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DETERMINATION OF THE PHYSICAL AND CHEMICAL CHARACTERISTICS OF ENVIRONMENTAL PARTICULATES CONTAINING RADIONUCLIDES

by

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INTERIM REPORT

ROAP 21BAS, Tasks 03, 04, 13

ROAP 21AMI, Task 18

"Determination of the Physical and Chemical Characteristics of Environmental Particulates Containing Radionuclides"

A. J. Cummings, J. A. Hodgeson, and E. W. Bretthauer

June 1974

I CONCLUSIONS

Significant results obtained for Tasks 03, 04, and 13, ROAP 21BAS, and Task 18, ROAP 21AMI, to date are:

A. In-house work has shown that it is feasible to collect, locate, and isolate alpha emitting particles collected on air filters. An Interim Progress Report on this work is shown in Appendix A. This work also showed that large conglomerate particles (10-20 μm) were subject to fragmentation into smaller (1-2 μm) particles.

B. Preliminary chemical and physical analyses of airborne plutonium particles collected in Area 11 of the Nevada Test Site (NTS) have been made by GE Vallecitos Nuclear Center. Analytical data on air filters included size, specific activity, chemical composition, and isotopic ratios. The data collected revealed the presence of at least four classes of particles: (1) small, high-activity in an organic matrix, (2) large, low-activity in a silicate matrix, (3) large, low-activity in an organic matrix, and (4) small, high-activity oxides.

C. A purchase order has been given to GE Vallecitos Nuclear Center, through the Air Force Technical Applications Center at Patrick Air Force Base, to perform detailed chemical and physical characterizations of individual plutonium and

americium containing particles collected on filter paper during atmospheric sampling.

D. A dichotomous air sampler for fractionating atmospheric particulates into respirable vs non-respirable ranges, has been ordered and will be used to collect air samples for the study.

E. Initial air sampling has begun at NTS. A remote location southwest of Las Vegas has been chosen as a background collection site.

F. Requirements for the physical and chemical characteristics of environmental iodine-129 will not be satisfied under present contractural arrangements. Instead an intensive literature review of the state-of-the-art knowledge has been performed. A report derived from the review is being prepared.

II RECOMMENDATIONS FOR FY-75

Plans for the coming fiscal year relative to this task are:

A. Efforts will be continued to obtain from the AEC laboratories and the Department of Defense material relative to this project. Significant findings from the literature review will be incorporated into a final report, which will also include results from the particle study being conducted

by GE Vallecitos Nuclear Center.

B. In-house screening of local background samples will be conducted to determine level of atmospheric fallout of plutonium. Some selected samples will be forwarded to the GE Vallecitos Nuclear Center for detailed analysis.

C. Preliminary screening will also be conducted of the NTS samples. A few selected samples will be forwarded to the GE Vallecitos Nuclear Center for detailed analysis.

D. Dichotomous air samples will be collected in the environs of other nuclear facilities, e.g., Rocky Flats, Hanford Atomic Facility, and the Savannah River Nuclear Facility. If suitable arrangements can be made, samples will also be collected directly from each source. Such information should give insight into the effects of resuspension and aging on plutonium particulates.

III INTRODUCTION

The output of ROAP 21BAS consists of a series of research reports on environmental transport of the radionuclides plutonium and iodine-129 which define parameters for

population exposure models. These parameters will be related to the various types of environments typical of regions near emission sources. While plutonium will receive primary emphasis, iodine-129 and americium-241 will be studied concurrently.

Most previous investigations dealing with environmental radionuclide pollution have been confined to the determination of the concentration of specific radionuclides at certain locations and in certain media by simply assaying the radioactivity of the collected samples. Relatively little is known about the physical and chemical characteristics of airborne plutonium or the effect of time and of soil and climatic parameters on these characteristics after deposition. This knowledge, however, is essential for any prediction of transportation phenomena in terrestrial and aquatic environments and for biological availability determination.

Our approach to these tasks has been (1) to review currently available information on the chemical and physical properties of radioactive plutonium, iodine, and americium in the environment; (2) to conduct sampling at various types of nuclear facilities involved in the nuclear fuel cycle both at the source and in the surrounding environs, and (3) to have detailed particle characterizations performed under contract. By sampling at the source and in the local

environs, information can hopefully be derived on the effects of aging and resuspension on plutonium containing particles. One critical problem that must be emphasized at this point is that of obtaining approvals to sample source terms at any of the more important nuclear fabrication facilities within the United States. In addition, all nuclear fuel reprocessing plants are closed and not expected to open for at least one year.

IV RESULTS AND DISCUSSIONS

A. In-House Particle Study

A number of large diameter, 18.5 cm, air filters from a previous REECo air sampling project were obtained for analysis as a preliminary step to field sampling. Several of these samples were subjected to autoradiographic techniques to determine the presence of radioactivity. Those areas showing activity were subjected to a cursory in-house analysis. This analysis consisted of dissolving those indicated areas of filter material in carbon tetrachloride and filtering off the contained particles. The dried filtrate was subjected to alpha counting for confirmation of radioactivity and to microscopic examination. Several particles were physically isolated for individual observation and photography. The results of this preliminary investigation are discussed in an interim progress report (Appendix A). In this report it is shown that it was possible to locate the

gross position of an alpha emitting particle on filters, and to isolate and manipulate these particles for further study, i.e., observation and gamma and/or alpha spectroscopy. Some of the larger particles (10-20 micron diameter) were very fragile and tended to shatter into fragments on the order of 1 to 2 microns. This observation suggests that during aging larger particles may degrade into particles in the respirable range.

B. Extramural Particle Study

A single 18.5 cm filter was submitted to the GE Vallecitos Nuclear Center for detailed examination. A 6.5 cm^2 section was processed by the GE Vallecitos Nuclear Center and 32 radioactive particles out of at least 55 observed were randomly selected for further analysis. All 32 radioactive particles were microscopically sized. Mass spectroscopy and electron probe analysis were performed on 12 of the sized particles. The analysis indicated that the observed geometric diameter ranged from <0.5 to 17.0 μm with an activity of 4 to 26 femtocuries/particle, and further, that these particles could be grouped into

four distinct classes (See Appendix B). The size, activity and chemical composition of those particles are summarized in Tables I & II of Appendix B.

A purchase order was accepted by the Air Force to provide the services of GE Vallecitos Nuclear Center for the subsequent analysis of air filter samples obtained under this task. The work to be performed by GE Vallecitos Nuclear Center can be summarized as follows:

1. Approximately 100 individual air samples will be submitted and examined for radioactivity by autoradiographic techniques.

2. The contractor will select approximately 25 of the above air samples for detailed analysis.

3. The detailed physical analysis will consist of the following:

a) Total number of alpha-active particles present.

b) Estimate of alpha activity per particle.

c) Estimate of physical size ranges of alpha-active particles.

d) Density estimate from size and chemical characterization (see Section 4 below).

e) Photographs of at least 10 particles.

f) At least 2 submicron particles will be examined by electron microscopy to determine size and shape.

g) Particles of diameter greater than 1 micron will be sized and shaped by optical microscopy.

4. A detailed chemical analysis will consist of:

a) Gross elemental composition determination by mass spectroscopy and electron probe analysis.

b) Ion microprobe analysis will be performed on at least 2 particles per filter.

c) Isotopic composition of any uranium, americium, and plutonium present will be determined.

d) Elemental distribution of at least one large particle will be determined by scanning electron probe analysis.

e) Gross chemical composition by electron microprobe will be attempted on the two submicron particles selected for step 3-f above.

f) Crystal structure for at least two particles will be determined by x-ray and/or electron diffraction.

C. Field Sampling

The field sampling program was begun by placing two high volume (air flow $\simeq 0.5$ m^3/min) air samplers in Area 8 at the NTS at the surface of ground zero of a previous air blast. A remote mountain peak 15 miles southwest of the city of

Las Vegas has been chosen as a sampling area to obtain background samples. A southwesterly direction was chosen due to the prevailing winds in this section of the country. This area allows placement of our sampler in a region that should be relatively free of contamination from the NTS and from locally released particulates. The site is presently being used by the Union Pacific Railroad as a communication site.

These air samples, as well as others later obtained, are now being screened by EPA personnel. Some selected samples will later be shipped to GE Vallecitos Nuclear Center for detailed analysis. The pre-screening consists of subjecting the filters to gross alpha counting and in-house autoradiography. The 37 mm filters are counted directly; particles collected on 185 mm filters are resuspended and refiltered on 37 mm filters prior to counting. The detector used is a zinc sulphide screen and photomultiplier-tube pulse counter. The device is at least 49% efficient with a background of 0.18 counts per minute (170 femtocuries). This background is sufficiently low to permit screening of most filters. Selected 6.5 cm^2 sections from four 185 mm filters have been resuspended and screened and found to contain greater than 1.5 picocuries of alpha activity/filter.

A two-stage dichotomous air sampler based on a design developed at the National Environmental Research Center, Research Triange Park, NC, has been ordered and should be in the

field by approximately August 1, 1974. This sampler, using 37 mm diameter millipore type AA filters, separates the particles into two groups, (1) those less than 2 μm in diameter, and (2) those in the 2 to 20 μm diameter range. This separation is designed to yield fractions of respirable versus non-respirable particulates directly. A third and separate filter simultaneously collects a total particulate fraction. The dichotomous sampler has the advantage over conventional cascade impactors of a sharper size fractionation. The particle re-entrainment problem, i.e., carry-over of large particles to lower stages known to occur with Anderson samplers, is avoided. In addition, the smaller size of the filter collection area is much more amenable to the intended subsequent analysis.

D. Literature Surveys

A literature review is being conducted on the physical and chemical properties of plutonium containing particles in the environment. Those publications available are listed in Appendix C.

An extensive literature survey on the physical and chemical characteristics of environmental iodine-129 has been conducted and a report summarizing this review will be published separately.

V APPLICATION TO ROAP 21AMI, TASK 18

The subject task calls for, (a) developing methodology for characterizing individual particulates in the environment, and (b) defining the shape, density and specific activity of plutonium particulates which are airborne, in soils, and on plants. Thus the physical and chemical characterization work called for under these tasks is quite similar to the requirements of ROAP 21BAS, Tasks 03, 04 and 13. Only the nature of the samples differs. ROAP 21BAS will require airborne samples collected at or in the vicinity of nuclear facilities. ROAP 21AMI, in addition, calls for soil and plant samples collected from the environment. However, the effort to this point has concentrated only on developing the methodology for isolating and characterizing individual particles, and such work could most easily be performed using air filter samples. Thus, the reports for tasks in both ROAPs have been combined.

During FY-75 soils and plant samples will be collected simultaneously and at the same location that air samples are being taken. These samples will also be submitted for analysis under the contract with GE Vallecitos Nuclear Center. Concurrent sampling will allow a comparison of plutonium containing particulates isolated from soil, plants, and air.

APPENDIX A

PROGRESS REPORT FOR ROAP 21AMI - TASK 17

DEVELOPMENT OF METHODOLOGY FOR DETERMINATION OF PHYSICAL CHARACTERISTICS OF AIRBORNE PARTICLES CONTAINING PLUTONIUM

by

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July 3, 1973

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DEVELOPMENT OF METHODOLOGY FOR DETERMINATION OF PHYSICAL CHARACTERISTICS OF AIRBORNE PARTICLES CONTAINING PLUTONIUM

by

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and E. W. Bretthauer

I. INTRODUCTION

Plutonium activity can be detected in air samples taken in areas where the soil has been contaminated with plutonium. The possibility that this resuspended plutonium represents a hazard on inhalation depends, primarily on the respirable characteristics of the particles.

The basic problem is to determine the physical characteristics (size, shape, density, mass, distribution, specific activity, etc.) of airborne debris bearing particles or chemical combinations of plutonium isotopes. The method we have developed can be broken down into three categories: (1) detection, (2) separation, and (3) characterization. It was developed for examination of microsorban filters obtained from high velocity impact air samplers located at the Nevada Test Site (NTS). These filters have a retentivity of 100% for particles of three microns or larger.

The work reported herein describes background activities and the procedures developed for characterization of individual plutonium containing particulates collected from air samples.

II. DEVELOPMENT OF PROCEDURE

The procedure developed is described below and flow-charted in Figure 1. Section III contains the stepwise procedure.

A. Detection

1. Autoscintography

A typical fifteen-centimeter impacted filter is shown in Figure 2. The first step is to produce an autoradiograph of the filter to determine the location of radioactive particles. The autoradiograph is produced on

a photographic emulsion thru light emission generated by alpha-particle bombardment from the plutonium upon zinc sulfide (ZnS) crystals. A sandwich is made of (1) the air filter, (2) a sheet of ZnS, and (3) a photographic emulsion[1,2]. This sandwich is then enclosed in a light-tight package and left for a period of time (see Figure 3). The exposure time is determined by the light sensitivity of the emulsion and the radioactive strength of the particle. Little has been done as yet to optimize this step of the process. Typical particles require 21 days of exposure when Kodak X-ray type film is used. This time has been reduced to a one-to-two day exposure using fast film such as Kodak Royal Pan (ASA 400) There is sufficient background of data to determine gross activity levels at this step[2,4]; however, additional work must be done. Recordings of film density as a function of known activity levels would have to be made. This would require acquisition of a densitometer and particles of known size and activity. Then, for various film types, a study would be made of the film grey scale as a function of radioactivity level. The most difficult step would be in obtaining known, fractional nanocurie plutonium samples and a densitometer of fractional micron spatial resolution. A preliminary calibration attempt is shown in Figure 4b. This autoradiograph represents a drop of 5,000 dpm plutonium solution. The solution consists of particles of 0.1 to 0.3 micron diameter in suspension. The exposure was on Kodak Royal Pan film for 16 hours. The activity is so great that it is impossible to determine the location of a single particle. Discrete points are visible; however, the number of particles per point has not been determined. We are attempting to refine this approach by depositing a small amount of the solution on an electron microscope grid. Observation and autoradiography of the grid may allow determination of spot size for a single particle.

The radiograph from the above step is then examined for activity. Figure 4a shows the results of a typical exposure with three points of activity readily seen.

2. First Cut and Count

The area of interest containing the source of radiation is removed from the air filter for further processing. A circle of

approximately 1 cm in diameter centered on the particle is cut from the sample with a cork borer and subjected to alpha spectrometry. The alpha count at this step is included only to determine whether or not the particle is contained in the filter aliquot. Typically, particles are embedded in the filter material and give a distorted spectrum when counted at this point. If the count shows activity, the filter is prepared for visual microscopic observation, photography, and resuspension.

B. Resuspension

1. Refiltration

The cut sample from step A is dissolved in reagent grade carbon tetrachloride and refiltered onto a 2.5 cm, 0.3 micron micropore filter (preliminary attempts at fractional filtration have been made but were found to be unnecessary since the same results are achieved by going immediately to the smallest pore size filter).

2. Visual Observation, Autoradiography, Removal and Alpha Count

The second filtered sample is now observed visually through a light microscope and particles of size 20 microns and larger are photographed and removed to a glass microscope slide for analysis. The filter is then set up for a second autoradiograph to locate remaining radioactive particles. Figure 5a shows typical particles removed at this stage. Considerable difficulty has been encountered with fracturing of these large particles into small ($\leq$5 micron) fragments (Figures 5b and 5c). We have found it possible to obtain stereophotographs of the larger particles. The stereophotos, however, are of only minor interest due to the shallow depth of field inherent in microscope lenses. Figure 5d is a stereopair of a copper particle. When properly viewed, some depth of field is seen.

Removal of the particles is aided through the use of a Sensaur Pneumatic micro-manipulator and Zeiss photomicroscope (see Figure 6). After removal, they may be subjected to X-ray diffraction, electron probe analysis and alpha spectroscopy. The analytical instruments require the particle to be attached to a glass probe. Figure 7 shows some such particles. The probe is formed from a pulled glass rod. A short, thin section of the pulled glass is waxed into a fine capillary tube and mounted in the

micro-manipulator head. Rubber cement is added to the probe tip and the particle picked up from the filter paper. The chosen particle and probe tip are deposited on a microscope slide by melting the wax on the capillary tip.

The second autoradiograph is developed and examined for activity. Those portions showing activity are then subjected to the same process of observation, photography, and removal as described above.

C. Characterization

1. Size Determination

An estimate of particle size can be made from visual observation through the microscope. A calibrated grid reticle is available for use in the eyepiece. Alternately, size can be compared to glass spheres or objects of known diameter. Photographs of known size particles or grid scales can be stereoscopically superimposed over particle photographs to estimate size. Due to particle irregularities such measurements are only rough estimates. A review of the literature shows that particle size can be related to autoradiography spot diameter[3,4,5]. This, however, is true only for particles of pure elements. This diameter could be roughly determined visually through the microscope. There is a recording electrophoresis densitometer available for use, although it has not been tested for resolution in the micrometer range. We will evaluate several types of film for spot size versus radioactivity to correlate with other work. Size determination will then be based on visual and autoradiography spot size measurements.

2. Specific Activity

Specific activity is defined as radioactive disintegrations per unit time per unit mass. This measure has a twofold application as it may be applied to either gross alpha counting or alpha spectroscopy. The latter case requires a determination of the various alpha emitters present on or in a particle. The parameters of the spectrometer, i.e., resolution and counting efficiency, allow relation back to the energy and type of the radionuclides. Then, since the specific activity of these nuclides is known to a good degree of accuracy[6], the mass of each nuclide present

may be calculated. These individual masses have further significance to be considered in the section II C 3 (Mass/Density Measurement).

The relation of specific activity to mass as determined by gross counting (no isotope identification) technique is quite different. These data will be used to determine aerodynamic and inhalation characteristics of particles and to facilitate calculation of radiation dosage from inhalation of a given volume of air.

3. Mass/Density Measurement

The specific activity and alpha spectroscopy measurements discussed previously allowed a determination of the amounts and types of radionuclides. These can be related to total particle mass, assuming the nuclides to be attached to an unknown substance, through electron probe and X-ray diffractometer analysis. These measurements can give an estimate of the constituents of a particle and relative percentages. The three measurements then combine to determine the make-up of a particle as to types of element, amount of each and total particle weight. Density is then determinable from weight and size. A second means of mass determination will be developed to supplement electron probe and diffractometer measurements.

4. Electron Probe and X-Ray Diffraction

a. Debye-Scherrer X-Ray Powder Diffraction Method

The X-ray diffraction analysis of small particles necessitates the use of a film method in order to integrate the intensity of the diffraction arcs by the Debye-Scherrer Powder method.

The Debye-Scherrer camera used for particles is 57.3 mm in diameter and 35 mm in width. The particle is mounted on a small glass fiber with an appropriate adhesive by use of a micro-manipulator. The fiber and particle are placed in a mount in the center of the camera and the camera is loaded with a strip of non-screen X-ray film. The sample is exposed to copper Kα X-radiation for a period of one to fifty hours. If the particle is large enough and has a crystalline structure, the X-ray film, upon development, will contain a series of diffraction arcs. The position and intensity of the arcs are used to determine the crystalline

structure and relative amounts of elements. The crystalline structure and the elemental analysis will usually indicate the chemical compound(s) in the sample.

Limitations of the Debye-Scherrer method when used for particles are:

1. The particle must be large enough (10-20 microns depending upon the sample) to produce a diffraction pattern.

2. A sample containing a large amount of iron will have a high background due to copper Kα induced fluorescence from the iron.

3. Quantitation is not possible because this laboratory does not have a densitometer.

b. Electron Microprobe

The electron microprobe yields qualitative and quantitative information on areas one to fifty microns in diameter on a sample surface. The information is obtained by bombarding a sample under vacuum with a focused electron beam to produce characteristic fluorescence X-rays from the elements in the sample. The X-rays energies are determined by energy dispersive techniques in a single crystal goniometer containing a thin window gas-flow proportional counter. The data for qualitative analysis are collected on a strip chart recorder in units of goniometer degrees versus peak height in counts per second. The goniometer values in degrees plus the known 'd' value of the analyzing crystal in the goniometer applied to the Bragg equation gives the X-ray energy. The total spectrum and associated peak heights indicate the type and quantity, respectively, of elements in the sample.

The limitations of the probe are:

1. The sample must be of low volatility.

2. The minimum size particle is about two microns in diameter.

3. The sample must be optically flat for quantitation.

III. STEPWISE PROCEDURE

The step-by-step procedure thus far developed is listed below.

A. Large Filter Autoradiograph

B. Removal of Active Areas

C. Dissolution and Refiltration of Each Active Area

D. Separate Large Particles from Filtrate

1. Large Particles

    a. Microscopic Observation and Photography

    b. Alpha Spectroscopy

    c. Electron Probe and X-Ray Diffractometer Analysis

2. Filtrate

    a. Autoradiography

    b. Microscopic Observation and Photography

    c. Removal

    d. Alpha Spectroscopy

    e. Electron Probe and X-Ray Diffractometer Analysis

E. Analysis of all of the above data to determine types of nuclides present and the size and shape of particles bearing this nuclide.

IV. SUMMARY

Some success has been experienced in the characterization of particles containing plutonium. We have found it possible to locate the gross position of particles by autoradiography and further to observe and handle these particles. These techniques have been developed through a cooperative effort by the EPA-NERC-LV staff, particularly Mrs. Betty Mitchell and Mr. Dale Modine. Further effort is required in the areas of autoradiography, electron probe, and X-ray diffraction before final results can be tabulated. Preliminary features of the particles characterized will be published in a subsequent report.

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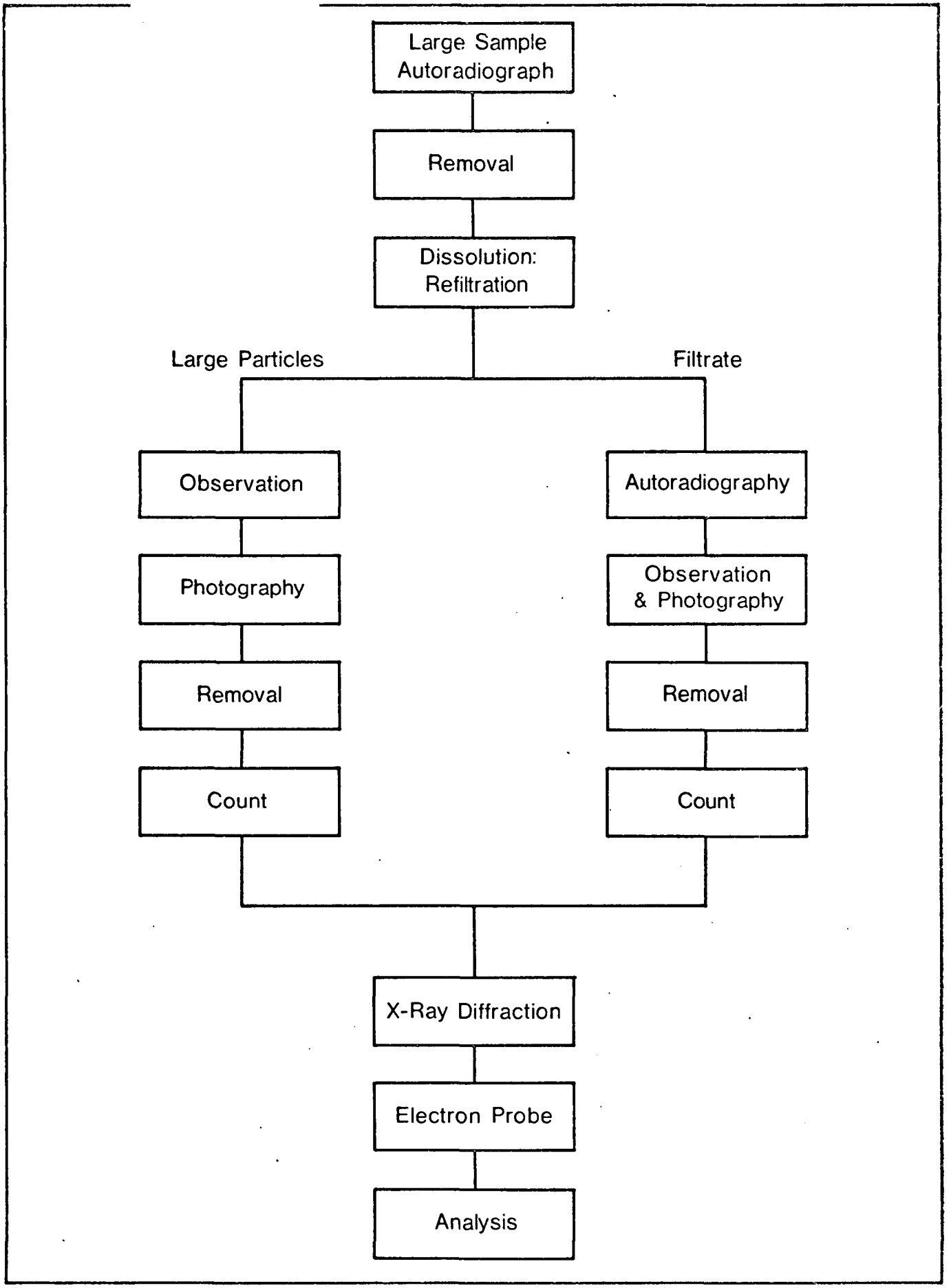

Figure 1
Process Flow Chart

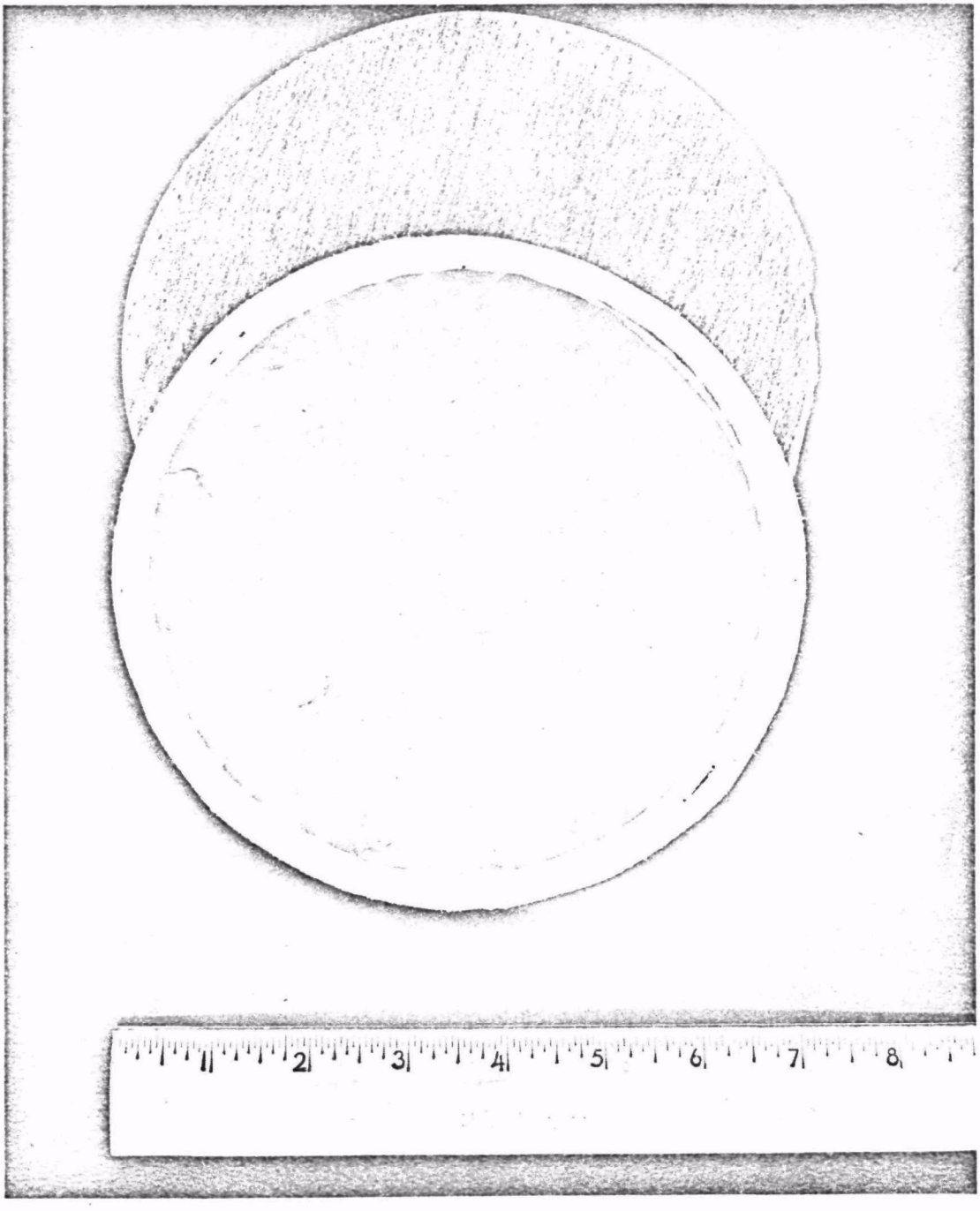

Figure 2
Air Sample Filter

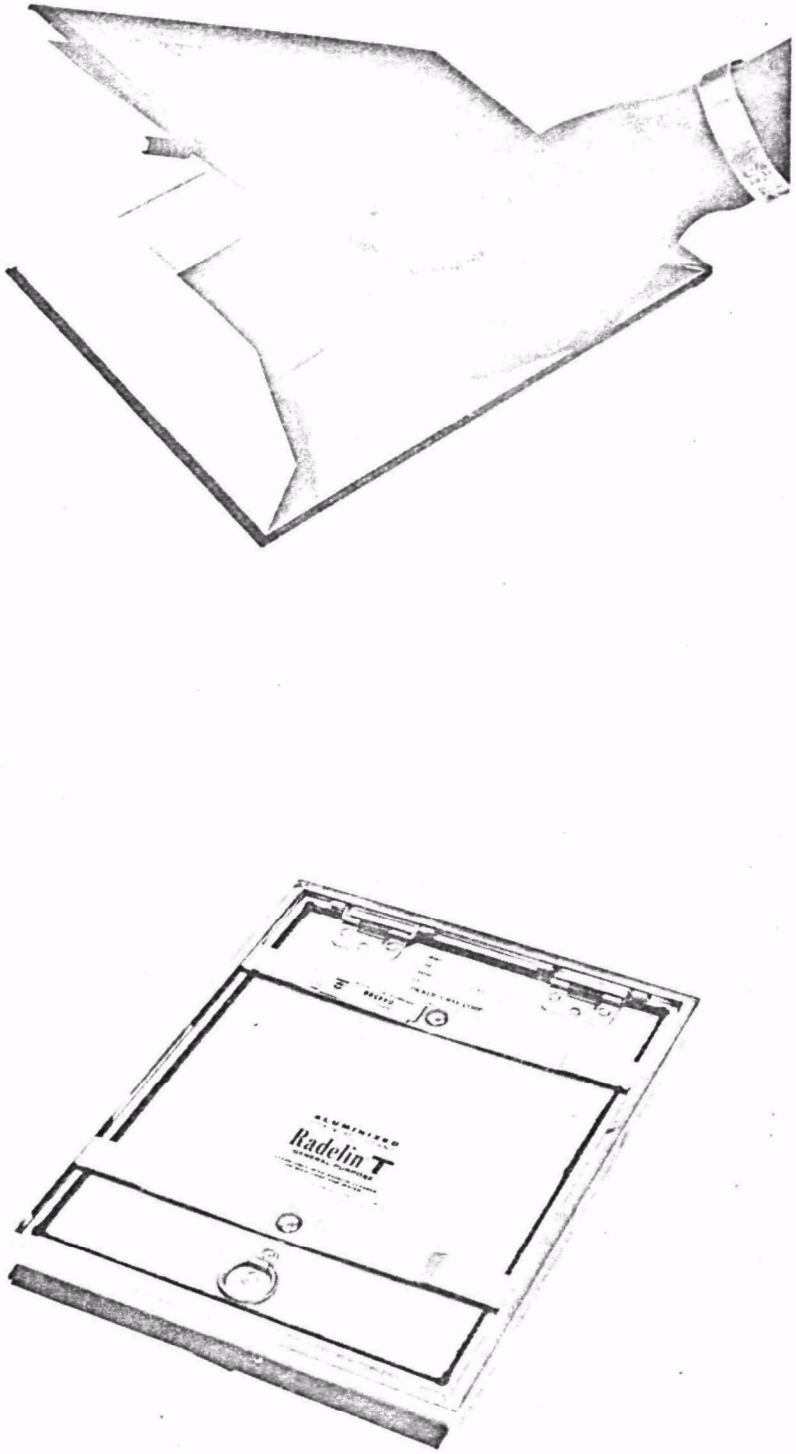

Figure 3
Autoradiograph Sandwich

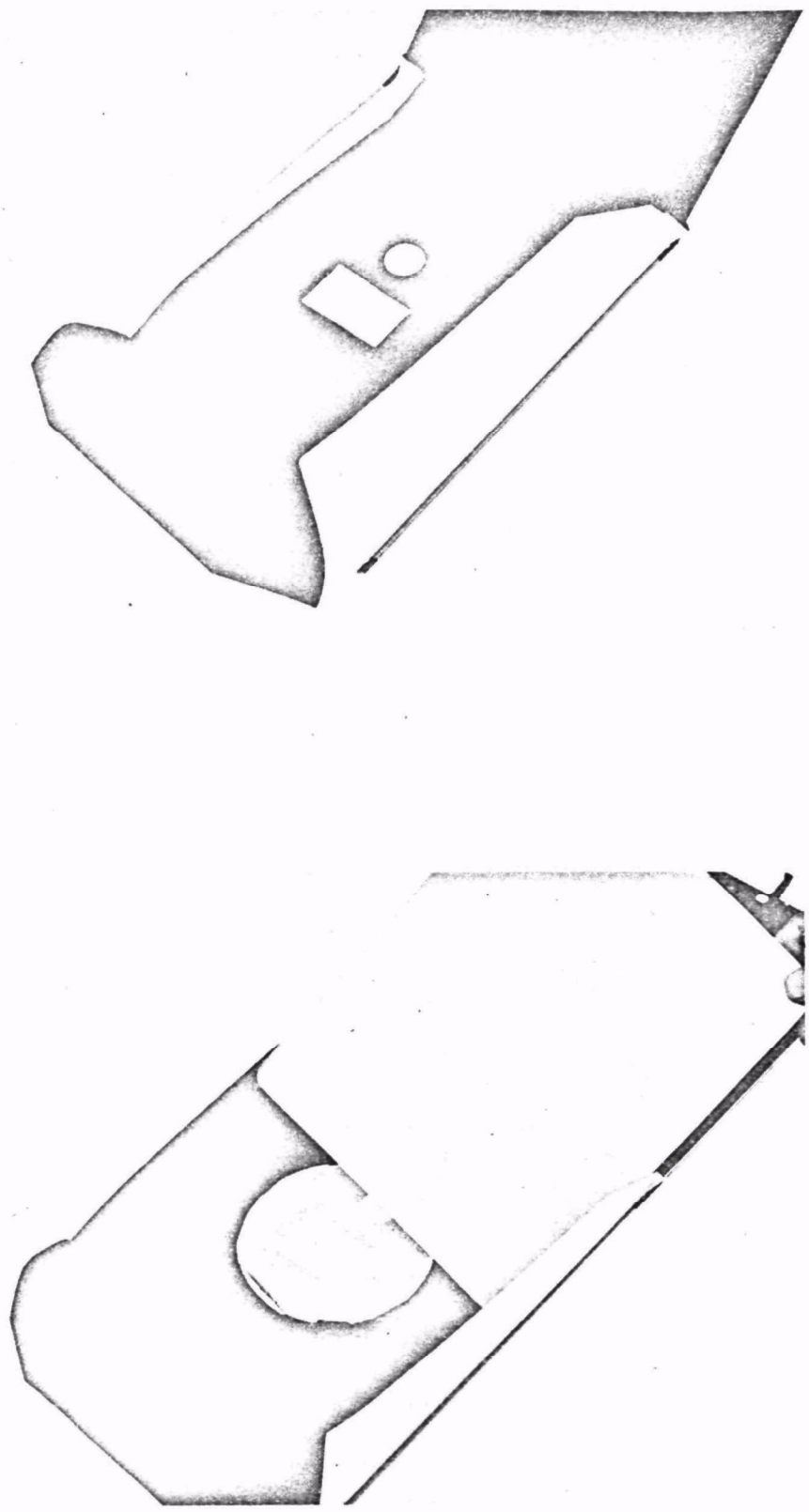

Figure 3
Autoradiograph Sandwich

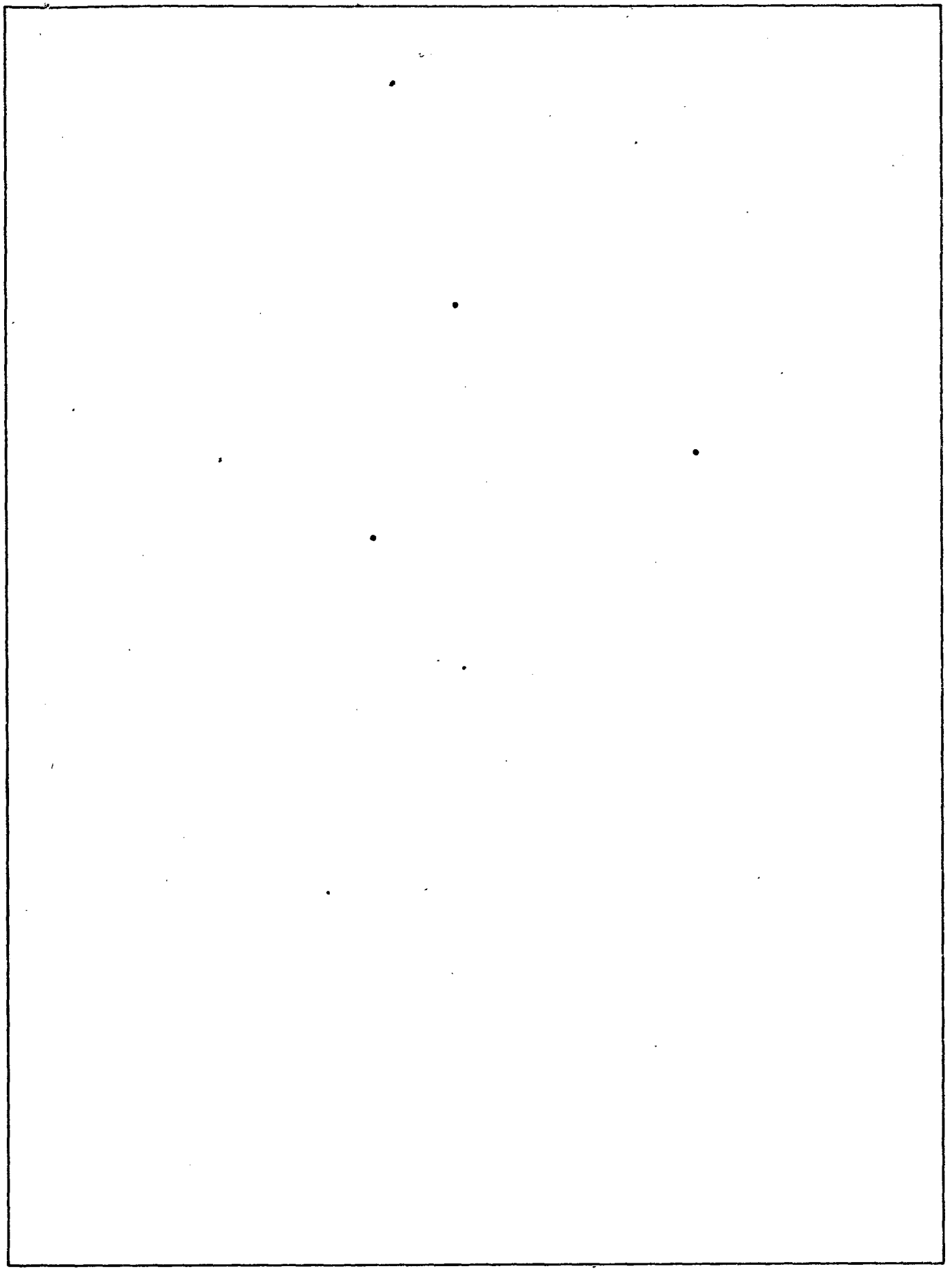

Figure 4a
Autoradiograph of 15 cm Filter

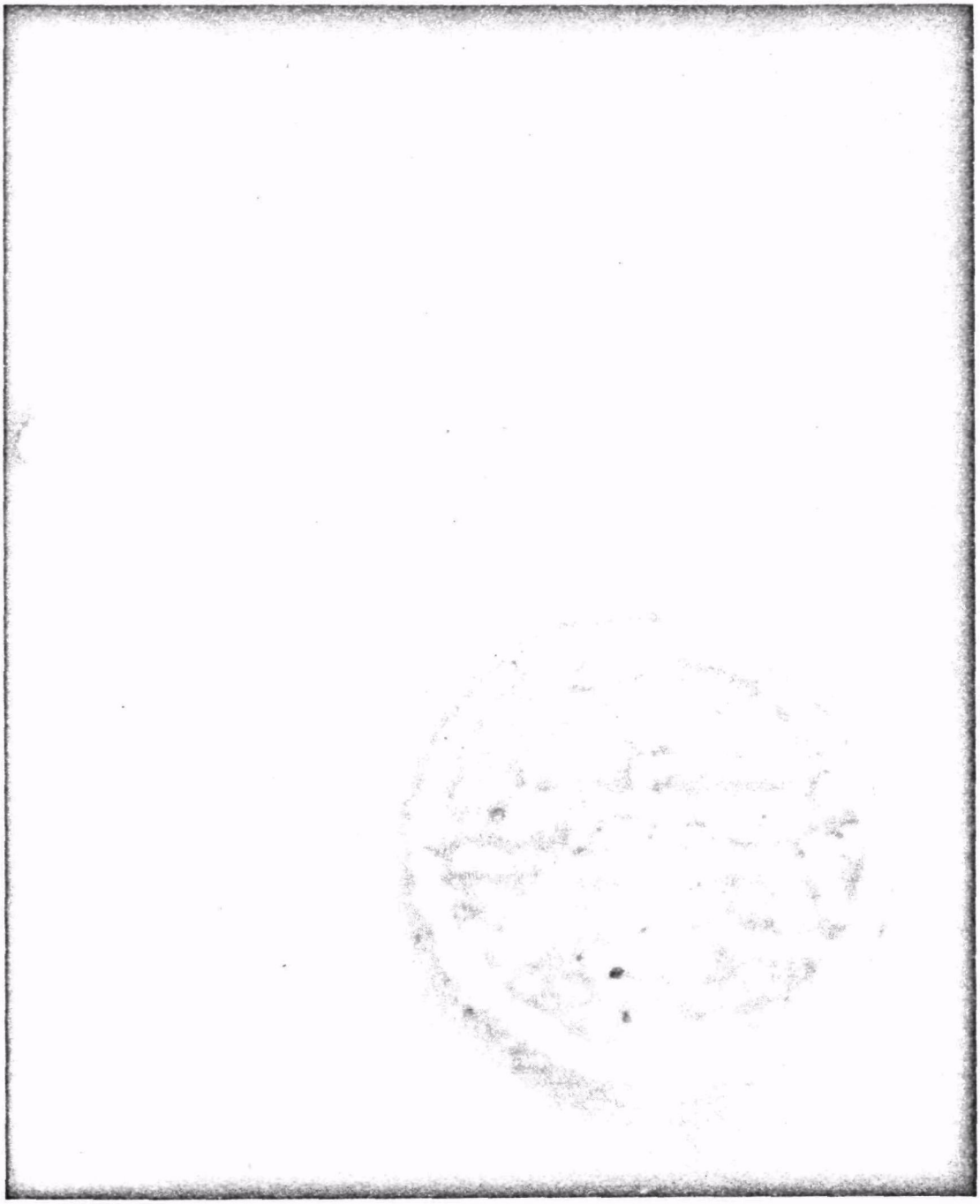

Figure 4b
Autoradiograph of Plutonium Particle in Solution

Figure 5a
Typical Large Particles

Figure 5b
Large Particle Before Fracture

Figure 5c
Large Particle After Fracture

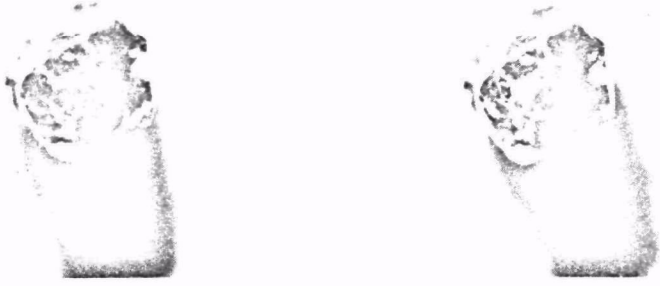

Figure 5d
Stereo Pair; Copper Particle

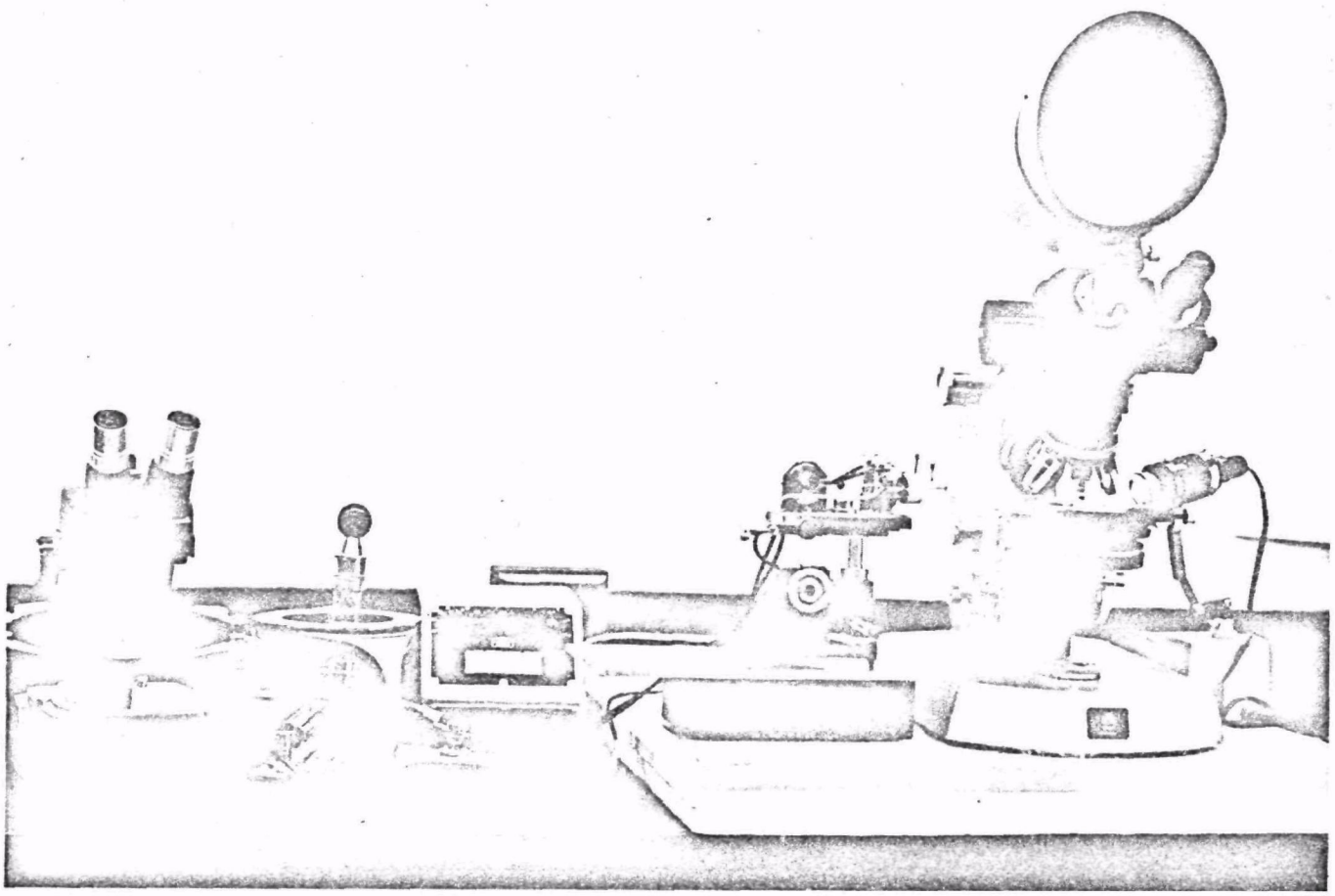

Figure 6a
Microscopy Facility

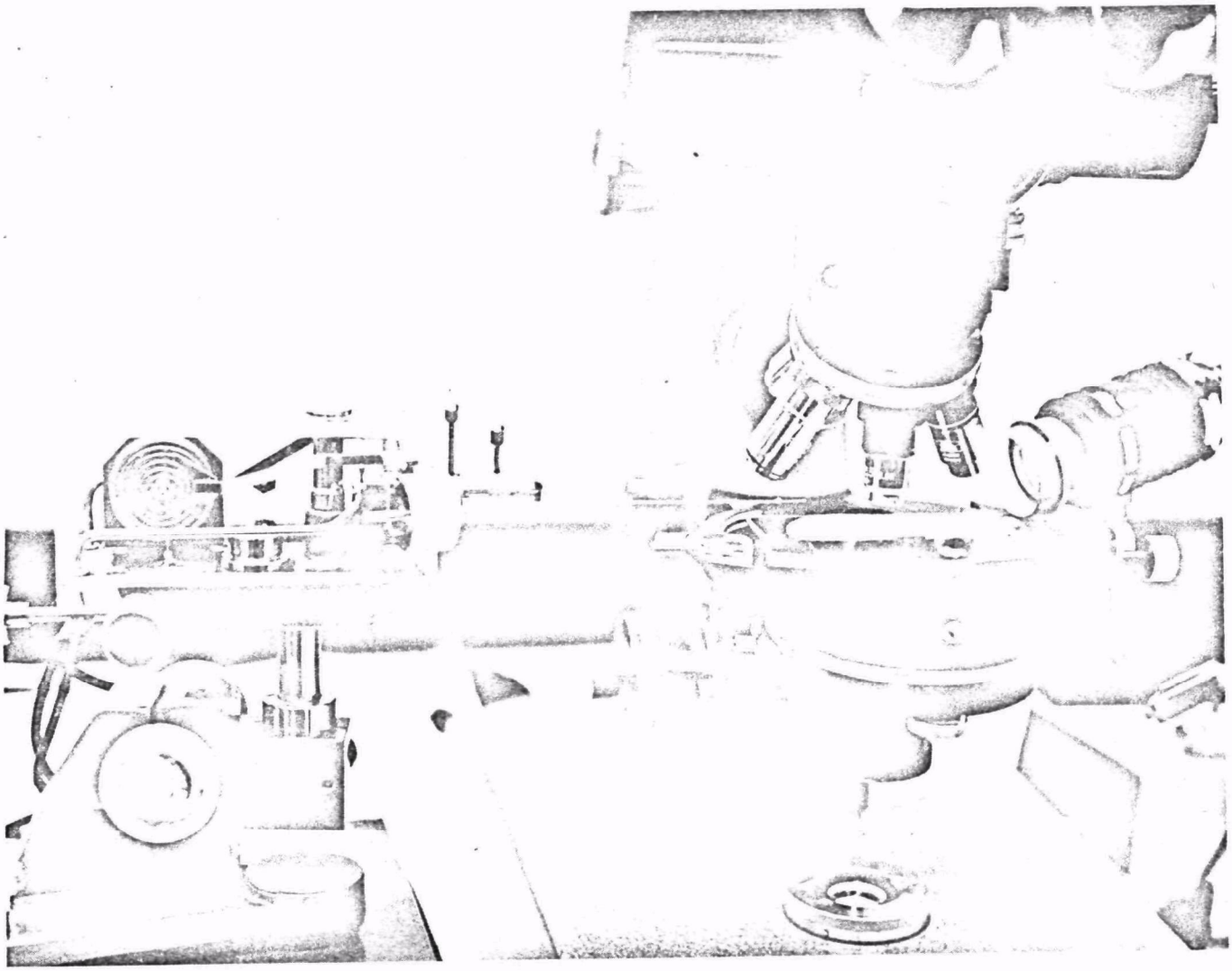

Figure 6b
Microscope and Micromanipulator

Figure 7a
Tooled Particles

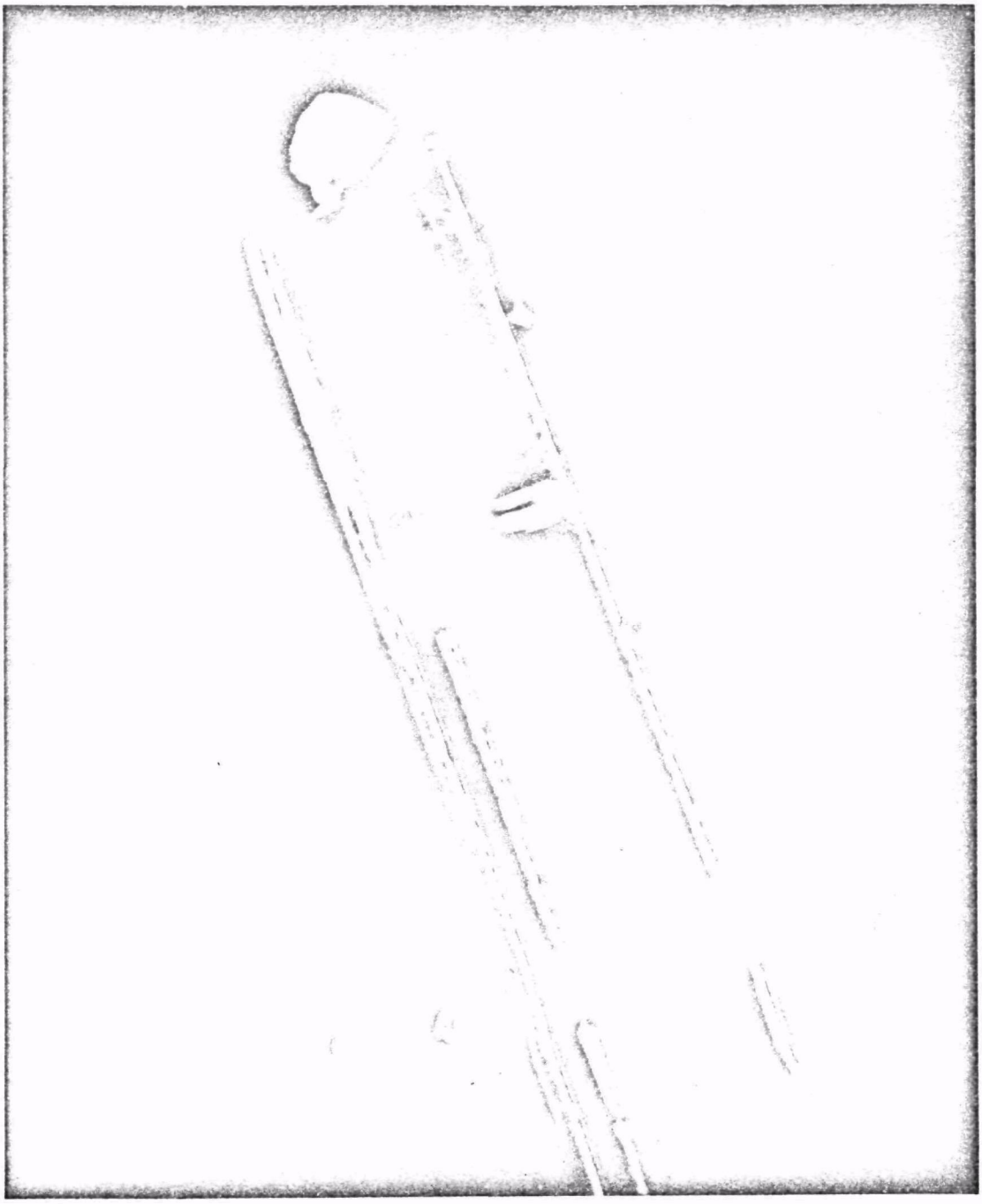

Figure 7b
Tooled Particles

APPENDIX B

PRELIMINARY REPORT ON THE CHEMICAL AND PHYSICAL PROPERTIES OF AIRBORNE PLUTONIUM PARTICLES AT THE NEVADA TEST SITE

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INTRODUCTION

Knowledge of the physical and chemical characteristics of airborne plutonium particles is necessary to accurately assess the biological availability of this radionuclide in the environs of the Atomic Energy Commission's Nevada Test Site (NTS). There has been much speculation as to the chemical forms and size of airborne plutonium at the NTS; however, there appears to be little published data to support these speculations. This preliminary report suggests that there are at least four physical/chemical forms of airborne plutonium, that most of the plutonium particles would reach the alveoli, and that at least one of the physical/chemical forms may become more biologically available with time.

PROCEDURE

Sample Collection:

Microsorban filters with a collection efficiency of 100% for particles greater than three micrometers diameter were used to sample airborne plutonium particles in Area 11 of the Nevada Test Site. The sampling rate of each air sampler was approximately 0.5 m^3 per minute, each sampler was positioned vertically one meter above the ground facing the explosion site of a previous safety shot.

Sample Analysis:

An autoradiograph was made of several microsorban filters by sandwiching it between pieces of x-ray film which were subsequently developed to determine the general location of the alpha-active particles. A 6.5 cm^2 section from a single filter containing alpha-active particles was subsequently cut out and processed in freon in an ultrasonic bath to separate the particles from the filter material. These particles were spread out on cellulose nitrate film, which was exposed for 72 hours and then analyzed by track etch techniques for alpha tracks to quantitate the alpha activity per particle. Thirty-two of these particles were selected at random and sized by optical microscopy: Mass spectrometric and electron microprobe analyses were performed on twelve of the sized particles.

RESULTS

The resultant tracks on the cellulose nitrate film indicated that there were at least 55 alpha-active particles present in the 6.5 cm^2 sample. The results obtained by optical microscopy and alpha track counting are shown in Table I. These results indicate that the observed size range of the particles was <0.5 to 17.0 μm, and that the gross alpha activity ranged from 4 to 260 fCi/particle.

Results of the mass spectrometric and electron microprobe analyses of the particles are shown in Table II. The data reveal the presence of at least four classes of particles. The first class (I) included the relatively small, high-activity particles composed

of plutonium-uranium oxide, which were included in an organic matrix. Previous experience with this type of particle in this laboratory indicated that the organic matrix was extremely fragile and fractured with the slightest manipulation.

The second class (II) included the larger, lower-activity silicate particles in which plutonium was heterogeneously distributed in the particle in percentages too small to detect by electron microprobe.

The third class (III) included the relatively large, low-activity organic particles in which plutonium was also heterogeneously distributed with a concentration of <1%.

The fourth class (IV) includes the relatively small, high-activity oxide particles in which the plutonium was homogenously distributed throughout the particle.

DISCUSSION

Due to the extreme fragility of Class I particles, it may be inferred that various meteorological phenomena such as wind, rain, freezing, and thawing may cause this class of particles to be easily fractured resulting in smaller particles which may be both more respirable and more subject to redistribution with time.

From the results of the 32 particles analyzed, approximately 90% are less than 10 μm diameter. When plotted on log-probability paper the geometric mean diameter appears to be 1.5 μm.

A plot of alpha activity versus size yields a scatter diagram suggesting that plutonium is not homogenously distributed in these air filter samples. Another approach to this apparent discrepancy

is to use the specific activity concept. The particle with the highest activity is 50% Pu (Table II). Because of the large difference in the half-lives of plutonium and uranium, essentially all of the alpha activity is assumed to be due to plutonium.

For particle #805:

Volume = 310 μm^3

Diameter = 8.2 μm

Activity = 261 fCi

Pu-239 Specific Activity = 6.13×10^{-2} Ci/g

Density (assumed) = 11.5 g/cm^3

Calculated Values of Mass and Volume are:

Mass of Pu = $(2.61 \times 10^{-13}$ Ci$)(6.13 \times 10^{-2}$ Ci/g$)^{-1}$ =

4.26×10^{-12} g

Volume = $(4.26 \times 10^{-12}$ g$)(11.5$ g/cm$^3)^{-1}$ =

3.07×10^{-13} cm^3 = 0.307 μm^3

The calculated volume is about 1/1,000 the observed volume suggesting that most of this particle is something other than plutonium.

With a geometric mean diameter of 1.5 μm, the particles should be respirable [1]*. A calculated aerodynamic diameter [2],[3] based on this mean value is

$$D_a = (P)^{\frac{1}{2}} D_g = (\text{density})^{-1} \text{ (geometric diameter)}$$

$$D_a = (11.5)^{\frac{1}{2}}(1.5) = 5 \ \mu m$$

"

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* A 5 μm diameter was set in this report as the upper limit on respirability.

The maximum allowable exposure to soluble ^{239}Pu in air as set by the AEC can be calculated as follows:

Concentration guide (CG) limit [4] =

2×10^{-12} $\mu Ci/cm^3$ = 2 pCi/m^3

Reduced CG = 6×10^{-14} $\mu Ci/cm^3$ = 60 fCi/m^3

Standard week = 168 hours

Work week = 40 hours

Breathing rate, standard week = 140 m^3/week

Breathing rate, work week = 33.3 m^3/week

Average activity of particles (GE data) = 46 fCi/particle

a) limit for ^{239}Pu in pCi/week

$L = (2\ pCi/m^3)(33.3\ m^3/week) = 67$ pCi/week

b) for the average activity of 46 pCi/particle, this limit in number of particles is

$N = (6.7 \times 10^4\ fCi/week)(46\ fCi/particle)^{-1} =$

1457 particles/week

c) reducing the CG by a factor of three for a suitable population sample gives [5]

$L_r = (6 \times 10^{-2}\ pCi/m^3)(33.3\ m^3/week) \simeq 2$ pCi/week

$N_r = (2 \times 10^3\ fCi/week)(46\ fCi/particle)^{-1} = 43$ particles/week

d) the particle inhalation rate at the reduced CG limit is

$R = (43\ particles/week)(33.3\ m^3/week)^{-1} = 1.3\ particles/m^3$

Assuming a retentivity of 50%, the average person would inhale 2 pCi/week and retain 1 pCi/week.

The air sampler used to collect these particles operated for 24 hours at 0.56 m^3/min and, therefore, filtered particles from 806 m^3 of air. Based on 8.5 alpha-active particles per cm^2, the total filter contained about 1,500 particles or nearly 2 particles/m^3. On the basis of the above calculation, this is about equal to the reduced AEC CG.

ACKNOWLEDGMENT

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Table I

Size and Activity of Particulates

Particle Number	Diameter (μm)	Gross Alpha Activity (fCi/particle)
101	<0.5	52
102	<0.5	52
103	0.5	31
104	1.0	23
105	0.5	16
106	1.0	29
107	<0.5	06
108	<0.5	19
109	<0.5	08
110	1.5	52
111	<0.5	12
112	8.8	15
113	8.0	05
114	10.5	78
115	3.5	30
116	1.2	29
118	<0.5	14
119	2.3	78
120	11.5	04

121	1.5	09
122	1.0	16
124	17.0	52
125	<0.5	06
126	<0.5	06
138	2.5	104
802	3.5	52
803	8.5	10
804	3.0	143
805	8.2	261
806	2.5	65
807	3.2	78
808	3.0	21

Table II

Chemical and Physical Forms of Particulates

Particle Number	Composition of Particle Containing Plutonium	Description of Particle	Class
804	48% Pu, 33% U, 19% O_2	<0.5 μm oxide particle in an organic matrix whose diameter is 3.0 μm	I
806	48% Pu, 33% U, 19% O_2	<0.5 μm oxide particle in an organic matrix whose diameter is 2.5 μm	I
805	50% Pu, 31% U, 19% O_2	<0.5 μm oxide particle in an organic matrix whose diameter is 8.2 μm	I
123	14% Si, 5% Al, 38% O_2, 34% Fe	Silicate whose diameter is 6.0 μm	II
803	24% Si, 7% Al, 45% O_2, 9% Fe, 8% K	Silicate whose diameter is 8.5 μm	II
124	31% Si, 4% Al, 47% O_2, 12% Fe, 2% Mg, 2% Ti	Silicate whose diameter is 17.0 μm	II
112	29% Si, 9% Al, 48% O_2, 4% Mg, 3% Fe, 3% K, 1% Ca	Silicate whose diameter is 8.8 μm	II
120	97% C, 2% O_2	Organic whose diameter is 11.5 μm	III
113	95% C, 3% Si, 1% Al	Organic whose diameter is 8.0 μm	III
808	38% Pu, 22% U, 22% O_2, 14% Al, 1% Fe	Oxide whose diameter is 3.0 μm	IV
138	45% Pu, 21% U, 23% O_2, 10% Al	Oxide whose diameter is 2.5 μm	IV
802	3% Pu, 79% U, 17% O_2	Oxide whose diameter is 3.5 μm	IV

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APPENDIX C

LITERATURE SURVEY - Plutonium Particulates

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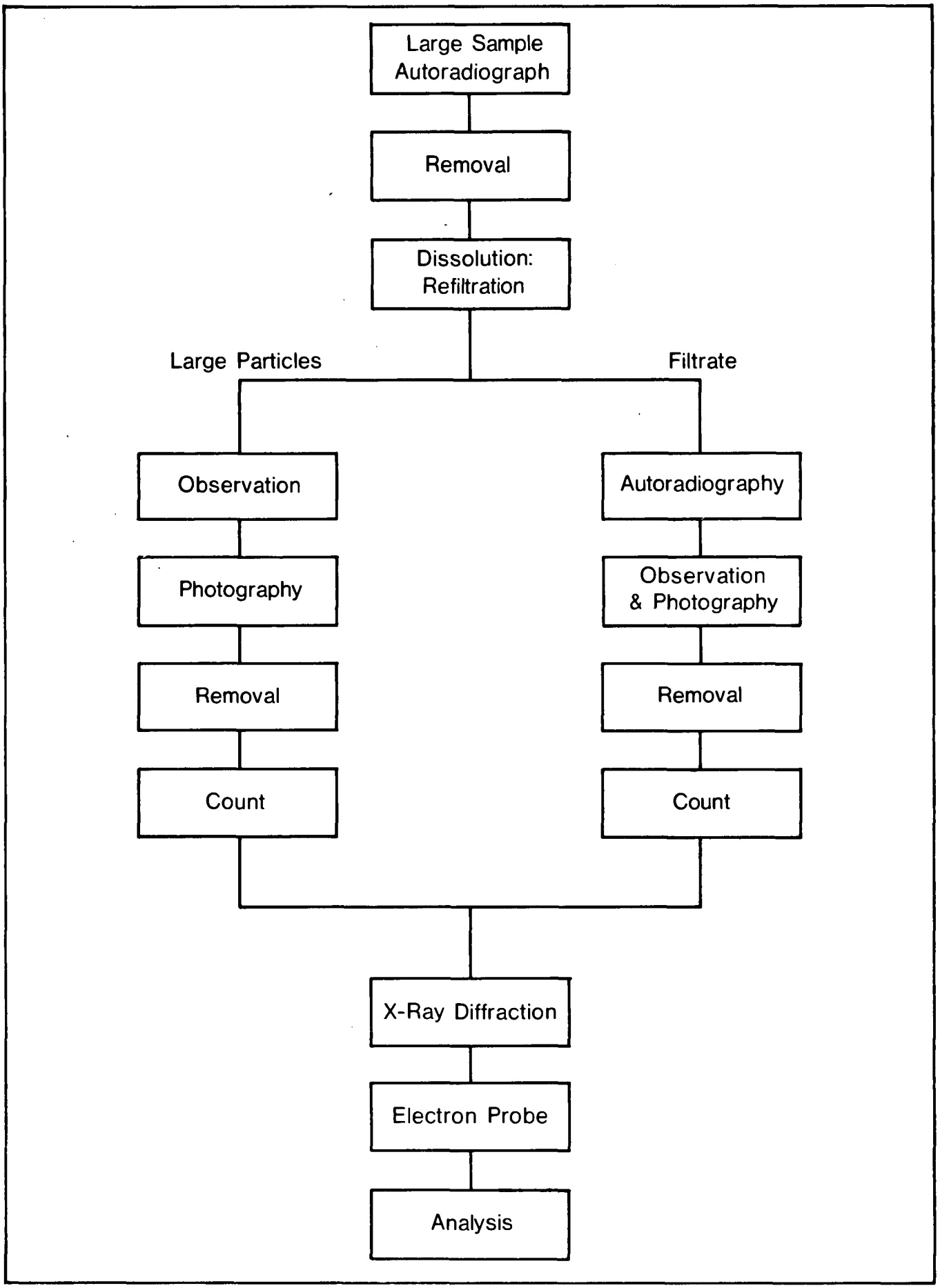

Figure 1
Process Flow Chart

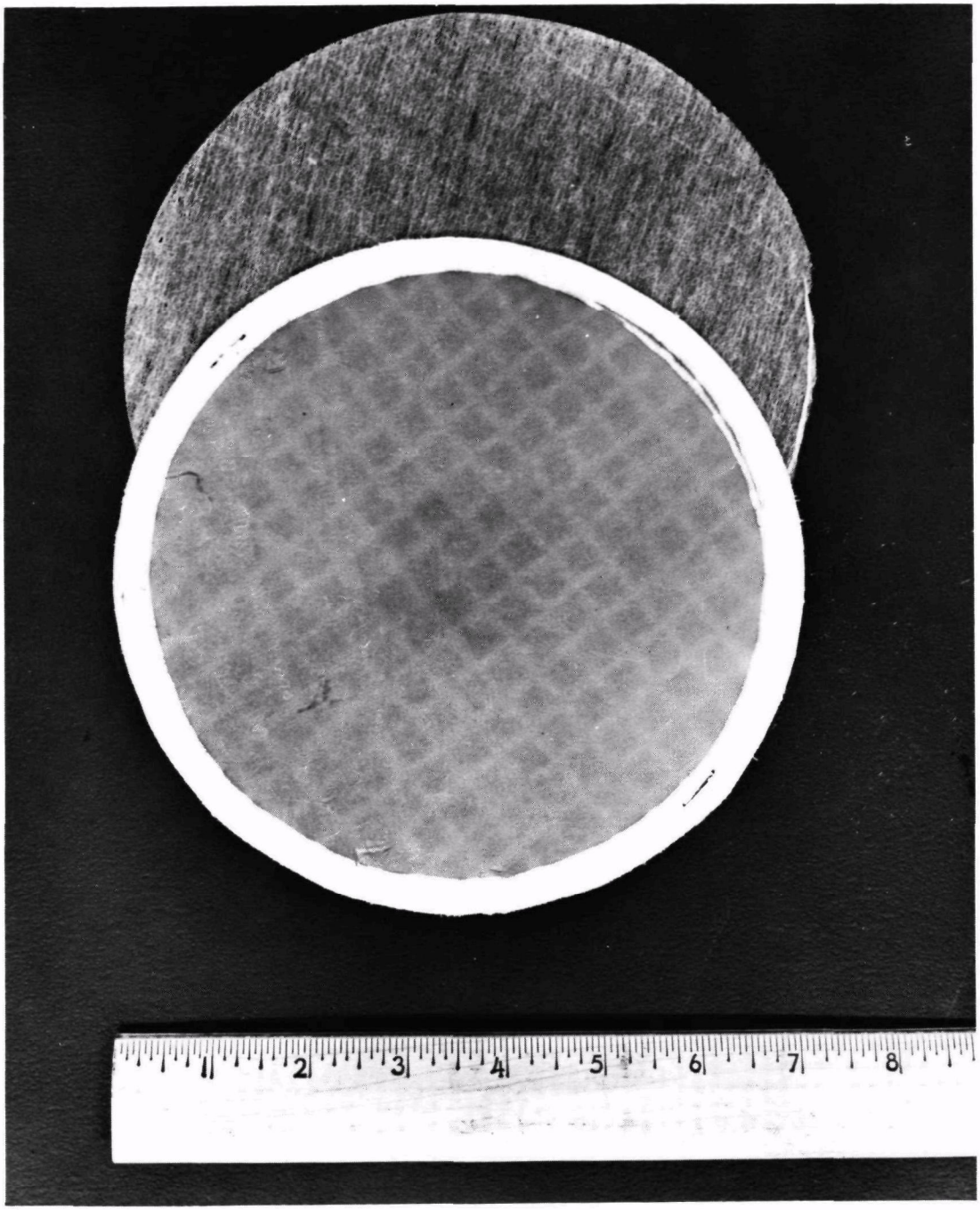

Figure 2
Air Sample Filter

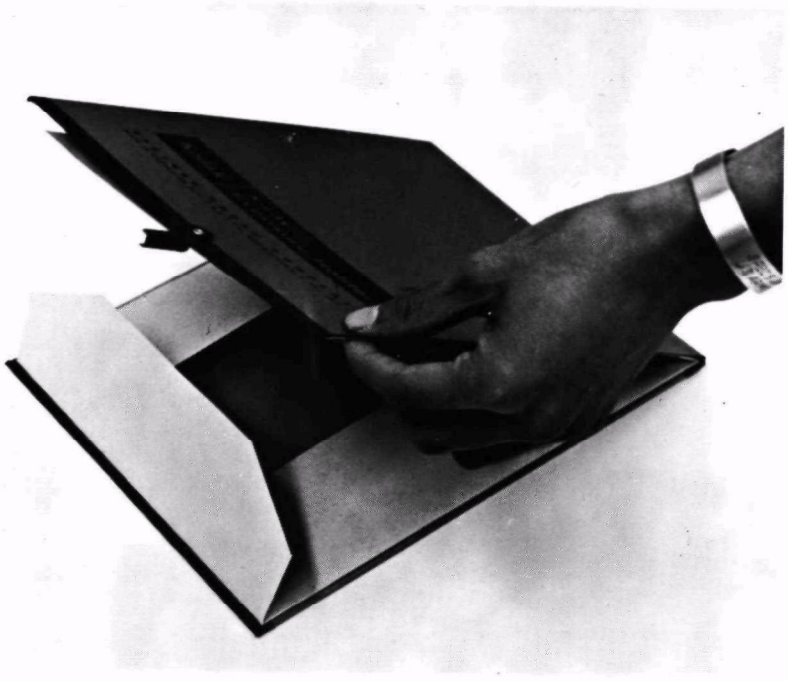

Figure 3
Autoradiograph Sandwich

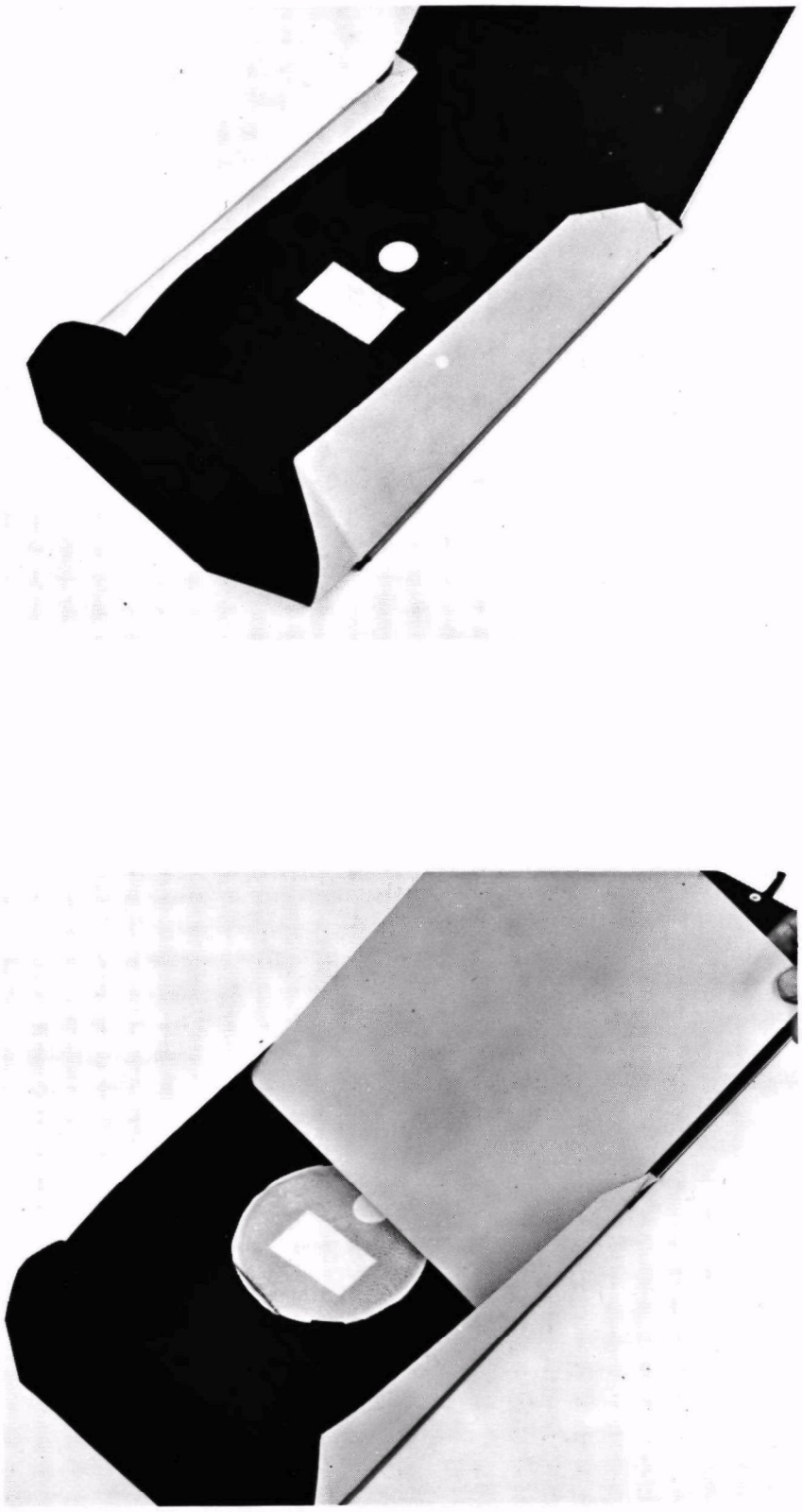

Figure 3
Autoradiograph Sandwich

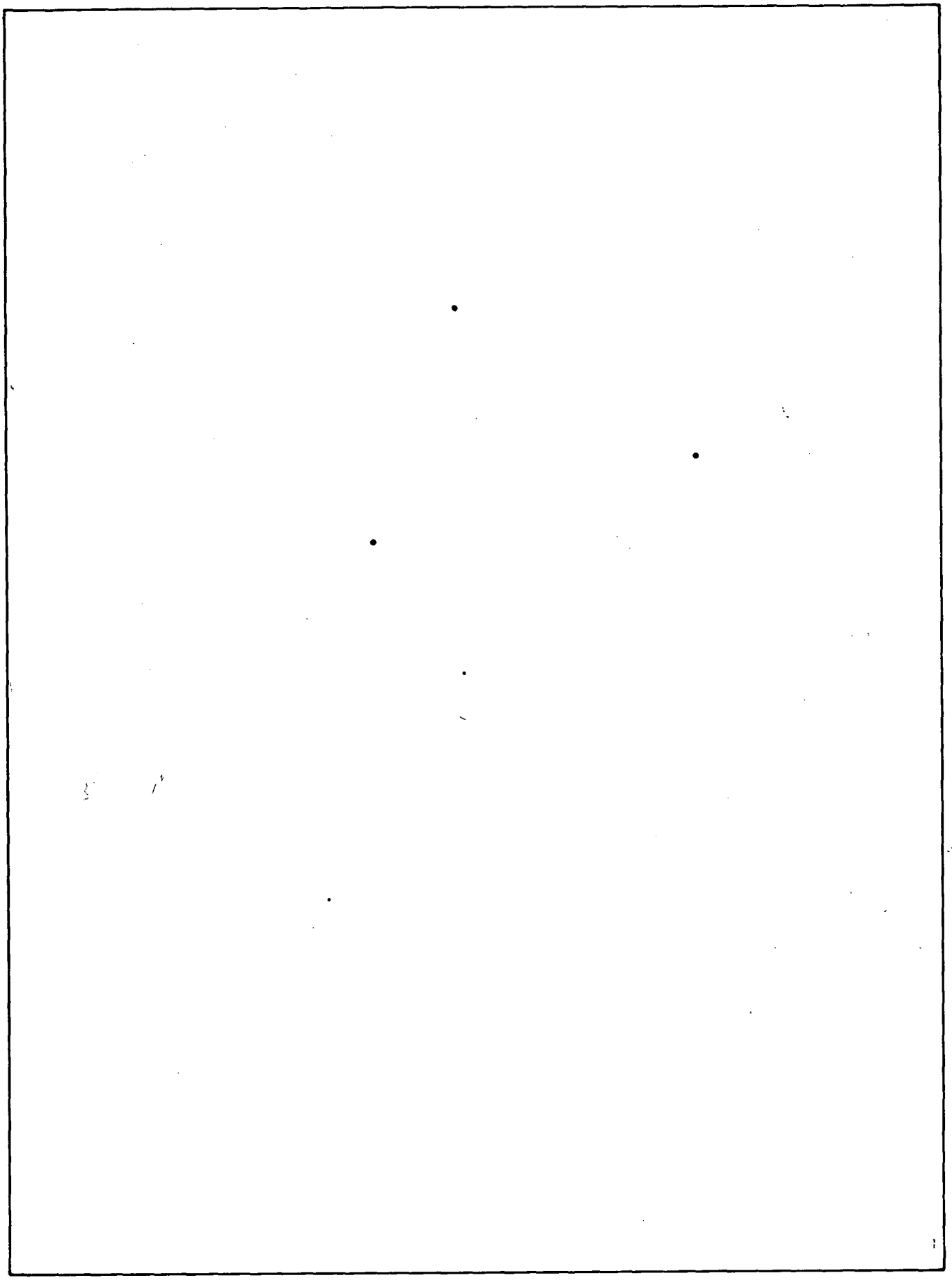

Figure 4a
Autoradiograph of 15 cm Filter

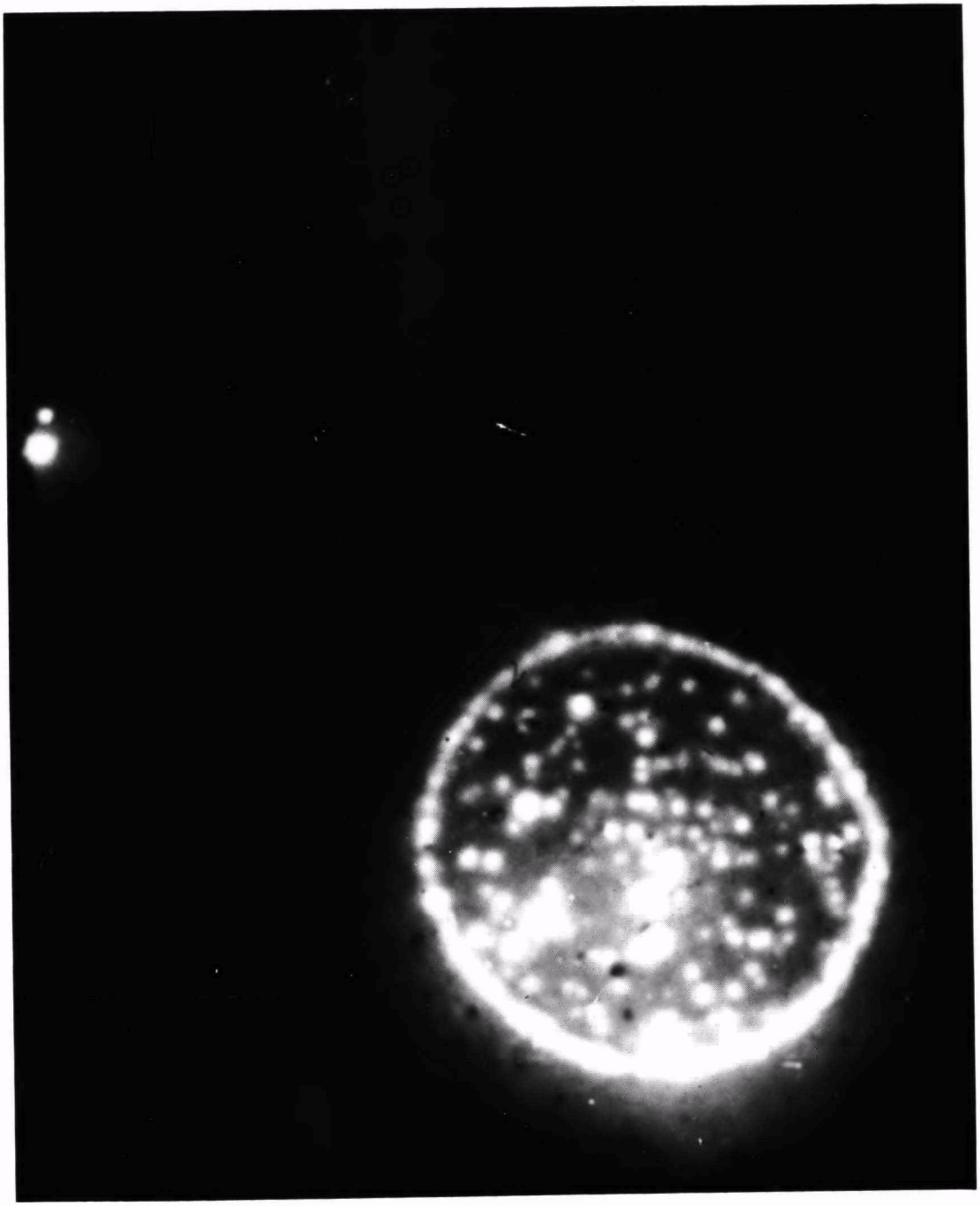

Figure 4b
Autoradiograph of Plutonium Particle in Solution

Figure 5a
Typical Large Particles

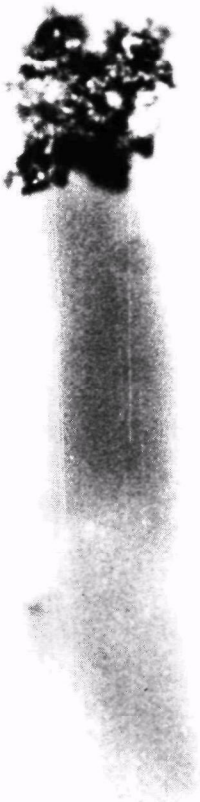

Figure 5b
Large Particle Before Fracture

Figure 5c
Large Particle After Fracture

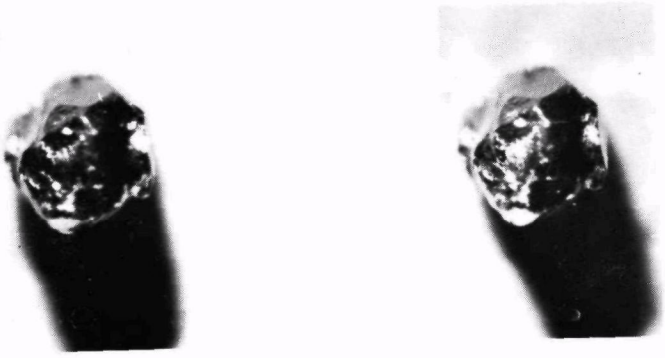

Figure 5d
Stereo Pair; Copper Particle

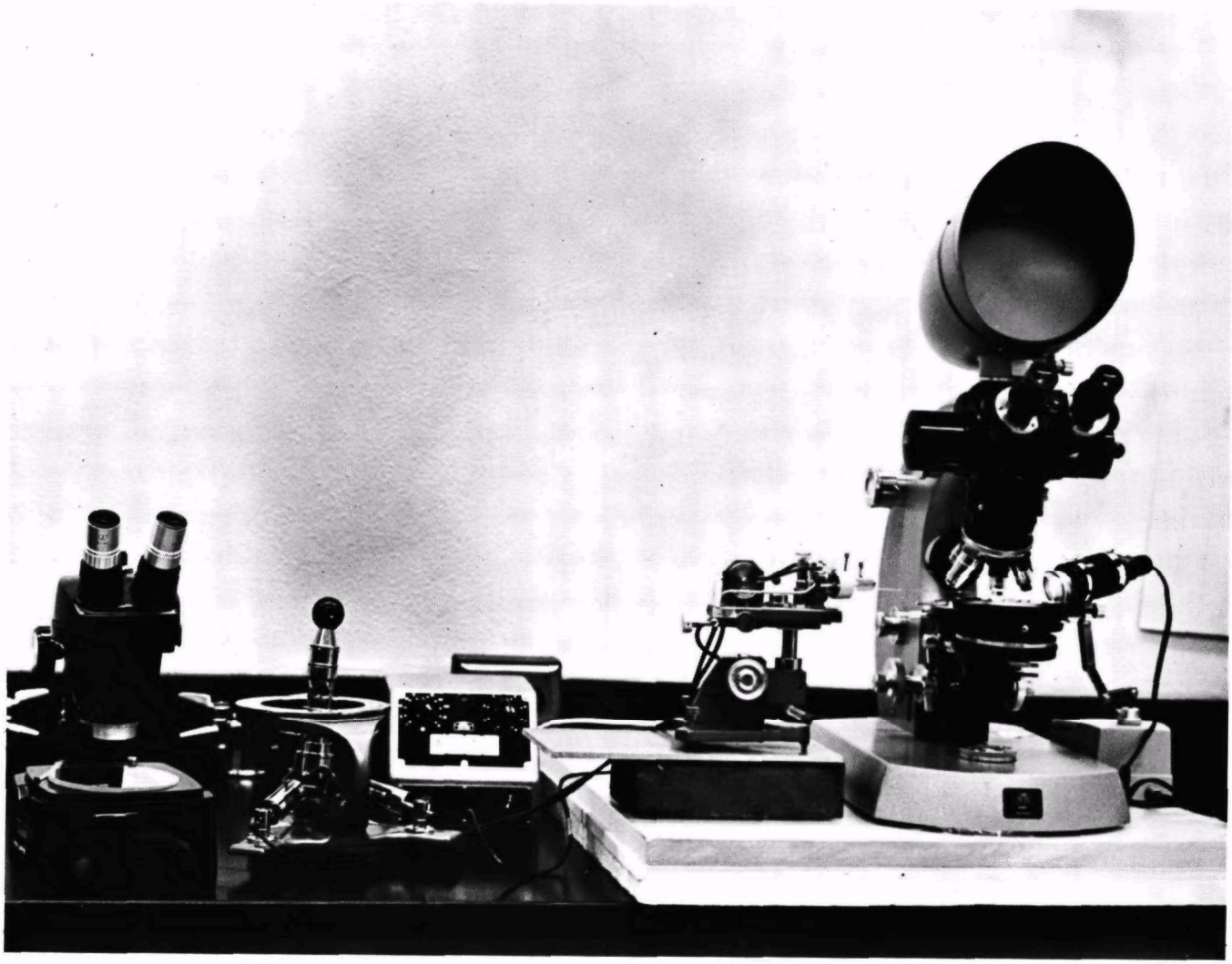

Figure 6a
Microscopy Facility

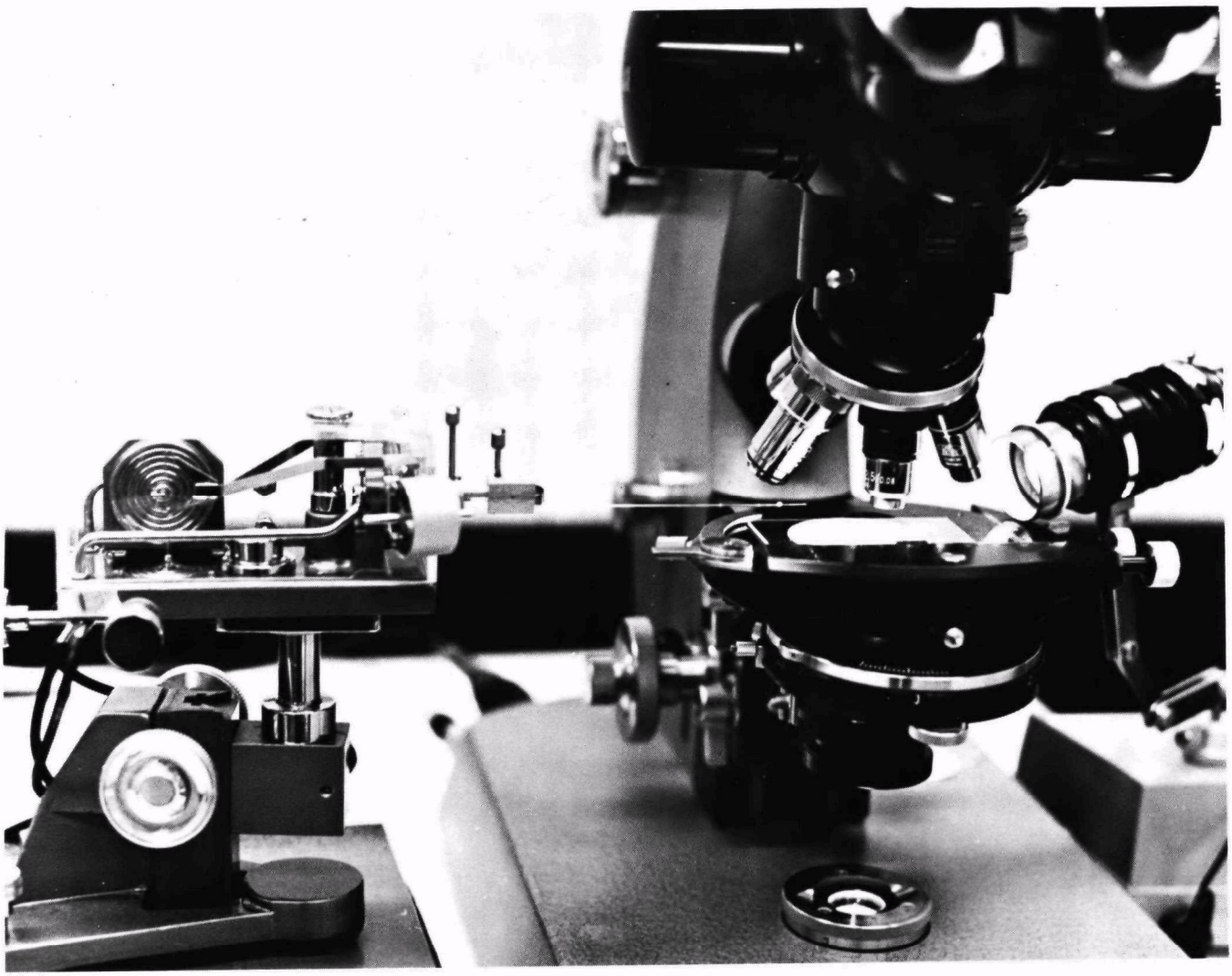

Figure 6b
Microscope and Micromanipulator

Figure 7a
Tooled Particles

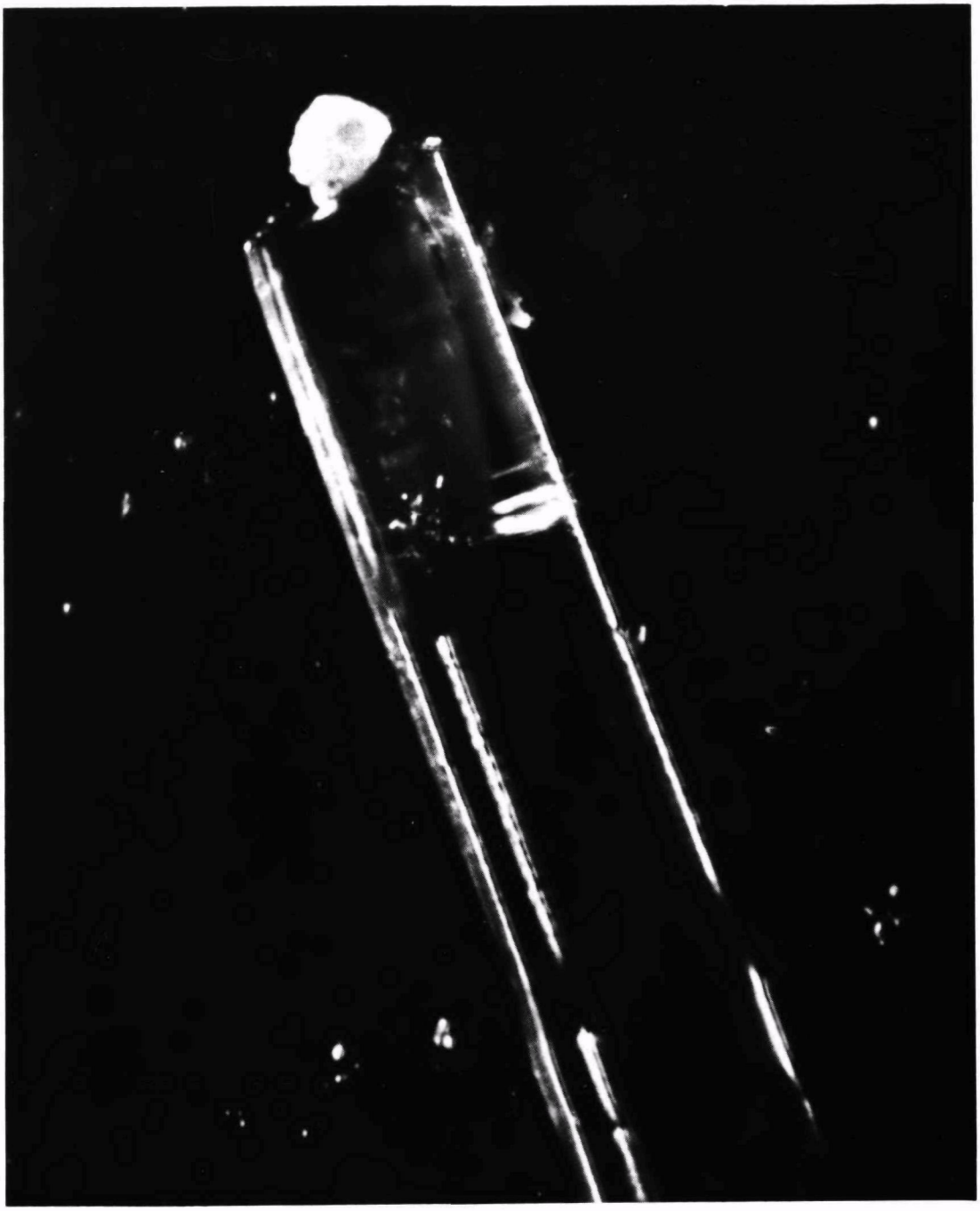

Figure 7b
Tooled Particles