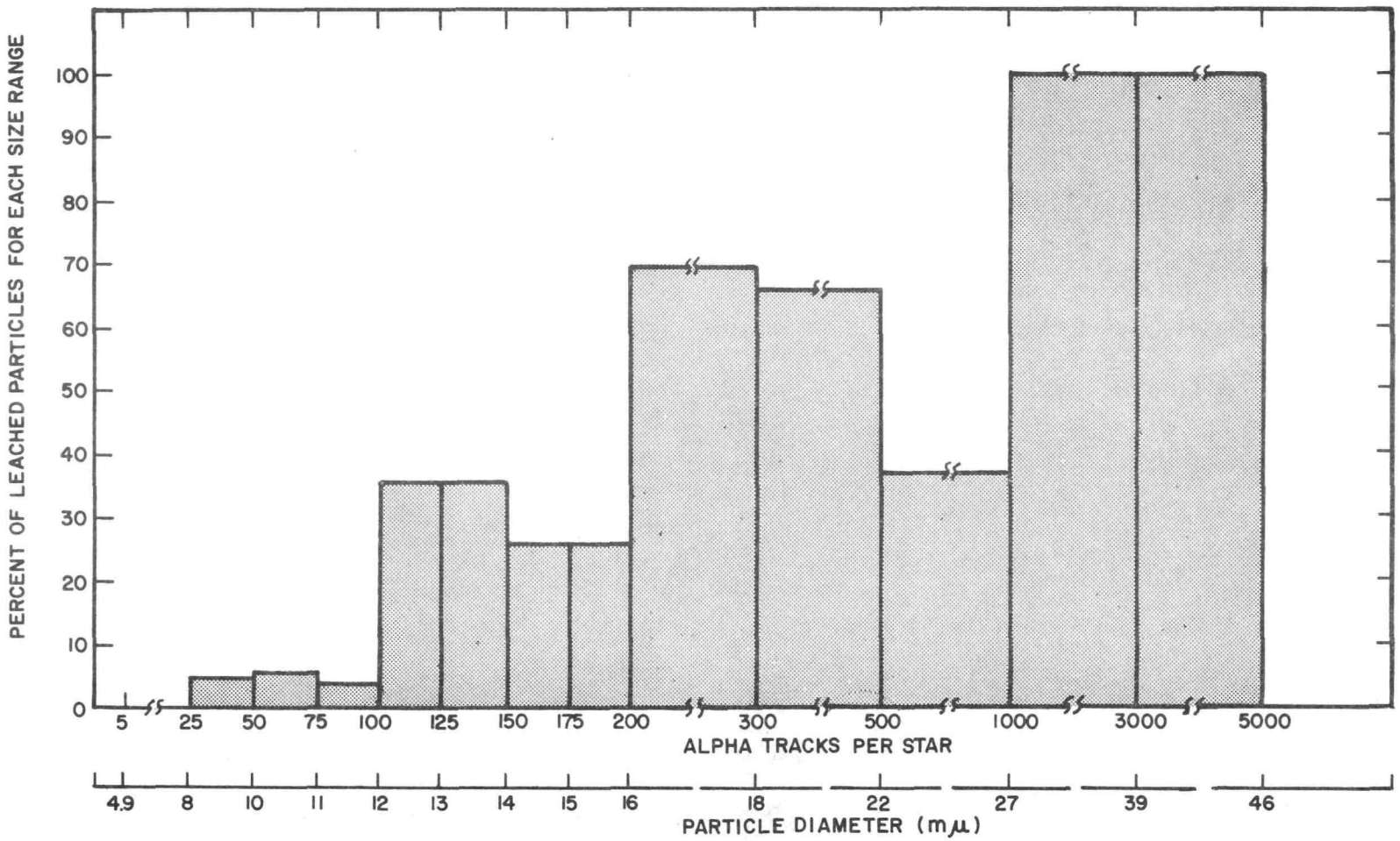


Figure 7. Histogram showing percent of leaching in each size range of SNAP-9A particles detected on sample slides exposed to thick-nuclear emulsion for one year. Of the four A-1404C sample slides which were examined, a total of 44 leached particles was obtained.

Table V. Scanning Results of Leached Stars Found on Four Sample Slides from Filter A-1404C which were Exposed for One Year in Thick-Nuclear Emulsion

Slide Designation	No. of Alpha Tracks in Leached Star (A)	No. of Alpha Tracks in Vicinity of Leached Star (B)*	No. of Alpha Tracks if Particle were Not Leached (A + B)	Equivalent $^{238}PuO_2$ Diameter of Unleached Particle (mμ)	% of $^{238}PuO_2$ in Original Particle Removed by Leaching $(\frac{B \times 100}{A})$
A-29	19	8	27	8.0	29
A-25	21	7	28	8.1	25
A-27	22	16	38	8.9	42
A-27	23	17	40	9.1	42
A-25	30	14	44	9.4	31
A-25	50	2	52	9.9	3
A-25	60	4	64	10.0	6
A-25	75	4	79	11.0	5
A-25	75	1	76	11.0	1
A-26	75	30	105	13.0	28
A-25	80	11	91	12.0	12
A-26	85	29	114	13.0	25
A-25	100	5	105	13.0	4
A-25	100	12	112	13.0	10
A-25	100	14	114	13.0	12
A-25	100	18	118	13.0	15
A-26	100	26	126	13.0	20
A-27	100	26	126	13.0	20
A-27	100	61	161	15.0	37
A-29	100	12	112	13.0	10
A-25	150	3	153	14.0	1
A-27	150	28	178	15.0	15
A-26	175	5	180	15.0	2
A-27	200	12	212	16.0	5
A-27	200	41	241	16.0	17
A-27	200	37	237	16.0	15
A-27	200	46	246	17.0	18
A-27	200	94	294	18.0	31
A-26	250	6	256	17.0	2
A-26	250	64	314	18.0	20
A-26	250	44	294	18.0	14
A-27	250	7	257	17.0	2
A-29	250	16	266	17.0	6
A-29	250	15	265	17.0	5
A-26	300	10	310	18.0	3

*The leached single tracks have been corrected for background tracks.

Table V. (Cont.)

Slide Designation	No. of Alpha Tracks in Leached Star (A)	No. of Alpha Tracks in Vicinity of Leached Star (B)*	No. of Alpha Tracks if Particle were not Leached (A + B)	Equivalent $^{238}PuO_2$ Diameter of Unleached Particle (mμ)	% of $^{238}PuO_2$ in Original Particle Removed by Leaching $(\frac{B \times 100}{A})$
A-27	300	35	335	19.0	10
A-26	350	21	371	19.0	5
A-29	500	27	527	22.0	5
A-27	800	40	840	25.0	4
A-25	1000	38	1038	27.0	3
A-26	1000	58	1058	27.0	5
A-26	3000	94	3094	39.0	3
A-27	5000	184	5184	46.0	3
A-29	5000	213	5213	46.0	4

their primary form or have become attached to larger airborne particulate material before sedimentation or washout by rain.

Such an objective was applicable in this case because the health hazards associated with the abort of a SNAP device in the upper atmosphere depend, among other factors, upon the specific activity (activity per unit mass) of the particles resulting from the event. Since agglomeration and coagulation of the SNAP debris particles may occur during their residence in the troposphere, the size distribution of the SNAP abort material in the stratosphere (as inferred from alpha tracks registered on photographic emulsions) may not be the same as the size distribution (and, hence, the specific activity) of the particles that enter the biosphere. It is, therefore, of interest to determine if agglomeration has indeed affected the size distribution.

Before processing sample slides with SNAP-9A particles for photoreversal studies, it was necessary to determine the approximate number of alpha disintegrations require by a ^{238}Pu particle to produce a (photoreversed) clear area that could be recognized by optical microscopy. After this had been established, the optimum exposure time to thin-film nuclear emulsion required for any ^{238}Pu particle could be calculated from its alpha activity as found by the hollow-star technique (see Appendix Section A-8). The photoreversal experiments required to establish the norm were performed on ablated ^{238}Pu-containing particles from Sandia.[4] These particles were selected because they have larger equivalent $^{238}PuO_2$ diameters than the SNAP-9A particles and, therefore, require shorter exposure times.

As reported previously, the first photoreversal experiments with these ^{238}Pu-containing particles from Sandia indicated that ~ 100 to 124 alpha disintegrations were required to produce a photoreversed clear area.[5] Electron microscopic examination of the clear areas, however, indicated that no ablated ^{238}Pu-containing particles were present. To confirm this, those grids which should have contained the photoreversed particles were exposed to a nuclear-emulsion plate for 12 hours. Subsequent scanning of this plate indicated that no ^{238}Pu activity was present.

The immediate conclusions were that the Sandia particles were insufficiently encapsulated in the PVC film and had remained on the sample slides or had dropped into the water during flotation. One alternate possibility was considered: in the normal stripping operations performed before electron microscopy, the sample slides are exposed to hydrofluoric acid fumes in order to loosen the PVC film from the glass slide so that the film may be floated onto the water surface. If it is assumed that the hydrofluoric fumes are reaching and reacting with the ^{238}Pu-containing particles, the lack of ^{238}Pu particles in the photoreversed clear areas may be due to exposure of the sample slides to the acid fumes. Thus, the second photoreversal experiment, as described below, was designed specifically to evaluate the effect of hydrofluoric fumes on the ^{238}Pu particles.

Ablated ^{238}Pu-containing particles from Sandia, suspended in a solution of methyl ethyl ketone, were pipetted onto six blank microscope slides and ashed in

an LTA-600 Low-Temperature Asher to remove organic residues. The six slides were then covered with thin-film PVC (1%) and exposed to nuclear-emulsion plates to determine the number of ^{238}Pu-containing particles on each slide. The equivalent $^{238}PuO_2$ diameters of these particles as calculated by the hollow-star technique ranged from 60 mμ to 1 μ. Before flotation, three of these slides were exposed to hydrfluoric acid; the other three were not.

All the PVC films containing the particles were floated off onto six "new" (blank) slides. The "old" slides (less the floated-off PVC films) were again covered with a thin film of PVC and exposed to nuclear emulsion plates. The "new" slides with their PVC films were also exposed to nuclear emulsions.

After processing, all nuclear emulsion plates were scanned for hollow stars. The scanning results indicated the following:

- All nuclear emulsion plates exposed to the slides before PVC films were removed by flotation showed hollow stars.

- Hollow stars were found on the nuclear emulsion plates which had been exposed to the three "old" slides which had their PVC films removed after exposure to hydrofluoric acid. The three nuclear emulsion plates exposed to the "old" slides that had not been bathed in acid fumes showed no hollow-star development after the particle-containing PVC films had been floated off.

- The three nuclear emulsion plates exposed to the "new" slides with the PVC films that had been bathed in acid fumes showed no hollow stars. The three nuclear emulsion plates exposed to the "new" slides with the PVC films that had not been bathed in acid fumes showed the same number of hollow stars that had been observed originally, before the particle-containing PVC films were floated off.

The particles in the PVC films not exposed to hydrofluoric acid fumes were retained in these films during flotation, while those particles on the sample slides exposed to hydrofluoric acid remained on the slides during flotation. However, this conclusion applies only to the particles in the 60 mμ to 1 μ equivalent $^{238}PuO_2$ diameter range which were investigated.

To determine the efficiency with which PVC (1%) can sustain encapsulation of particles less than 60 mμ in diameter (the equivalent $^{238}PuO_2$ diameter range of SNAP-9A particles), the same procedure was followed with ^{238}Pu-containing particles from Sandia, which ranged from around 20 to 100 mμ in diameter. The results of this experiment indicated that these smaller particles remained on the original slides whether or not the film had been exposed to acid fumes before float-off.

In the light of these results, the need for modification of this sample-preparation procedure was obvious. Several methods, some of which do not involve ashing the filter paper directly on the slide, were investigated.

3.3 PVA-SiO Layer Technique

For this experiment, five blank microscope slides were covered with a thin layer of PVA (polyvinylalcohol); a thin layer of SiO was vacuum-deposited on this. It was thought that the layer of SiO would serve as a protective coating for the PVA when a portion of filter paper was placed on the coated slide and ashed in an LTA-600 Low-Temperature Asher. After ashing, a few drops of PVC (1%) were added to the ash, forming a thin film. It was expected that, since PVA dissolves in water, the thin PVC film segments would separate from the slides easily during float-off, without the aid of hydrofluoric acid.

It was found that no cracks had developed in the layer of SiO during ashing and, therefore, that the PVA was adequately protected. However, the films could not be floated off. The films on four of the prepared slides would not float off at all, and although the fifth film separated along the edges of the slide, it soon folded over on itself because the PVA did not dissolve quickly enough. However, it is thought that the basic procedure has not been conclusively tested, since PVA was the only material used. Experiments to evaluate materials other than PVA were suspended until the results of the study described in section 3.4 were known. If successful, that experiment would preclude the PVA approach because its method of preparation is simpler.

3.4 Particle-PVC Suspension Technique

This experiment was conducted to determine the feasibility of suspending ^{238}Pu particles in a solution of PVC (1%) and pipetting the solution onto microscope slides. It was thought that this would encapsulate the particles of interest more completely and prevent them from remaining behind when film segments were floated off. The difference between this approach and the procedure employed in previous photoreversal experiments is significant. In preparation for earlier experiments, the ablated ^{238}Pu-containing particles from Sandia were suspended in a solution of methyl-ethyl-ketone, pipetted onto blank microscope slides, and ashed in an LTA-600 to remove the organic residue. After ashing, these slides were covered with a thin film of PVC (1%). Since the results of these photoreversal experiments showed that the ^{238}Pu particles (particularly those smaller than 60 $m\mu$ in diameter) were not removed from the microscope slides during float-off, it was considered probable that the PVC (1%) added to the ashed residue was not encapsulating the ^{238}Pu particles sufficiently.

To investigate this, ^{238}Pu particles suspended in a solution of PVC (1%) were pipetted onto three microscope slides. Nuclear emulsion plates were exposed to these slides for 24 hours to determine by the hollow-star technique the number and the locations of the particles on each slide. After this had been determined, the PVC films were floated off the slides. A few drops of PVC (1%) were pipetted onto the "old" slides and nuclear emulsion plates were again exposed to these slides. This flotation procedure was repeated with these films three more times. The results of scanning the nuclear emulsion plates indicated that approximately 35% of the particles (numerically) were lost between the original sample slides and the final slides which now contained the thin films of PVC (1%). The results, however, are only qualitative, in that hollow-star calculations were not made to determine whether there was any loss of Pu from the individual particles during float-off.

This method appears much more promising than the PVA-SiO layer technique. In experiments to investigate this technique further, it is recommended that individual particles be hollow-star counted to be sure that no portion of the ^{238}Pu remains behind during float-off.

3.5 PVC Sandwich Technique

This experiment was to evaluate the feasibility of positioning the SNAP-9A particles between two thin films of PVC to ensure complete encapsulation, thus retaining the particulates in the film during float-off. With this approach, the ^{238}Pu particles in an isopropyl alcohol solution would be pipetted onto microscope slides already covered by a thin film of PVC. After the alcohol had evaporated, the particles would be covered by a second thin film of PVC.

The usual procedure for preparing ashed samples for photoreversal work consists of pipetting a 1% solution of PVC onto the microscope slide containing the ^{238}Pu particles. Electron microscopy with these films, however, indicates that, in some instances, the PVC film is too thick for the electron beam to penetrate. Therefore, it was desirable to find a new method for preparing thinner films of PVC than are obtained by pipetting the PVC (1%) solution onto the microscope slides with the ashed sample.

The technique used to make very thin films of PVC was adapted from an article by B.D. Pate and L. Yaffe.[6] They used VYNS resin (a polyvinyl-chloride-acetate copolymer) with cyclohexanane as the solvent. Since VYNS in cyclohexanane does not spread satisfactorily on a water surface, they developed a new method for fabricating thin films.

Essentially their technique is as follows: A wooden barrier is placed in contact with one end of a sink filled with water. One to two millimeters of the resin solution is pipetted between the barrier and the side of the sink. The barrier is released from the resin and lifted from the water; the barrier is then lowered lightly onto the solidified film and moved away over the water surface, drawing out a thin film of resin, which feeds out of the solution band until the far side of the sink is reached or the solution is exhausted. The film thickness is governed by the speed at which the barrier is drawn across the water surface, the thinnest films being obtained with the highest speeds.

With this technique, film thicknesses from ~ 36 to 140 mμ were easily obtained and subsequently mounted on blank microscope slides. The double films of PVC were then covered with thin-film nuclear emulsion, exposed to photoreversal solutions, floated off the microscope slides, and prepared for electron microscopy. This part of the experiment was successful in that the double films of PVC containing processed thin-film nuclear emulsion were not too thick to prohibit electron microscopy. Future experiments with radioactive particles will be performed to determine if photoreversed particles are still lost during float-off.

3.6 Scanning Electron Microscope

This experiment consisted of a preliminary investigation to determine the usefulness of the scanning electron microscope for viewing photoreversed submicron particles encapsulated in thin films of PVC (1%). The interesting feature of this technique is the possibility of viewing the photoreversed submicron particles

directly on the microscope slides, thus precluding the necessity of floating them off.

Figure 8 shows two electron micrographs of photoreversed nuclear weapon debris particles which are encapsulated in PVC (1%) film and mounted on electron microscope grids. Although these particles are much larger than the ^{238}Pu particles involved with the SNAP-9A filter collections, the figure indicates that further experiments would be useful. To be investigated specifically is the effective resolution of the scanning electron microscope when viewing 25- to 50-mμ-diameter particles encapsulated in PVC (1%). Figure 9 illustrates the resolution with which submicron Al_2O_3 particles encapsulated in a thin film of PVC (1%) appear.

The direct-viewing technique with the scanning electron microscope could be of immediate use, since there are at present six SNAP-9A abort particles $\geq$ 25 mμ on several microscope slides being exposed to thin-film nuclear emulsion. Even though these sample slides were prepared for photoreversal by the normal ashing procedure described earlier in this report, it is very probable that the ^{238}Pu particles would be lost during float-off. However, after processing, it may be possible to view the larger ^{238}Pu particles directly on the microscope sample slides with the scanning electron microscope.

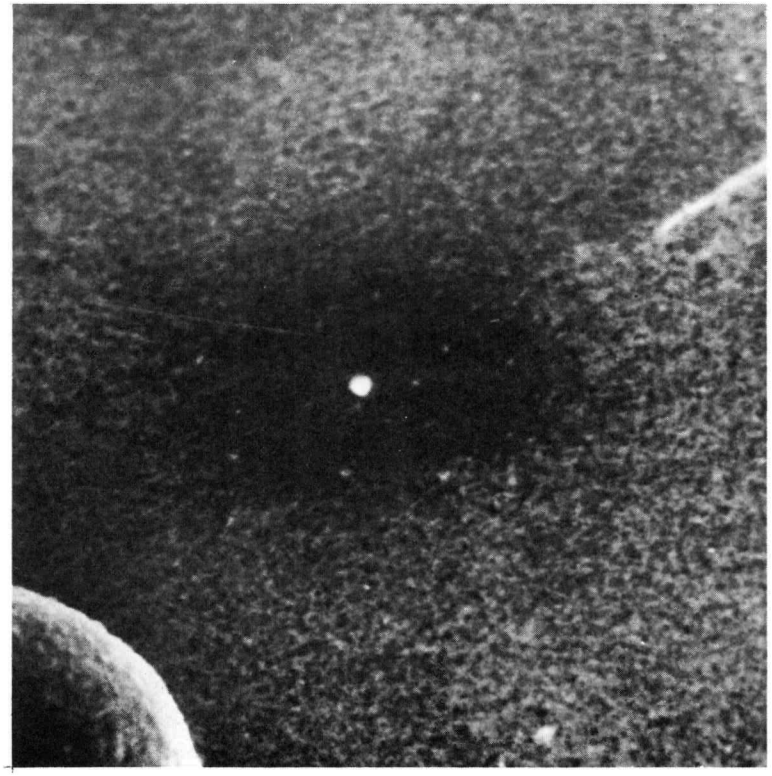
3.7 SCINTILLATION-SCREEN STUDIES

All remaining portions of the IPC filter samples (see Table I) containing ^{238}Pu particles which were received from HASL for SNAP-9A particle analysis were ashed, covered with collodion (4%), and exposed to the scintillation-screen/x-ray film arrangement for 12 days (see Appendix Section A-9).

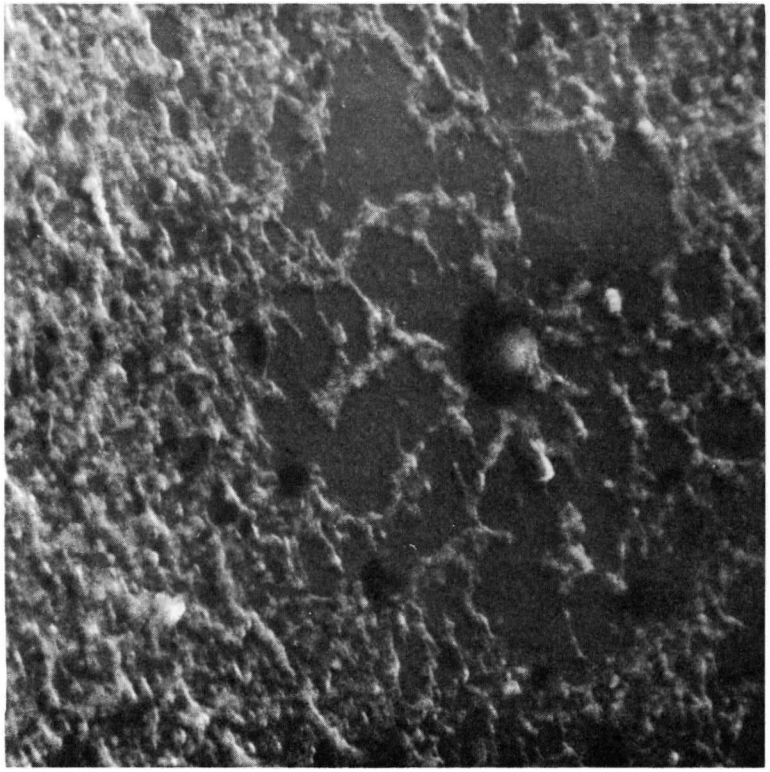
As explained[5], this technique provides a faster and more economical method for locating SNAP-9A particles greater than 50 mμ in diameter. Specifically, the optical scanning usually required for the hollow-star or thick-nuclear-emulsion methods is avoided.

The primary purpose of this work was to locate the larger SNAP-9A particles for further experiments with the photoreversal and fission-fragment tracking (see Appendix Section A-10) techniques. However, only two particles larger than 50 mμ were located. One out of every ten sample slides on which no particles larger than 50 mμ were found after exposure to the scintillation-screen/x-ray film arrangement was re-examined by the hollow-star technique. The maximum equivalent $^{238}PuO_2$ particle diameter found on these slides, as calculated by the hollow-star technique, was $\sim$ 36 mμ.

A secondary objective of this work was to evaluate the use of the scintillation screen as a means to decrease the exposure time necessary for SNAP-9A particles to produce clear areas in thin-nuclear emulsion during photoreversal. It was thought that SNAP-9A particles exposed to the scintillation-screen/thin-film nuclear-emulsion arrangement would produce larger clear areas in the thin-nuclear emulsion more rapidly than if the scintillation screen were omitted.

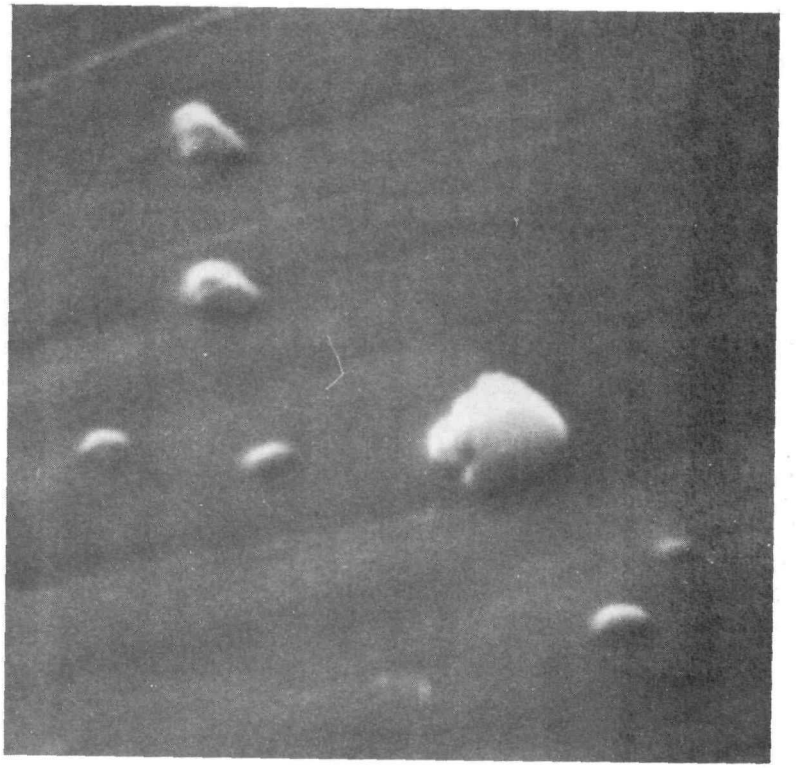


3000 X



10,000 X

Figure 8. Oscillographs of Photoreversed Weapon-Debris Particles



30,000X

Figure 9. Oscillograph of Al_2O_3 Particles Encapsulated in PVC (1%)

As reported previously[5], two experiments were proposed to evaluate the use of scintillation screens for this purpose.

In the first experiment, two flown control slides from Filter A-1155C were exposed for 30 days by the scintillation-screen/thin-film nuclear emulsion method. Previously, the thick-nuclear emulsion method applied to samples originating with this same filter indicated the presence of alpha stars with a maximum of five tracks. Scanning the two flown control slides processed for the experiment failed to show any photoreversed clear areas indicating stars with more than five tracks.

In the second experiment, three sample slides from Filter A-1499B were exposed to nuclear emulsion plates for one week. After this, the sample slides were subjected to a scintillation-screen/thin-film nuclear emulsion exposure for one month. Calculations of the ten hollow stars found from scanning the nuclear emulsion plates indicated that the largest equivalent $^{238}PuO_2$ particle diameter was ~ 36 mμ. All hollow stars on the nuclear emulsion plates were scribed and eventually used in an attempt to locate the photoreversed clear areas on the processed sample slides.

During exposure of the three sample slides to thin-film nuclear emulsion, portions of the scintillation screen separated from the sample slides and prevented the development of clear areas by the scintillation-screen/thin-film nuclear emulsion method. Although the hollow-star location technique was employed for those areas on the processed sample slides which appeared to have been in contact with the scintillation screen during the exposure period, no photoreversed clear areas were observed. The results of the optical and electron microscopy performed on the ten particles are shown in Table VI. As this table indicates, several of the locations where photoreversal should have produced clear areas were covered with extraneous debris. This debris can be assumed to have obstructed the development and observation of clear areas.

As reported previously[5], the data tabulated in Table VI result from an effort to relate the sizes of the clear areas caused by photoreversing six ablated $^{238}PuO_2$ particles from Sandia to the total number of alpha disintegrations they emitted during a 22-hour exposure to thin-film nuclear emulsion. It was tentatively concluded from this experiment that a total of from ~ 100 to 125 alpha disintegrations are necessary to produce a photoreversed clear area that can be recognized by optical microscopy. However, because the ablated $^{238}PuO_2$ particles themselves were missing from the clear areas, an actual particle size/clear area size comparison could not be made.

A study of the total alpha disintegrations of ^{238}Pu particles as listed in Table VII suggests that all but one of the ten particles used in the scintillation-screen/thin-film-nuclear-emulsion experiment, as shown in Table VI, could have been recognized by optical microscopy without the aid of the scintillation screen. The failure to observe any clear areas, however, indicates that the

Table VI. Pertinent Data and Results of Scintillation-Screen/Thin Nuclear Emulsion Study with Ten SNAP-9A Abort Particles from Filter A-1499B

Slide No.	Particle Designation	Equivalent $^{238}PuO_2$ Diameter (mμ)	Total Alpha Disintegrations during 30-day Exposure	Remarks*
1	1	32	279	Extraneous debris present
2	1	36	402	Not covered by thin-film nuclear emulsion during preparation for photoreversal exposure
	2	24	123	Lost during float-off
	3	24	123	Unable to locate clear area in electron microscope
	4	26	162	Extraneous debris present
3	1	20	72	No work done with this particle
	2	24	123	Extraneous debris present
	3	26	162	Extraneous debris present; unable to locate clear area in electron microscope
	4	28	213	Extraneous debris present
	5	28	213	Lost during float-off

*Float-off operations in preparation for electron microscopy were attempted on only the four particles with the least adjacent extraneous debris.

Table VII. Photoreversal Results of Six Sandia ^{238}Pu Particulates Covered with PVC and Exposed to Thin-Nuclear Emulsion for 22 Hours

Particle Designation	Equivalent $^{238}PuO_2$ Diameter (mμ)	Total Alpha Disintegrations during Exposure Time	Diameter of Clear Area
1	146	832	~ 17
2	100	267	~ 4
3	75	111	~ 3
4	74	110	~ 1
5	72	99	*
6	70	91	*

*Only a general lightening of an ~ 8 to 10 μ area was obtained.

extraneous debris on the sample slides was enough to mask the development of photoreversed clear areas recognizable by optical microscopy with or without the aid of the scintillation screen.

It is suggested that further experiments be conducted, with measures to avoid including extraneous debris on the sample slides and to ensure total and lasting contact of the scintillation screen with the thin-film nuclear emulsion during the exposure period.

3.8 SILICONE OIL

Efforts were made to develop a suitable method for detecting SNAP-9A particles on silicone-oil-coated phenolic slides in a Litton Model A247 Sampler. The work described in previous Quarterly Reports[5, 7, 8] was performed with several second-stage silicone-oil slides exposed to nuclear-emulsion plates for 42 and 97 days in an attempt to obtain a numerical size distribution by the hollow-star technique.

Scanning the nuclear emulsion plates indicated that the largest concentrations of ^{238}Pu particles were in distinct clusters, due to the configuration of the sampler intake nozzles. This configuration cuased a concentration of particles into small areas on the silicone-oil-coated slides.

The results of the 42-day nuclear-emulsion-plate exposure indicated that this was insufficient time for many of the ^{238}Pu-containing particles to produce recognizable hollow stars. On the other hand, the 97-day nuclear-emulsion-plate exposure caused a maze of overlapping alpha tracks which prevented an efficient counting of tracks. Therefore, it was considered impractical to use the hollow-star technique to obtain a numerical size distribution.

Although silicone oil is a very good medium for collection and retention of particulate matter, electron-microscopic examination of submicron particles imbedded in this material is not possible. Either the submicron particles must be separated from the silicone oil, or the silicone oil must be diluted before electron microscopy is attempted.

With this in mind, it was proposed in a previous report[5] to conduct an evaluation of ultracentrifugation as a technique to separate the submicron Pu-containing particles from the silicone oil. If such separation proved feasible, examination of these particles by electron microscopy was thought to be possible.

To test the ultracentrifugation technique, the following centrifugation-dilution steps were performed:

Preliminary Preparation

Silicone oil on a 1-3/4 by 2-1/2-cm portion of blank impactor slide was washed into a centrifuge tube with hexane. This liquid was diluted with more hexane up to 52 ml.

Step 1: Liquid was stirred for 5 minutes.
Step 2: Liquid was centrifuged for 30 minutes at 6000 rpm.
Step 3: One drop from center of liquid column was placed on slide, covered with 4% collodion, and prepared for electron microscopy.
Step 4: All of the liquid except the lower five ml in the centrifuge tube was extracted. The remaining five ml was diluted with hexane up to 52 ml.

The centrifugation-dilution procedure was followed four times. The four sample extractions prepared for and examined by electron microscopy revealed that the fourth dilution was sufficient for particle work with a transmission electron microscope.

None of the second-stage impactor slides with ^{238}Pu particles imbedded in silicone oil were processed by this technique, primarily because of the difficulties with the float-off procedure, as explained in section 3.2. The centrifugation-dilution method should be tried, however, in conjunction with the scanning-electron microscope and PVC-sandwich techniques, described in sections 3.5 and 3.6.

3.9 LITTON GRID STUDIES

Three electron microscope grids from Flight 3036, performed on August 23, 1966, by Litton Industries[9], were received at Tracerlab for determination of the presence of SNAP-9A abort particles. It was planned to locate the particles by the hollow-star technique and examine them in the electron microscope.

The three grids were exposed to a nuclear-emulsion plate for 30 days. Examination revealed only one hollow star. Calculation indicated an equivalent $^{238}PuO_2$ diameter of about 17 mμ. Optical examination showed that the hollow-star center fell so close to the edge of the grid that it could not be observed in the electron microscope.

The grids were subsequently exposed to another nuclear emulsion plate for 60 days. Optical examination showed no further development of hollow stars.

4.0 CONCLUSIONS

Several conclusions based on experiments not directly related to photoreversal studies are stated below:

- The results of the leaching study suggest that as the equivalent $^{238}PuO_2$ diameters of SNAP-9A particles decrease, the percentage of the original particle leached increases.

- The scintillation-screen/thin-film-nuclear-emulsion studies should be conducted again, with the precautions necessary to ensure lasting contact of the scintillation screen with the thin film of nuclear emulsion during the entire exposure period.

- The application of the centrifugation-dilution technique to separate the ^{238}Pu particles from the silicone oil to permit electron microscopic examination of these particles appears promising.

Several conclusions based on the photoreversal studies were reached. The particle-PVC solution technique, developed to ensure encapsulation of the photoreversed ^{238}Pu particles during float-off, was successful in that 65% (numerically) of the particles remained with the PVC (1%) films during float-off. However, the evaluation of this technique is qualitative, since no hollow-star calculations were made for individual particles to determine if there are any losses to the ^{238}Pu particles during float-off.

Two other techniques -- the PVC-sandwich and the scanning electron microscope approaches -- appear more promising because they provide complete retention of particles in the PVC films, while with the particle-PVC solution method, there is some particle loss during float-off. However, this loss is not serious, provided there is no loss of portions of individual ^{238}Pu particles during float-off.

Although no SNAP-9A particles have yet been studied to understand the changes they may undergo in their descent through the atmosphere, most problems encountered in photoreversal preparation have been solved. The photoreversal technique should prove to be the best method to locate these particles as well as beta-active fallout particles for study by electron microscopy.

5.0 RECOMMENDATIONS

It is recommended that the sample slides with SNAP-9A particles now being exposed in thick-nuclear emulsion be processed and scanned after a two-year exposure to determine the ^{238}Pu content in the three to five-track star range. It is further recommended that these same slides be used to augment the present leaching study results.

Although the contract requirements have been fulfilled, the photoreversal experiments could not be completed in the time allotted. Since the specific objective of these additional experiments is to view the submicron SNAP-9A abort particles to determine whether they are in their primary form or have become attached to larger airborne particulates before washout by rain, Tracerlab recommends that the photoreversal investigations be completed.

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APPENDIX
PARTICLE ANALYSIS EQUIPMENT AND TECHNIQUES

A-1. LTA-600 LOW-TEMPERATURE ASHER

In the Tracerlab LTA-600 Low-Temperature Asher, radiofrequency-excited oxygen reacts with a single layer of filter paper on a microscope slide, producing CO_2 and H_2O and leaving an extremely thin ash of filter paper in addition to the previously-collected inorganic particulates. Oxygen gas at a pressure of 450 microns of Hg is admitted to the reaction chambers at a flow rate of 15 cc/min. The ashing temperature is approximately 150° C. After half an hour at 70 watts of rf power, the ashing is complete. Even though the ash is almost completely transparent in the optical microscope and appears quite fragile, it is firmly enough attached that the slides can be carefully handled without loss of sample.

A-2. THICK-NUCLEAR EMULSION

Previous methods for counting alpha tracks to obtain measurements of radioactive aerosol particles have been limited to collections on membrane filters or impactor slides. The membrane filters or impactor slides are placed in contact with the emulsion during the exposure period.[1a, 2a, 3a, 4a] After the emulsion has been processed, the alpha tracks produced by particles collected on the impactor slides are observed in an optical microscope. Immersion oil is added to the membrane filter slides to make the membrane filter transparent so that alpha tracks on these slides may be counted. This method was improved by placing the membrane filter in contact with the emulsion-coated slide and transferring the particles from the filter to the emulsion as the filter was stripped away before exposure.[5a]

We modified these techniques to allow investigation of radioactive particles collected in IPC by adding liquid Ilford L-4 nuclear emulsion to the ashed samples on microscope slides.

Under darkroom conditions, with a Wratten Series OA Safelight, a few grams of Ilford L-4 are added to a 40-ml centrifuge tube and melted at 50° C in a water bath. About 15 drops of emulsion are pipetted onto each slide of ashed IPC; after the emulsion has dried, the slides are stored in a light-tight box at 5° C for the exposure period.

After the desired exposure time, the emulsion is processed under darkroom conditions as before. The slides are first allowed to come to room temperature. They are placed in Kodak D-19 developer, diluted 1:1, for 5 minutes, in Kodak indicator stop bath for about 10 seconds, and in Kodak rapid fix for twice the time required for the emulsions to clear. The slides are then rinsed for 30 minutes in running tap water and dried in a dust-free area. Each slide is scanned in an optical microscope at 150X for alpha stars.

A-3. ALPHA TRACKS AND ALPHA STARS

Alpha tracks result from emulsion grains of silver halide which have been made developable by the passage of a charged alpha particle. When a silver halide crystal absorbs energy from a moving charged alpha particle, this mycrocrystal, under a chemical reducing agent, will convert to metallic silver more rapidly than will the non-irradiated crystals in the emulsion. Under the optical microscope, the alpha tracks appear as a straight line (with a maximum length of 25 μ) composed of opaque grains of metallic silver. Tracks radiating from a common particle form an alpha star, with the unseen Pu particle at the center.

A-4. CALCULATION OF EQUIVALENT SPHERICAL DIAMETERS FROM TRACK COUNTS IN THICK-NUCLEAR EMULSION

Assuming spherical particles and a composition of pure $^{238}PuO_2$, the sizes of the SNAP-9A particles were estimated with a modification of the formula derived by Leary[1a]:

$$d = \left(\frac{10^{-2} \times C}{t}\right)^{1/3}$$

where d = particle diameter in microns
C = number of alpha tracks in the star, assuming a factor of 2 for relating alpha tracks to disintegrations
t = length of exposure in minutes

A-5. GRAPHICAL DIFFERENTATION[6a]

Histogram data may be converted to a cumulative curve to smooth out any irregularities caused by an insufficient number of measurements.

This is justifiable because the limited number of particles which can be counted or measured results in pronounced statistical fluctuations. The best method is to construct a smooth cumulative curve, not from the histogram directly, but from the experimental data (Figure A-1). By this method, random errors in measurement and fluctuations in the particle number per fraction can be smoothed. The differential curve (Figure A-2) can then be constructed by graphical differentation from the integral curve. This smooths out such physically improbable rises as that at the extreme right of the histogram.

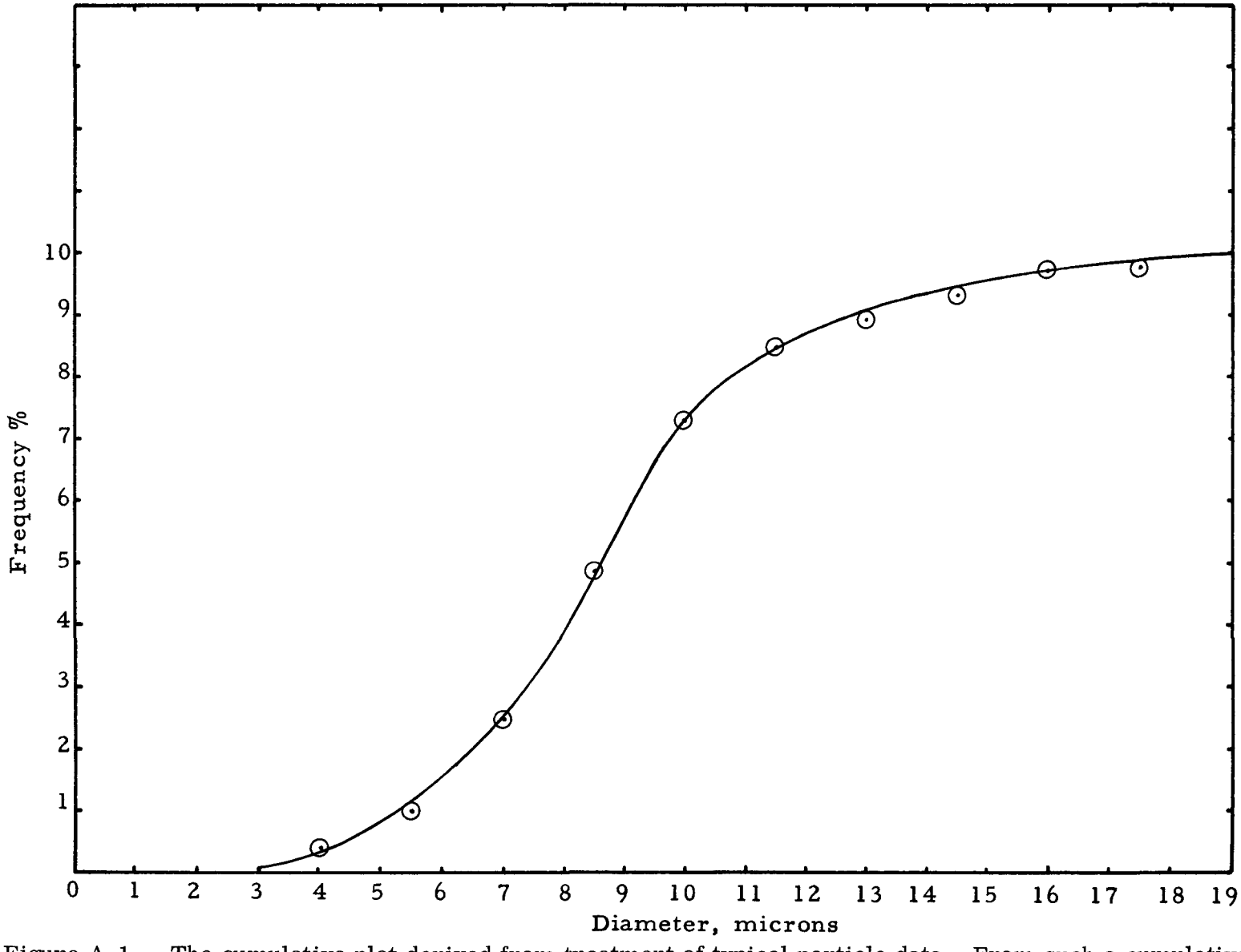

Figure A-1. The cumulative plot derived from treatment of typical particle data. From such a cumulative curve irregularities in the experimental points caused by insufficient measurements can be smoothed out by differentation.

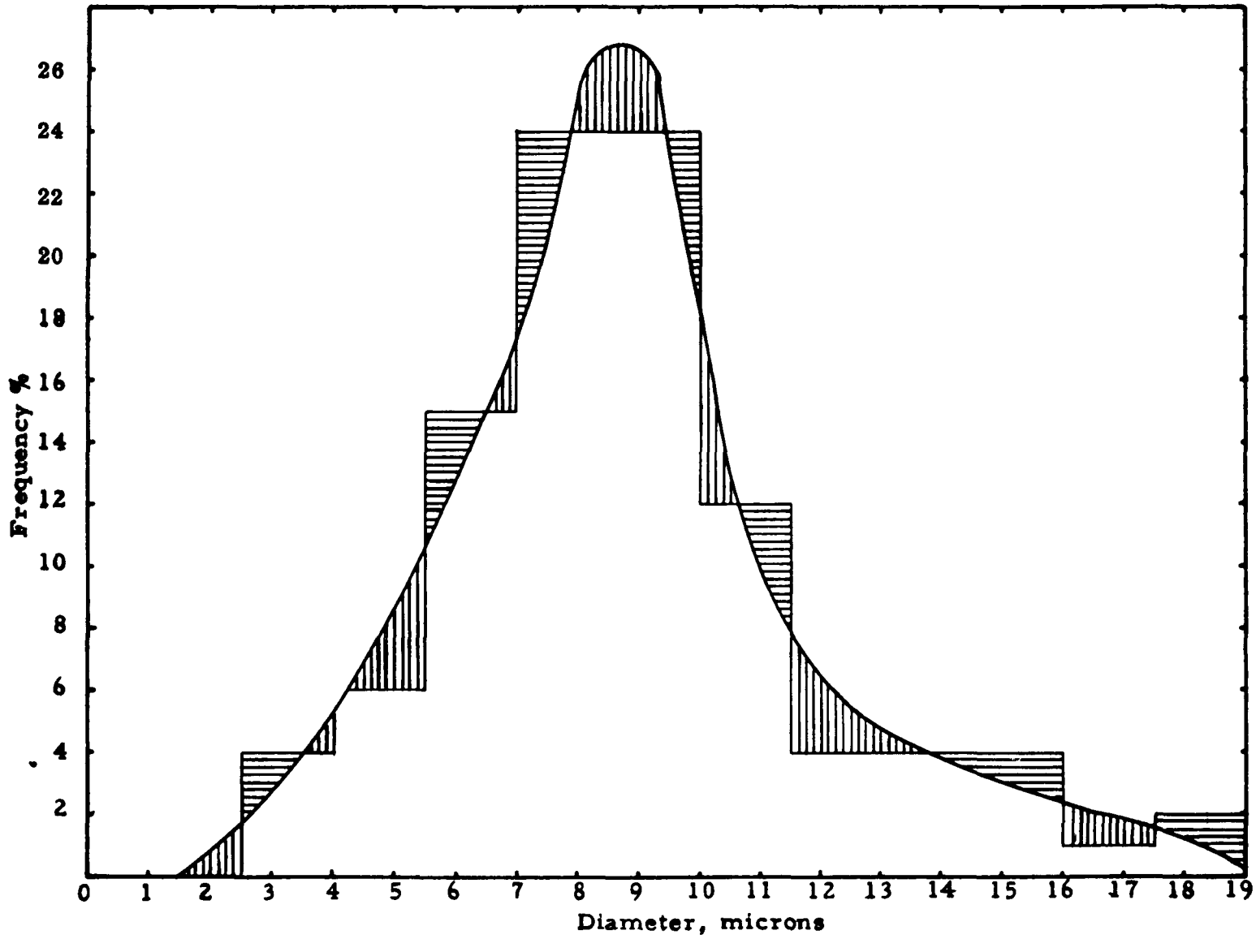

Figure A-2. A histogram and size frequency curve constructed by differentation from the cumulative plot. The histogram represents the experimental data and the smooth curve represents the differentiated curve from the cumulative plot.

A-6. Z-TEST[7a]

This is a test of the hypothesis that two means ($\bar{x}_1$ and $\bar{x}_2$) of different normal distributions are equal when both standard deviations are known. The statistics used for testing that $\bar{x}_1 = \bar{x}_2$ (or $\bar{x}_1 - \bar{x}_2 = 0$) are a function of the difference between the means. The acceptance region for the two-sided test procedure is given by the interval ($-K\,\alpha/2$, $K\alpha/2$) where α is the level of significance and $K\alpha/2$ is the $100\,\alpha/2$ percentage point of the normal distribution. The 95% confidence level (or five percent significance level) was used as the acceptance region. The statistical number was $\leqq 1.96$ for the acceptance of the hypothesis $\bar{x}_1 = \bar{x}_2$.

Example: The average single-track value per Filter A-1583 sample slide was 220 ± 218; the average value for the blank slides exposed with these slides was 92 ± 52. To test if these means are the same, the Z-Test was used:

$$Z = \frac{\bar{x}_1 - \bar{x}_2 - d}{\left[\frac{\sigma_1^2}{n_1} \pm \frac{\sigma_2^2}{n_2}\right]^{1/2}} = \frac{220 - 92}{\left[\frac{218^2}{30} \pm \frac{52}{6}\right]^{1/2}} = 2.57$$

A-7. PHOTOREVERSAL TECHNIQUE

(a) Sample

Submicron radioactive, alpha-emitting particles are encapsulated in a thin film of PVC (1%) on a microscope slide.

(b) Preparation of Emulsion

Under darkroom conditions, with a Wratten Series OA Safelight, 10 grams of Ilford L-4 emulsion is placed in a 250-ml beaker. Twenty milliliters of distilled water is added to this, and the mixture is placed in a water bath at 50° C for 10 minutes; then it is placed in the freezing compartment of a refrigerator. The emulsion has cooled to the right consistency when a thin film of emulsion can be drawn over a 4-cm loop of platinum wire, which is dipped into the emulsion and removed slowly. The film thus formed is carefully placed on a slide; the film adheres to the slide, and the wire loop is removed When the emulsion is dry, the slide is placed in a light-tight box and stored in a refrigerator at 5° C for exposure.

(c) Development

After exposure, the slide is developed for 15 minutes in Kodak D-19 developer (diluted 1:1) and rinsed in stop bath for 30 seconds. The reduced silver is then bleached for 2 minutes in a solution of 2 grams of potassium permanganate and 2.5 ml of concentrated sulfuric acid in 500 ml of distilled water; then it is cleared for 2 minutes in a solution of 25 grams of potassium meta bisulfite in 500 ml of distilled water. After the film has been rinsed in distilled water for 2 minutes and exposed to normal light for a few seconds, it is developed

a second time in Kodak D-19 developer for 5 minutes, rinsed in stop bath, and cleared in Kodak rapid fix for approximately 5 minutes. Finally, it is rinsed in running water for 20 minutes and dried before being optically scanned for clear areas.

A-8. HOLLOW-STAR TECHNIQUE

The hollow-star technique is used to measure the alpha activity to permit calculation of the equivalent $^{238}PuO_2$ diameters of particles. The exposure arrangement uses shims to maintain a known distance between the particles and the slide of thick-nuclear emulsion.

Figures A-3 and A-4 show how this arrangement is used to make the desired calculations. The shim distance AA' and the exposure time t (in minutes) are known. By optical microscopy, the center A' of the hollow star is located. Also, an alpha track such as BE, at a convenient distance from the center (about 400 μ), is selected, and the distance A'B is measured with a filar micrometer. By counting the single alpha tracks in the circle of radius A'B (including those intersecting this circle), N is determined.

To calculate the solid angle Ω in which these tracks were produced, the following relationship is used (see Figure A-3):

$$\text{Tan } \theta = \frac{A'B}{A'A} \qquad \theta = \text{Tan}^{-1} \frac{A'B}{A'A}$$

The solid angle Ω in which N tracks are produced is given by:

$$\Omega = \int_0^{\theta} 2\pi \sin\theta d\theta = 2\pi(1 - \cos\theta)$$

The number of alpha disintegrations D_t from the particle during time (t) is calculated by:

$$D_t = \frac{4\pi}{\Omega} \times N = \frac{2N}{1 - \cos}$$

The alpha activity D_m (in dpm) of the particle is calculated by:

$$D_m = \frac{D_t}{t}$$

Likewise, the equivalent diameter d (in μ) of a $^{238}PuO_2$ particle is calculated by:

$$d = \left(\frac{10^{-2} D_t}{2t}\right)^{1/3}$$

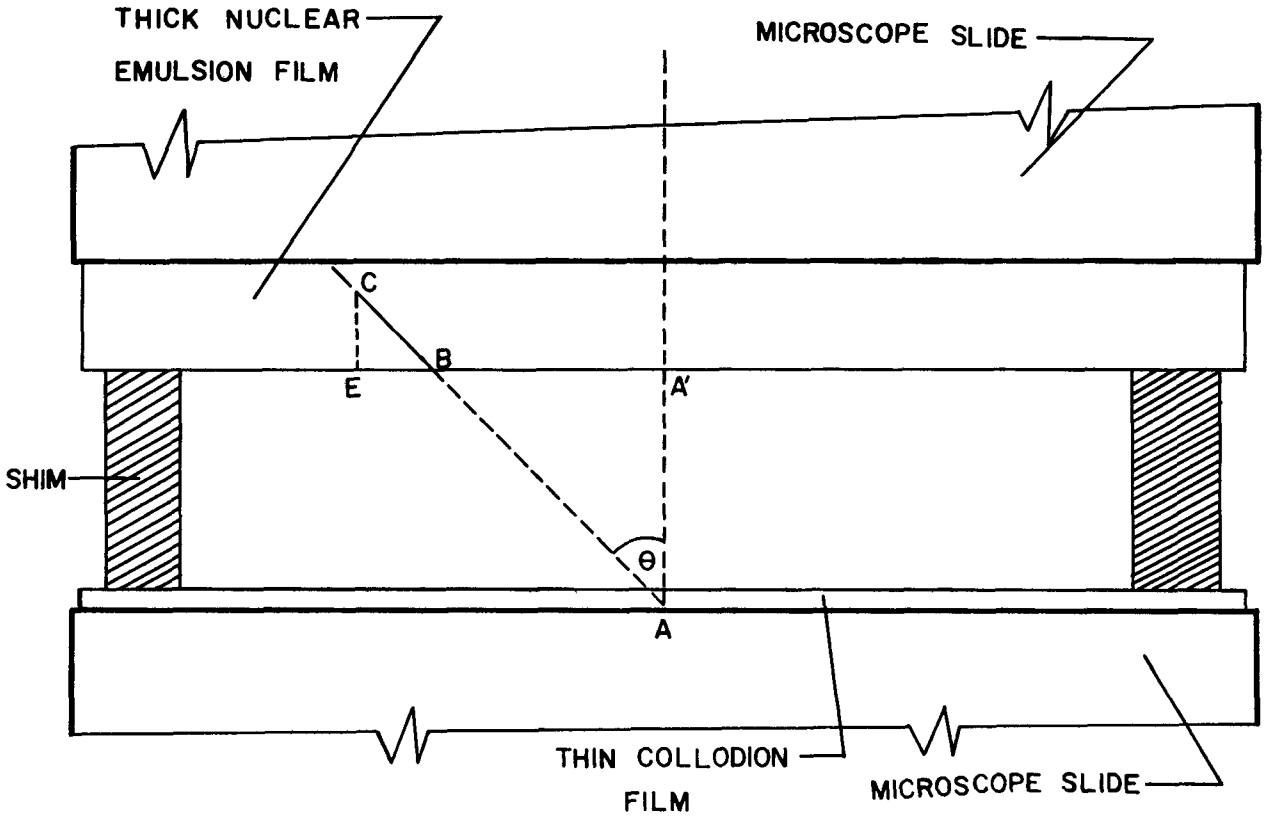

Figure A-3. Side view of geometric configuration used for calculation of ^{238}Pu disintegrations from hollow-star data.

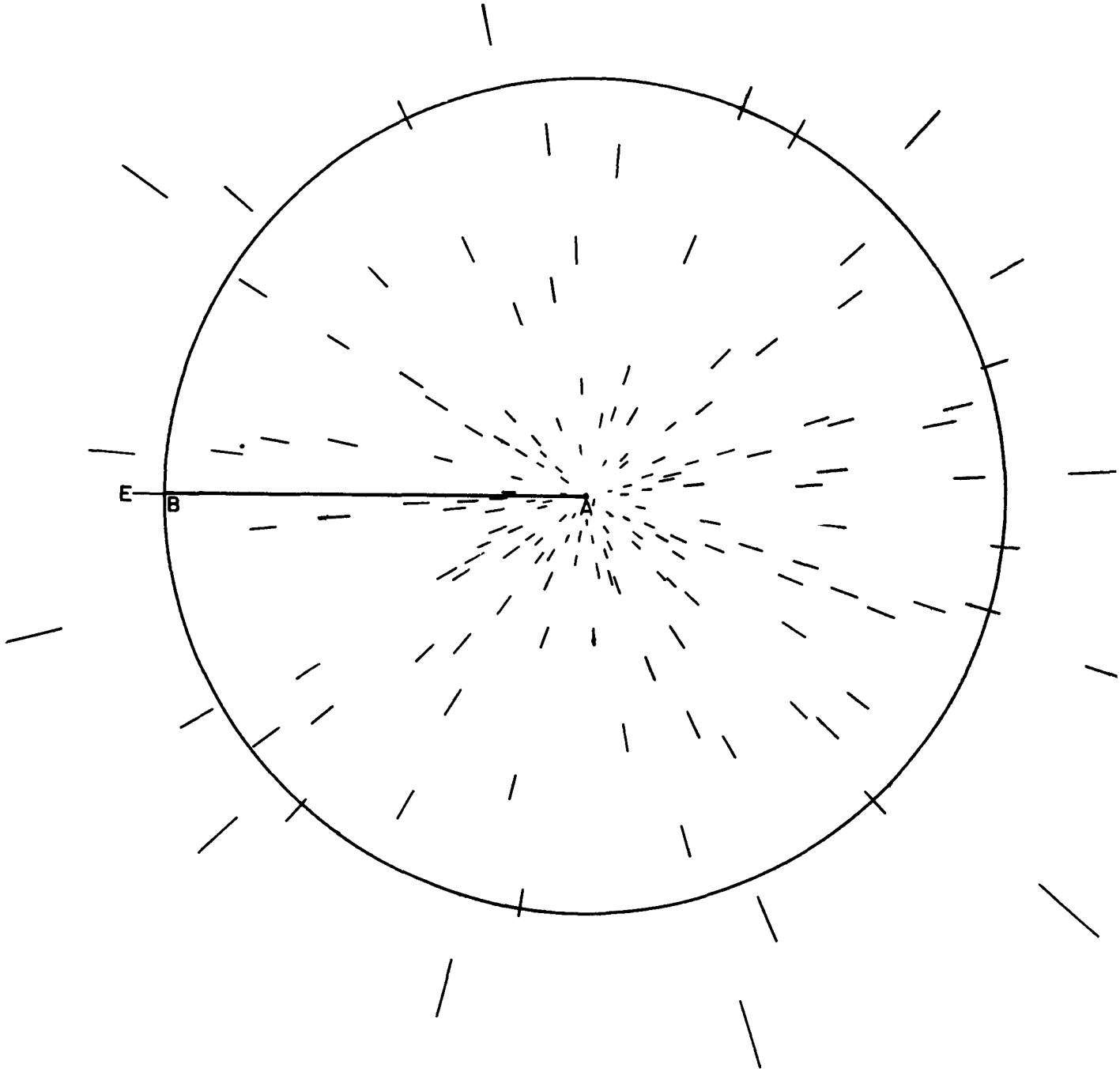

Figure A-4. Top view of geometric configuration for calculation of ^{238}Pu disintegrations from hollow-star data.

A-9. SCINTILLATION-SCREEN/X-RAY FILM ARRANGEMENT FOR LOCATING LARGE SNAP-9A PARTICLES IN ASHED IPC FILTER SAMPLES

Because of the low alpha activity of the SNAP-9A particulates, x-ray film must be exposed for extremely long times. Although the thick-nuclear emulsion and hollow-star techniques can also be used for this work, with these techniques the nuclear emulsion plates must be scanned with an optical microscope. Therefor, the scintillation-screen/x-ray-film technique, a faster and more economical method, was used to locate the larger SNAP-9A particles. With this method, the optical scanning required with the thick-nuclear emulsion or hollow-star techniques was avoided.

An ALPH-A-TAK scintillation screen, manufactured by the Har-D-Cal Company, Williston, South Carolina, was used. It consists of a 1-mil polyester film coated with zinc sulfide (silver-activated). The phosphor density ranges from two two three mg/cm^2. With this arrangement, the alpha emitted from the SNAP-9A particles on the sample slide strike the scintillation screen, producing a flash of light that exposes the x-ray film.

A-10. FISSION-FRAGMENT TRACKING TECHNIQUE

This technique is based on A.G. Malmon's findings[8a] concerning electron-microscopic observation of non-etched fission-fragment tracks recorded in cellulose nitrate by neutron irradiation of uranyl acetate.

Malmon's experiment was duplicated at Tracerlab with fine ^{235}U powder encapsulated in a thin film of cellulose nitrate. The preliminary results from this study are encouraging. Scanning the irradiated cellulose nitrate film with the electron microscope revealed numerous fission-fragment tracks surrounding the ^{235}U particles in an arrangement resembling a hollow star of alpha tracks.

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