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METHODOLOGY FOR THE ISOLATION AND CHARACTERIZATION OF INDIVIDUAL PLUTONIUM-BEARING PARTICLES IN ATMOSPHERIC EFFLUENTS FROM A NUCLEAR PROCESSING PLANT

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DISCLAIMER

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ABSTRACT

A method for isolating and characterizing individual particles containing fissionable materials is described. Polycarbonate membrane filters are used to collect the particles from atmospheric effluents from a nuclear processing plant. The filters are cast into films composed of a polycarbonate matrix containing the particles. Collected particles containing plutonium or other fissionable materials are identified by fission fragment tracks produced by irradiating the polycarbonate film with thermal neutrons. The nature of the fissionable material is identified by coating the film with nuclear track emulsion and measuring the ratio of alpha-particle tracks in the emulsion to the fission fragment tracks in the polycarbonate film. Single particles

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containing fissionable material are isolated by excising small squares of film containing them. Isolated particles are prepared for sizing and electron microbeam analysis by peeling the emulsion off the film square, placing the square on a beryllium sample-mounting block, and washing the polycarbonate away from the particle.

Results reported earlier show this method can be used to determine the size, ^{239}Pu-content, and elemental composition of micrometer-diameter particles. Tests using this method to analyze particles containing known plutonium and uranium isotope mixtures, show that with it the quantity and isotope composition of fissionable material on a single particle can be determined.

INTRODUCTION

Nuclear fuel processing facilities at the Savannah River Plant release minute (<1 mCi/yr) quantities of ^{239}Pu combined with other material in particulate form to the atmosphere.

A procedure was developed to identify particles containing fissionable material in process exhaust streams so that the nature and quantity of the fissionable material in each particle could be determined and to isolate these particles so that their elemental compositions, sizes, and structures could be determined. Two hundred and ninety-nine particles containing ^{239}Pu were collected from the exhaust of a fuel processing facility and characterized by the method discussed in this paper as part of a continuing program to provide further information concerning the transport and fate of released radioactive particles. The results have been reported previously.[1,2]

This report gives the methodology used in collecting and handling the particles. Tests were made using mock samples prepared from particles containing only low-irradiation plutonium or highly-enriched uranium. Test results showed that radiographic techniques incorporated in the procedure can be used not only to identify particles containing fissionable material, but also to determine the nature and quantity of the fissionable material in each particle.

METHODS AND MATERIALS

Particle Collection

Particles are collected by drawing a fraction of exhaust air through membrane filters. These filters are polycarbonate films with uniform-diameter pores. The pores are almost perfectly round cylinders normal to the surface. Thus the filters approach a two-dimensional screen with surface filtration.

The filters are 47 mm in diameter and 5 μm thick with 0.1-μm diameter pores, $3 \times 10^8/cm^2$, giving a filter porosity of 0.024. The filters are supported in a polycarbonate aerosol holder.* Air is drawn through the holder by a small diaphragm pump with a *Viton*** diaphragm at a rate of four liters per minute to give a face velocity at the filter of 3.8 cm/sec. At this flow, the

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* The aerosol holders and membrane filters were produced by Nuclepore Corporation, Pleasanton, California and obtained from them or Bio-Rad Laboratories, Richmond, California.

** Trademark of E. I. du Pont de Nemours & Company, Inc.

total efficiency for particle collection by the processes of impaction, diffusion, and interception, calculated according to Spurny,[3] is 100% for all particles with diameters of 0.001 μm (the diameter of gas molecules) are larger.

These membranes are particularly suited for the particle studies described here for two reasons: (1) The low ash weight (one-third that of typical cellulose membranes) minimize both induced radioactivity during neutron irradiation and spurious results during electron microprobe analyses. (2) The sensitivity of polycarbonate to fission fragment irradiation makes this membrane useful as an indicator of trapped particles that contain fissionable material.

Arrangement of the air sampling system is shown in Figure 1. As particles accumulate on the membrane filters, membrane porosity and air flow are reduced. To determine the fraction of the exhaust sampled, integrated air flow is measured with a dry type test meter* in series with the diaphragm pump. When nitrogen dioxide is present, exhaust gas is passed through two gas drying towers between the filter and the pump. The first tower contains indicating *Drierite*** to remove moisture from the air and save the *Ascarite*† in the second tower. The self-indicating *Ascarite*, in turn, absorbs nitrogen dioxide to protect the pump and the dry

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* Manufactured by the American Meter Division of Singer.

** Trademark of W. S. Hammond Drierite Company.

† Trademark of Arthur H. Thomas Company.

test meter. A small flowmeter is mounted on the exhaust side of the dry test meter to give an indication of the instantaneous flow rate through the system. Air from the meter is fed back into the exhaust system to prevent its release to the service area.

Film Preparation

Figure 2 shows the procedure for converting the particle-containing filter membrane to a polycarbonate film. After air is sampled, the radioactivity retained on each filter is measured before it is handled in the laboratory. Each filter is then dissolved in a 40% (v/v) solution of 1,2-dichloroethane in dichloromethane. The filters are folded and each placed in a 1-mL volumetric flask. A second clean, unused filter is placed in the same flask to give sufficient polycarbonate to form a 50-mm square film. Volume of the dichlorethane solution in the flask is adjusted to about 3/4 mL. This mixture is stirred until the polycarbonate filters dissolve. The flasks are stoppered and allowed to stand for 30 minutes to allow trapped air bubbles to rise to the surface.

The clear polycarbonate solution containing the particles is poured onto a clean, 50-mm (two-inch) square glass plate (see Figure 2). One edge of a second 50-mm square glass plate is used to spread the solution evenly over the surface of the first plate. The solution is stirred continuously with the second plate for about half a minute while the solution thickens. A 50-mm square, 1.6-mm thick acrylic support with a 45-mm diameter hole is placed

on top of the wet film. The support and plate combinations are placed in covered petri dishes for 16 hours while the films continue to dry. If the film is allowed to dry too rapidly in a moist atmosphere, water will condense on the newly forming surface to produce a frosted appearance which will interfere with microscopic examination of the particles.

After the films have dried for 16 hours, the glass plates are removed by dipping the support and plate combinations in distilled water and prying the supports from the glass with tweezers. (The glass pellets may be removed soon after affixing the acrylic supports, however the films will split after a few hours if not supported by the glass until thoroughly dry.) The supports are then allowed to dry overnight in a dust-free atmosphere.

Film Irradiation

To produce fission fragment tracks in the polycarbonate film by which particles containing fissionable material can be identified, the cast film is irradiated in a thermal neutron fluence of about 9×10^{14} neutrons per cm^2. Films are arranged for irradiation by stacking the supports on top of each other and sandwiching each film between two supports. Included in the stack are blank films that are prepared in the same way as the sample films from clean unused filters. The assembled stack is wrapped with cellophane tape. Wrapped with each stack are preweighed 25.4-mm diameter, 0.25-mm thick, Type 302 stainless steel disks. The induced radioactivity from 27-day ^{51}Cr in these disks is later

measured to determine the thermal neutron fluence to which the particles are exposed.

The packaged stacks are irradiated in a three-inch diameter hole in a light water-cooled, enriched uranium-fueled standard pile with graphite reflectors.[4] Following irradiation, the induced radioactivity of the stacks are allowed to decay several days before the packaged stacks are returned to the laboratory.

Film Etching

Fissionable material contents and isotopic compositions of particles on the irradiated file are determined by the procedure shown in Figure 3. Figure 3a shows the thermal neutron irradiation of a particle causing the fission of a nucleus and the production of two fission-fragment tracks in the polycarbonate matrix. These tracks can be seen only with difficulty using an electron microscope. To make the tracks visible with an optical microscope, the polycarbonate film is etched for fifteen minutes in 6N NaOH at 52 to 55°C. During this etching process, a portion of all polycarbonate surfaces is dissolved: the outer surface of the cast film, the surface around the particle, and especially that along the fission-fragment tracks, as shown in Figure 3b.

Emulsion Coating

To identify the fissionable material in each particle, the alpha particle emission rate is measured by autoradiographic techniques, i.e. coating the polycarbonate film with a photographic

emulsion (Figure 3c) which is developed after a predetermined exposure time. Alpha particle tracks are produced in the emulsion as microscopic rows of silver grains. Tracks are produced wherever there is emulsion, not only in the emulsion coating the surface of the polycarbonate film, but also that filling the fission-fragment tracks and surrounding the particle (Figure 3d).

The emulsion used to coat irradiated films is Kodak Type NTB nuclear-track emulsion (Kodak catalog number 165 4425). Under darkroom lighting (No. 2 Wratten filtered), a 4-oz jar of emulsion is partly immersed in a water bath maintained at 40°C until the emulsion melts (between 15 and 20 minutes). Slightly over half the molten emulsion is carefully poured into an elliptical, black, polyethylene cylinder in the water bath. The molten emulsion is tested by dipping a clean glass microscope slide into it and examining the coat on the glass under a safelight to determine whether bubbles are present. If present, they are scooped from the surface of the molten emulsion with a porcelain spoon.

The acrylic supports with polycarbonate films are coated with emulsion by holding the supports vertically by one corner and dipping them into the clear molten emulsion for about one second. The films are kept vertical until the excess emulsion has drained off. The coated films are then placed horizontally in a biochemical oxygen demand (BOD) incubator maintained at 28°C and about 80% relative humidity until the emulsion cools and gels (about 30 minutes).

Exposure

To determine the alpha particle emission rate for each aerosol particle, the polycarbonate films are stored for a determined period of time before being developed. Spun aluminum *Desicoolers** containing 60 grams of indicating *Drierite* are used to contain the films during this exposure of the emulsion to the particles. The *Desicoolers* are sealed with black adhesive tape and stored in a refrigerator between 4 and 5°C for the duration of the exposure, usually about one week.

Emulsion Processing

At the end of the exposure period, the alpha-particle tracks in the emulsion are developed and all substances other than tracks are removed from the emulsion. The emulsion is developed in a 1:1 solution of *Dektol*** developer for three minutes at 17°C.[5,6,7] This developer is chosen because it remains unusually free from muddiness, sludge, precipitation, and discoloration throughout the normal solution life. It also has a high capacity and uniform development rate.

Immediately following development, the film is rinsed in 28% (v/v) acetic acid for 10 seconds. The high acid concentration is used to prevent reticulation of the emulsion and its separation from the supporting polycarbonate film.

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* Trademark of Fisher Scientific Company.

** Trademark of Eastman Kodak Company.

The rinsed emulsion is fixed by placing it for five minutes in a 1:3 dilution of Kodak rapid fixer concentrate* containing 2.8% (v/v) hardener concentrate. This fix solution dissolves the undeveloped silver halides, forming salts with them that are not only soluble in the fixing bath but are also stable when diluted during washing. The rapid fixer contains ammonium thiosulfate, rather than the usual hypo (sodium thiosulfate), as a fixing agent, because ammonium thiosulfate complexes the silver more rapidly and has a greater capacity.

After silver halides are dissolved, all chemicals except the metalic silver are washed from the emulsion using a batch process. This removes the ammonium thiosulfate, which will react with (1) the metallic silver of the alpha particle tracks, causing them to slowly fade, (2) the silver thiosulfate complex, which will decompose to silver sulfide, and (3) all extraneous elements which, if left in the vicinity of the particles, will affect electron microprobe analysis. The emulsion-covered film, after washing, is placed in distilled water, and the chemicals in the emulsion and wash water are allowed to approach equilibrium for two minutes. The emulsion is then placed in a second container of distilled water while the water in the first container is changed. This process is repeated a total of eight times. After the water wash, the emulsion-coated polycarbonate films are placed in racks and allowed to dry in a dust-free atmosphere.

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* Kodak Photographic Products catalog numbers 146 4106 or 146 4114.

The entire process is carried out in a dark room where the temperature is maintained between 17 and 18°C. All solutions and the wash water are stored in the dark room so there will be no temperature gradient between solutions during processing.

Track Counting

Both the fission fragment tracks in the polycarbonate film and alpha particle tracks in the emulsion coating are counted by direct observation under a Zeiss photomicroscope.

The film is prepared for track counting by placing the acrylic support on a 50-mm square, 1.6-mm thick acrylic block. This backing for the film is necessary for, during microscopic observation, the slight movement of air caused by breathing would move the thin, unsupported polycarbonate film out of the focal plane of the objective. The block also serves to protect the film from damage by the microscope objective and to support it during marking and cutting operations.

Those particles having tracks are located under a Bausch and Lomb stereo zoom microscope using transmitted light and a magnification of 105X. When found, each particle with tracks is circled with a felt-tip marking pen containing water-resistant, quick-drying ink. After a particle has been marked, the support and block holding the film are moved to the photomicroscope where the fission fragment and alpha particle tracks are counted using transmitted light and a magnification of 1000X. Epiplan, flat-field objectives are used because they are corrected for uncovered

specimens and do not require cover glasses. The numbers of alpha particle tracks in the lower and upper emulsions are added to give the total number of alpha particle tracks observed.

Particles and tracks are photographed using both Polaroid 4- × 5-inch Land type 57 black and white film with a magnification of 869X and Kodak 35-mm, high-speed, color-reversal, *Ektachrome** type EHB film with a magnification of 256X. Three Polaroid pictures of tracks from a single particle — one with the focal plane in the lower emulsion, one in the polycarbonate film, and one in the upper emulsion — are given in Table 4.

Identification of Fissionable Materials

Table 1 gives the theoretical ratios of alpha-particle to fission-fragment tracks which would be produced from particles irradiated with a fluence of 8.64×10^{14} thermal neutrons/cm^2 when there is a seven-day interval between film casting and etching and during exposure to nuclear-track emulsion. (A description of the calculation of these ratios is contained in the Appendix.) The stipulation that etching follow film casting by seven days is included because spontaneous fissions will add to the number of fission fragment tracks during this period.

This identification procedure can be used to distinguish particle-bound plutonium from uranium. Table 1 shows that, of the six isotopic mixtures of uranium, only the highly enriched

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* Trademark of Eastman Kodak Company.

uranium mixture will give a number of fission fragment tracks comparable to that of the plutonium mixtures. Even if there should be enough uranium to produce fission fragment tracks, mixtures of these isotopes would not produce alpha particle tracks. The highly enriched uranium mixture will give an average of only one alpha particle track with every 1372 fission fragment tracks.

This procedure may be used not only to identify plutonium, but also to identify the plutonium isotopic composition in a particle. For example, a particle having 10 fission fragment tracks would also have 5 alpha particle tracks if the mixture were low-irradiation plutonium, 640 alpha particle tracks if it were high-irradiation plutonium, and 5080 alpha particle tracks if it were heat source plutonium. A ratio as high as 5080 would appear to emit only alpha particle tracks unless the track count were very high.

Table 1 includes (in addition to U and Pu track data) a number of curium and californium nuclides which could mimic the plutonium mixtures. The figures contained in this table also indicate that spontaneous fission should always be considered if nuclides other than those of plutonium and uranium are suspected. In this case, the polycarbonate film should be allowed to stand several weeks after casting and then be etched both before and after thermal neutron irradiation. Under these conditions, tracks due to spontaneous fissioning will appear in unirradiated films.

Measurement of Plutonium and Uranium Ratios

To demonstrate the effectiveness of this identification method in distinguishing between plutonium and uranium, samples of particles were obtained from two sources of known nuclide mixtures: one of low-irradiation plutonium and one of highly enriched uranium. Polycarbonate films were prepared containing particles from either one or the other source. The films were irradiated and coated with emulsion, emulsion was exposed and developed using this procedure. The number of alpha particle and fission fragment tracks with each particle were counted.

The data from 315 particles containing low-irradiation plutonium are given in Table 2 and those from 350 particles containing highly enriched uranium are given in Table 3. The data were ranked according to the number of observed fission fragment tracks per particle to determine whether the number of tracks influenced the measured ratios. The mean and standard deviation of the ratios in each track internal are also given.

From Tables 2 and 3 the mean ratio (alpha to fission fragment tracks) for low-irradiation plutonium is 9.1×10^{-1} while that for highly enriched uranium is 1.8×10^{-3}. Thus, ^{239}Pu can clearly be distinguished from ^{235}U using this procedure if there is a sufficient number of tracks. However, these ratios are 1.7 and 2.5 times the theoretical ratios given in Table 1. In the case of highly enriched uranium, all alpha particle tracks were observed as single tracks only, some of which may have been due to background

radiation; this would explain the higher mean ratio for uranium. With plutonium, the higher observed ratios are probably due to the geometry of the media in which the tracks are formed. The polycarbonate film in which the particles are embedded is thin enough for alpha particle tracks to be recorded in the emulsion on both sides. Thus, some of the fission fragments emitted in the vertical direction produce short tracks or no tracks. Likewise, some of the alpha particles emitted in the horizontal direction do not reach the emulsion and give no tracks. If the thickness of the polycarbonate film is less than $2R_\alpha R_f/(R_\alpha + R_f) \approx 2R_\alpha/3$, where R_α is the range of the alpha particles and R_f is the range of the fission fragments, then the observed ratio will be higher than the theoretical ratio.

Data in Table 2 are also plotted in Figure 6 where the circles represent the mean alpha particle to fission fragment ratios in each track interval; the bars, the standard deviation; and the numbers above the bars, the number of particles observed. In this figure there appears to be some influence of the number of tracks counted on the observed ratio. This apparent influence is probably due to the increasing difficulty in counting fission fragment tracks with increasing numbers.

Quantitative Radiographic Analysis

Alpha particle and fission fragment track counts will provide not only a ratio from which the fissionable material carried on the particles can be identified, but also an estimate of the

quantity of the radioactive nuclides present. One femtocurie (fCi) of ^{239}Pu will produce about 22 alpha particles in a week and, when irradiated with a fluence of 8.64×10^{14} thermal neutrons/cm^2, will produce about 40 fission fragments. In a mixture of low-irradiation plutonium, the number of fission fragments produced will be increased to 53 with between 28 and 33 alpha particles depending on the age of the mixture. Only about half of these particles will produce tracks, yet this radiographic technique is much more sensitive than electron microprobe analysis, which is not sensitive to less than 10 fCi[1].

Particle Isolation

After a particle has been identified and photographed and the tracks are counted, the particle is excised from the film in a polycarbonate square. For this, the support and block holding the film are returned to the stereo microscope. In transmitted illumination and at a magnification of 105X, two parallel cuts are made through the emulsion-coated film on either side of the particle using an ultra microlance. The film is then rotated through 90° and two more cuts made forming a square (see Figure 5a). The cut square is then probed in one corner by a 15-mm long, electrolytically sharpened, tungsten needle (made by placing a pair of 0.52-mm diameter tungsten wires in a 3N NaOH solution and applying a 60-Hz, 10-volt potential between them for 10 to 15 minutes). With this needle, the cut square containing the particle is lifted from the film and placed on a glass microscope slide

(see Figure 5b, 5c, and 5d). The polycarbonate square is freed from the needle by rotating it so the corner of the square opposite that stuck by the needle strikes the slide causing the square to rotate and fall.

The emulsion layers are then removed from the polycarbonate square by placing a cover glass on top of the square. (For this operation, the magnification is reduced to between 15X and 70X.) Water is introduced between the cover glass and slide using a glass microbrush made from a 20-μL glass disposable pipet. A very fine tungsten wire is doubled and threaded through the lumen forming a loop at one end. A small amount of quartz wool is placed through this loop, which is then drawn into the end of the pipet. The glass fibers are then cut off about 2 mm from the end of the pipet.

The microbrush is dipped in water until the water rises to the desired level in the lumen. The glass fibers are then touched to the edge of the cover glass to allow the water to flow from the brush to between the slide and the cover glass.

The emulsion is then removed by gently moving the cover glass a few mm from side to side (see Figure 5e); this rolls the swollen emulsion off the polycarbonate film. The emulsion is removed from the surface of the film, but not from the fission fragment tracks themselves, (see Figure 3e and 3f). The cover glass is carefully lifted from the glass microscope slide, taking care not to loose the polycarbonate square containing the particle.

Particle Mounting

To mount a particle, the polycarbonate square is placed in a selected grid location on a beryllium sample mounting block* (Figure 5f). These sample mounting blocks are 25 mm in diameter and 13 mm thick and fit the standard electron microprobe sample holders, which grip the sides and provide the necessary electrical contact. The top surface of the block is highly polished and contains a grid network of 1-mm squares inscribed on the surface. The squares are numbered in mirror image fashion both vertically and horizontally through the center.

With coaxial (reflected light) illumination and 15X magnification under a stereo microscope, the polycarbonate squares are moved from the microscope slide to the beryllium block as previously done, using an electrolytically sharpened, tungsten needle. The polycarbonate square is then dissolved and washed back from the particle using dichloroethane, leaving the particle usually connected to the main body of polycarbonate by a thin isthmus of plastic. This connection does not seriously effect the microprobe analysis and aids in later locating the particles and holding them on the beryllium block. A second glass microbrush is rinsed in dichloroethane to remove any foreign material and filled by immersing the bristled end in a second beaker of dichloroethane. Under a stereo microscope with coaxial illumination

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* Walter C. McCrone Associates, Inc. catalog number XIII-403-3.

and 105X magnification, dichloroethane from the brush is dispensed on the beryllium block just in front of the polycarbonate square until the square is engulfed in the solution. The microbrush is then used to push the solution back from the particle. Gelatin replicas of the fission fragment tracks remained with the particles. Some alpha particle tracks from the particles intersect with the fission fragment tracks causing a few silver halide crystals in the gelatin replicas to develop. These small silver grains remain with the particles to cause spurious silver analysis on the electron microprobe.

The beryllium block is returned to the photomicroscope where, using reflected light, a second Polaroid picture of each particle is made at a magnification of 556X to identify the particles after the gelatin has been removed.

The gelatin with each particle is oxidized by exposure to an oxygen plasma for three hours in a low-temperature asher.* In this asher a gas plasma is generated in oxygen using the energy of electrons in the gas. Power is supplied to electrons at 13.56 MHz by a radio-frequency (RF) generator. Normal ashing techniques require thermal energy to break down molecular oxygen to the atomic form, which then initiates oxidation. Since the energy to do this with a low-temperature asher is provided through the electrons instead of heat energy, the gelatin can be oxidized with atomic oxygen without the usual requirement of heat. The

* Manufactured by International Plasma Corporation.

only heat generated is that resulting from the exothermic oxidation reaction itself. Consequently, high temperature degradation, volatilization, or fusion of the inorganic constituents of the particles are eliminated. (While the gelatin oxidizes readily under these conditions, the polycarbonate is self-extinguishing and does not burn.)

The beryllium sample blocks, each containing 40 to 60 particles with the polycarbonate displaced to one side, are placed in a quartz reactor chamber. The chamber is sealed and evacuated to a pressure of about 0.5 torr with a two-stage, high-vacuum, air pump* having a free air pumping capacity of 500 liters per minute. (*Versilube*** F-50, oxygen compatible lubricant is used instead of normal vacuum pump oil and silicone rubber tubing is used for all flexible connections due to the high oxygen concentration.) The RF generator is turned on and the impedance matching network controls adjusted until a plasma is ignited and sustained. Oxygen is bled into the chambers allowing the pressure to slowly increase to about 5 torr. The gelatin is oxidized for 3 hr with a sustained plasma at a pressure of 5 torr and 450 watts power.

Figure 7 illustrates the last three stages in the preparation of one particle. The top picture is the particle in the polycarbonate film with emulsion stripped off. The middle picture is the same

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* Welch series 1397 Duo-Seal pump.

** Trademark of General Electric for silicone fluid with a viscosity of 50 centistokes at 100°F.

particle with the polycarbonate removed showing the gelatin replicas of the fission-fragment tracks. The bottom picture is a scanning electron micrograph of the particle after oxidation of the gelatin. In this picture, traces of the gelatin replicas and silver grains can be seen. Here, what had appeared to be a single particle is actually a conglomeration of at least five and possibly ten smaller particles.

Particle Sizing

To maintain control of particles after the gelatin track replicas are oxidized, the beryllium sample block is returned to the photomicroscope where each particle is located and photographed again under reflected light using Polaroid film and a magnification of 556X. Occasionally, reference is made to previous photographs to establish the exact location of a particle. As each Polaroid film is developed, it was marked with the particle identification number, location on the block, and magnification used. An arrow is marked on the film pointing to the particle, so there will be no mistake in what is intended for analysis.

The size of each particle is estimated from its third Polaroid picture. Eyepiece graticules are not used because photographs with known magnifications were available for each particle. An average of the smallest and largest dimensions of the photographed particle are measured in μm and divided by the magnification. The dimensions are not measured until after oxidation when the completely denuded particles can be photographed.

Elemental Analysis

To determine elemental composition of the particles, the particles are analyzed on a Cameca MS46 electron microprobe, equipped with four crystal, wave-length-dispersive spectrometers (take-off angle, 18°) and an *EDAX 701/MICROEDIT** energy-dispersive analyzer. X-ray intensities resulting from the electron bombardment of the particles and particle sizes and shapes are estimated. These estimates, along with estimated average densities, are used in the FRAME program[8] as modified for particles work by Armstrong[9] on a *UNIVAC*** 1110 computer. This calculation gives the particle composition in both element and oxide weight percents and atomic proportions based on 24 for oxygen atoms.

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* Trademark of EDAX International, Inc.

** Trademark of Sperry Rand Corporation.

APPENDIX

Track Ratio Calculations

To evaluate the possibility of determining the nature of fissionable material in particles by radiographic methods, the theoretical ratio of alpha particle to fission fragment tracks, which would be produced by a number of nuclides and nuclide combinations, were calculated for a specified set of conditions encountered in routine laboratory procedures.

Twenty-four nuclides were selected on the basis of their thermal neutron cross sections, their half-lives for spontaneous fission, and their presence in typical nuclide mixtures. These nuclides are listed in Table 4 along with their 2200 m/sec thermal neutron cross sections in 10^{-24} cm^2 from Benjamin[10] and Stehn, et al.[11] and their percent alpha decay and half-lives for alpha decay and spontaneous fission in days from Lederer.[12]

Uranium and plutonium are rarely found as single nuclides. Consequently, defined nuclide mixtures of the elements were used. These include six different mixtures of four uranium isotopes as defined by Blumkin and Von Halle[13]: natural uranium and depleted uranium from separation cascade tails containing over 99% ^{238}U but with 0.72% and 0.25% ^{235}U respectively; highly enriched uranium containing 90% ^{235}U; and power reactor fuel, low-burnup uranium from power reactors as the Yankee Reactor, and high-burnup uranium from power reactors as the Fort Calhoun Reactor with 4.0%, 2.5%, and 0.9% ^{235}U respectively. Table 5 gives the

percentages of each nuclide used in the different mixtures. The minor uranium isotope (^{234}U and ^{236}U) concentrations relative to the ^{235}U content in power reactor fuel and depleted uranium mixtures from separation cascades are altered by the isotopic composition of the feed uranium, the relative quantities of the feeds when more than one kind is used, and the magnitude of a partially enriched side withdrawal, if any. Therefore, the values were arbitrarily chosen as fairly representative of these two classes of uranium. Irradiation of uranium in a nuclear reactor results in the production of ^{236}U and a reduction in both ^{234}U and ^{235}U, although the quantity of ^{234}U relative to that of ^{235}U will be increased as seen in the composition of low- and high-burnup uranium. Enriching uranium for use in nuclear power reactors by recycling reactor tails uranium as feed to separation cascades will introduce ^{236}U into the cascade which otherwise would not be present. For the calculation of the ^{236}U in power reactor fuel, highly enriched uranium, and depleted uranium, cascade feeds were assumed to consist of low-burnup uranium and natural uranium with low-burnup uranium providing one-half of the total ^{235}U input.

Three different mixtures of seven plutonium isotopes as defined by Healy[14] were used: low-irradiation plutonium with a composition reasonably representative of plutonium used in weapons, high-irradiation plutonium typical of plutonium recycle in light water reactors, and heat source plutonium.[15]

The maximum number of fission fragment tracks which can be produced per atom for a particular nuclide is equal to $2I\sigma$, where I is the thermal neutron fluence in neutrons/cm^2 and σ is the thermal neutron cross-sectional area per target atom of the nuclide in cm^2/atom. This assumes that there will be two fission fragments per fission. This relationship was used to calculate the maximum number of fission fragment tracks, T_f, which could result from irradiating 10^{10} atoms with a fluence of 8.64×10^{14} thermal neutrons/cm^2. A target size of 10^{10} atoms was chosen because a sphere of plutonium dioxide 0.9 μm in diameter would contain 10^{10} atoms of plutonium. 8.64×10^{14} neutrons/cm^2 was selected as the fluence, because this was approximately the fluence to which the polycarbonate films were exposed. If σ is expressed in barns/atom, then

$$T_f = 2(8.64 \times 10^{14}\ n/cm^2)(10^{10}\ \text{atoms})(\sigma\ \text{barns/atom})(10^{-24}\ cm^2/\text{barn})$$

or

$$T_f = 17.28\ \sigma$$

For nuclide mixtures, the sum of the contributions of each ith nuclide was used

$$T_f = 17.28 \sum_i f_i \sigma_i$$

Where a nuclide decayed by spontaneous fission, the contribution of this process was calculated on the basis of the number of fission fragment tracks produced by 10^{10} atoms during a seven-day period or

$$T_f = \frac{2(10^{10}\ \text{atoms})(7\ \text{days})\ \ln 2}{T_{\frac{1}{2}}(\text{SF})\ \text{days}} = \frac{9.7 \times 10^{10}}{T_{\frac{1}{2}}(\text{SF})}$$

The same reasoning was used to calculate the maximum number of alpha particle tracks which would be produced in seven days from 10^{10} atoms from the relationship

$$T_a = \frac{F(10^{10}\ \text{atoms})(7\ \text{days})\ \ln 2}{T_{\frac{1}{2}}(\alpha)\ \text{days}} = \frac{4.85 \times 10^{10}\ F}{T_{\frac{1}{2}}(\alpha)}$$

where F = the fraction of the nuclide which decays by alpha particle emission. When isotopic mixtures were involved, the relationship became

$$T_a = 4.85 \times 10^{10} \sum_i \frac{f_i F_i}{T_{\frac{1}{2}}(\alpha)_i}$$

The isotopic mixtures of plutonium contain ^{241}Pu. Only 0.0023% of this nuclide decays by alpha emission to ^{237}U. The remainder decays by beta emission to ^{241}Am, which has a 433-year half-life. The amount of this americium nuclide in a mixture will reach a maximum of 0.887 of the initial ^{241}Pu atom percent in 74.6 years. ^{241}Am will add additional alpha tracks to those from plutonium. Thus, two calculations were made for each plutonium mixture: one for freshly purified plutonium and one for 75-year old plutonium containing the maximum ^{241}Am activity. In two of the three mixtures, this caused an increase in alpha particle to fission fragment ratios. However, with heat-source plutonium, the decrease in ^{238}Pu activity was not compensated for by the increase in ^{241}Am activity.

The alpha particle to fission fragment track ratios, T_a/T_f, were calculated. The nuclides were then arranged in order of increasing ratios and listed in Table 1.

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TABLE 1

Theoretical Number of Fission and Alpha Tracks from 10^{10} Atoms and the Ratio of Alpha to Fission Tracks

Nuclides	*Fission Tracks per 10^{10} Atoms*	*Alpha Tracks per 10^{10} Atoms*	*Ratio of Alpha to Fission Tracks*
Power Reactor Fuel (4% ^{235}U)	3.99×10^{2}	2.14×10^{-1}	5.36×10^{-4}
Low Burn-Up Uranium (2.5% ^{235}U)	2.50×10^{2}	1.50×10^{-1}	6.00×10^{-4}
Highly Enriched Uranium (90% ^{235}U)	8.98×10^{3}	6.54	7.29×10^{-4}
Natural Uranium (99% ^{238}U)	7.18×10^{1}	5.74×10^{-2}	7.99×10^{-4}
Depleted Uranium (~100% ^{238}U)	2.49×10^{1}	3.60×10^{-2}	1.44×10^{-3}
High Burn-Up Uranium (1% ^{235}U)	8.88×10^{1}	1.44×10^{-1}	1.62×10^{-3}
^{247}Cm	1.25×10^{3}	8.10	6.48×10^{-3}
^{242m}Am	1.31×10^{5}	4.20×10^{3}	3.19×10^{-2}
^{233}U	9.06×10^{3}	8.20×10^{2}	9.05×10^{-2}
^{245}Cm	3.73×10^{4}	1.56×10^{4}	4.17×10^{-1}
Low–Irradiation Pu (94% ^{239}Pu)	1.20×10^{4}	$(6.48\text{-}7.52) \times 10^{3}$	$(5.40\text{-}6.27) \times 10^{-1}$
^{247}Cm (SF)	<6.64	8.10	>1.22
^{251}Cf	8.30×10^{4}	1.48×10^{5}	1.78
^{248}Cm (SF)	5.78×10^{1}	3.37×10^{2}	5.84
High–Irradiation Pu (40% ^{239}Pu)	8.02×10^{3}	$(5.13\text{-}9.31) \times 10^{4}$	$(6.40\text{-}1.16) \times 10^{1}$
^{249}Cf	2.88×10^{4}	3.79×10^{5}	1.32×10^{1}
^{252}Cf (SF)	3.13×10^{6}	5.02×10^{7}	1.61×10^{1}
^{243}Cm	1.19×10^{4}	4.43×10^{6}	3.71×10^{2}
Heat Source Plutonium (80% ^{238}Pu)	2.40×10^{3}	$(1.22\text{-}1.07) \times 10^{6}$	$(5.08\text{-}4.46) \times 10^{2}$
^{250}Cf (SF)	1.56×10^{4}	1.02×10^{7}	6.50×10^{2}
^{246}Cf (SF)	1.56×10^{1}	2.76×10^{4}	1.76×10^{3}
^{241}Am	5.43×10^{1}	3.07×10^{5}	5.66×10^{3}
^{252}Cf	5.53×10^{2}	5.02×10^{7}	9.08×10^{4}
^{244}Cm (SF)	2.03×10^{1}	7.34×10^{6}	3.62×10^{5}

TABLE 2

Analyses of Particles Containing ^{239}Pu

Fission Tracks	*Number of Particles*	*Total Fission Tracks*	*Total Alpha Tracks*	*Ratio Alpha to Fission Tracks*	*Standard Deviation*
3-4	33	125	108	0.86	0.68
5-9	80	528	358	0.68	0.49
10-14	65	783	569	0.73	0.44
15-19	36	607	489	0.81	0.45
20-24	26	570	432	0.76	0.38
25-29	11	294	275	0.94	0.50
30-34	13	408	417	1.02	0.38
35-39	7	262	287	1.10	0.32
40-44	16	672	819	1.22	0.44
45-49	9	423	398	0.94	0.45
50-54	4	212	302	1.42	0.72
55-59	1	58	41	0.71	
60-64	6	371	362	0.98	0.30
65-69	2	136	124	0.91	0.08
80-84	2	160	172	1.08	0.46
85-89	1	86	86	1.00	
90-94	2	182	147	0.81	0.18
100-104	1	104	56	0.54	
Total	315	5981	5442	0.91	

TABLE 3

Analyses of Particles Containing ^{235}U

Fission Tracks	*Number of Particles*	*Total Fission Tracks*	*Total Alpha Tracks*	*Ratio Alpha to Fission Tracks*
3-4	124	435	3	0.0069
5-9	146	935	2	0.0021
10-14	39	460	0	0.0000
15-19	18	293	0	0.0000
20-24	10	214	0	0.0000
25-29	3	82	0	0.0000
30-34	4	126	0	0.0000
35-39	3	110	0	0.0000
40-44	3	125	0	0.0000
Total	350	2780	5	0.0018

TABLE 4

Thermal Cross Sections, Alpha Decay, and Half-Lives for Alpha Decay and Spontaneous Fission for Selected Nuclides

Nuclide	σ_{nf}^{2200}, 10^{-24} *cm*^{2}	*Alpha Decay, %*	$T_{1/2}$ *(α), days*	$T_{1/2}$ *(SF), days*
^{233}U	5.245×10^{2}	10^{2}	5.92×10^{7}	
^{234}U	$<6.5 \times 10^{-1}$	10^{2}	9.06×10^{7}	
^{235}U	5.771×10^{2}	10^{2}	2.60×10^{11}	
^{236}U		10^{2}	8.77×10^{9}	
^{238}U	0	10^{2}	1.65×10^{12}	
^{236}Pu	1.62×10^{2}	10^{2}	1.04×10^{3}	
^{238}Pu	1.73×10^{1}	10^{2}	3.21×10^{4}	
^{239}Pu	7.406×10^{2}	10^{2}	8.90×10^{6}	
^{240}Pu	3.0×10^{-2}	10^{2}	2.39×10^{6}	
^{241}Pu	1.015×10^{3}	2.3×10^{-3}	5.41×10^{3}	
^{242}Pu	0	10^{2}	1.41×10^{8}	
^{244}Pu		10^{2}	3.02×10^{10}	
^{241}Am	3.14	10^{2}	1.58×10^{5}	
^{242m}Am	7.600×10^{3}	4.8×10^{-1}	5.55×10^{4}	
^{243}Cm	6.90×10^{2}	10^{2}	1.10×10^{4}	
^{244}Cm		10^{2}	6.61×10^{3}	4.79×10^{9}
^{245}Cm	2.161×10^{3}	10^{2}	3.12×10^{6}	
^{246}Cm		10^{2}	1.76×10^{6}	6.21×10^{9}
^{247}Cm	7.23×10^{1}	10^{2}	5.99×10^{9}	$>1.46 \times 10^{10}$
^{248}Cm		10^{2}	1.44×10^{8}	1.68×10^{9}
^{249}Cf	1.665×10^{3}	10^{2}	1.28×10^{5}	
^{250}Cf		10^{2}	4.78×10^{3}	6.21×10^{6}
^{251}Cf	4.801×10^{3}	10^{2}	2.92×10^{5}	
^{252}Cf	3.20×10^{1}	10^{2}	9.66×10^{2}	3.11×10^{4}

TABLE 5

Atom Percents in Six Typical Mixtures of Uranium

Nuclide	*Natural Uranium*	*Power Reactor Fuel*	*Low-Burnup Uranium*	*High-Burnup Uranium*	*Highly Enriched Uranium*	*Depleted Uranium*
^{234}U	0.005	0.030	0.018	0.017	1.160	0.001
^{235}U	0.720	4.000	2.506	0.890	90.000	0.250
^{236}U	-	0.320	0.363	0.393	2.770	0.014
^{238}U	99.275	95.650	97.113	98.700	6.070	99.735

TABLE 6

Atom Percents in Three Typical Mixtures of Plutonium

Nuclide	*Low-Irradiation Plutonium*	*High-Irradiation Plutonium*	*Heat Source Plutonium*
^{236}Pu	-	5×10^{-6}	1×10^{-4}
^{238}Pu	0.0115	2.9	80.3
^{239}Pu	93.6	39.6	15.87
^{240}Pu	5.9	25.6	3.00
^{241}Pu	0.4	16.8	0.72
^{242}Pu	0.013	15.0	-
^{244}Pu	0.02	-	-

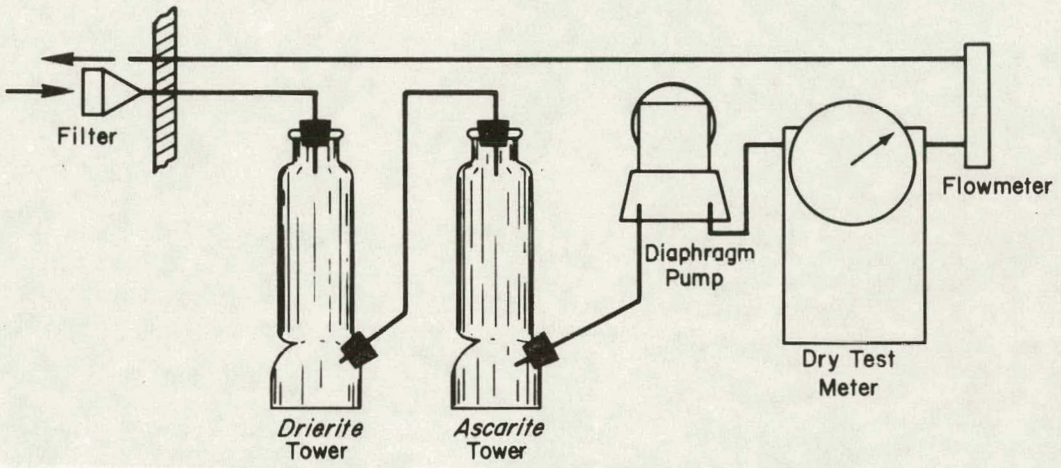


FIGURE 1. Arrangement of Sample Collection Equipment
Alpha Counter

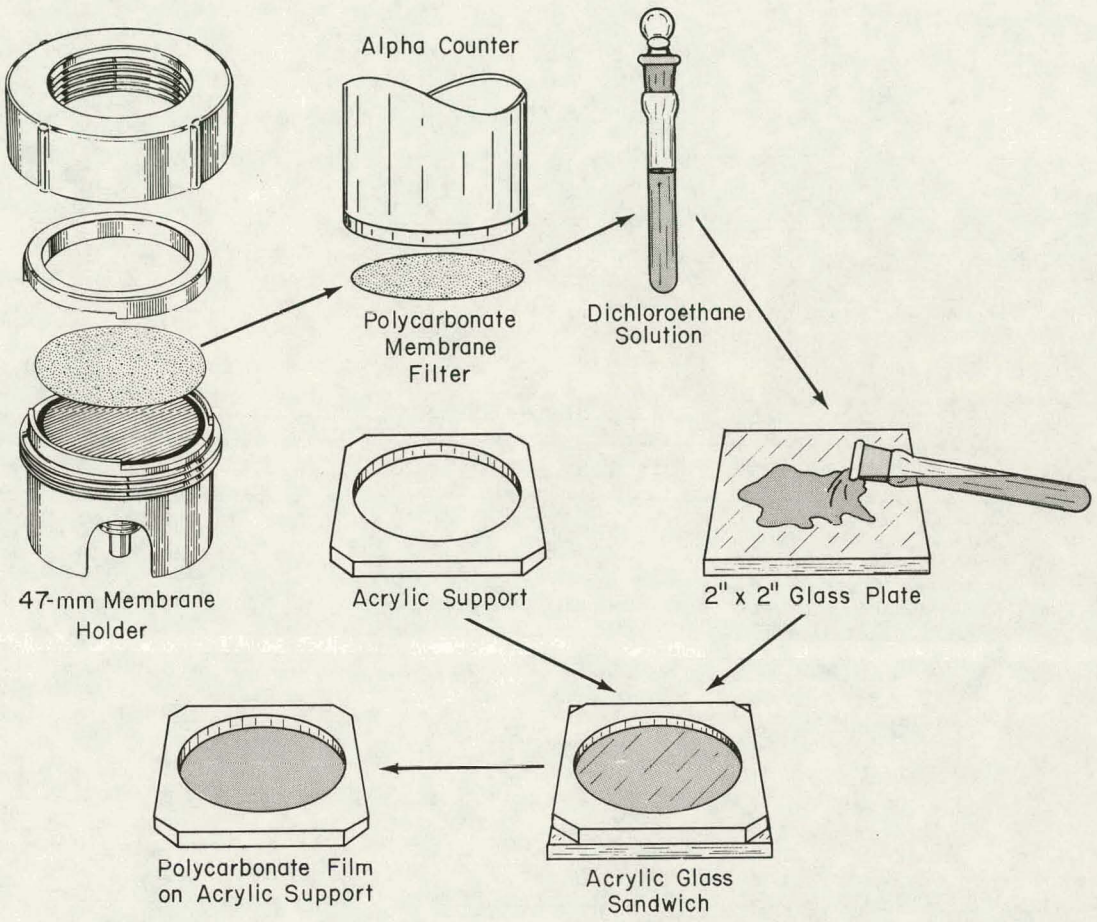


FIGURE 2. Procedure for Preparing Polycarbonate Films

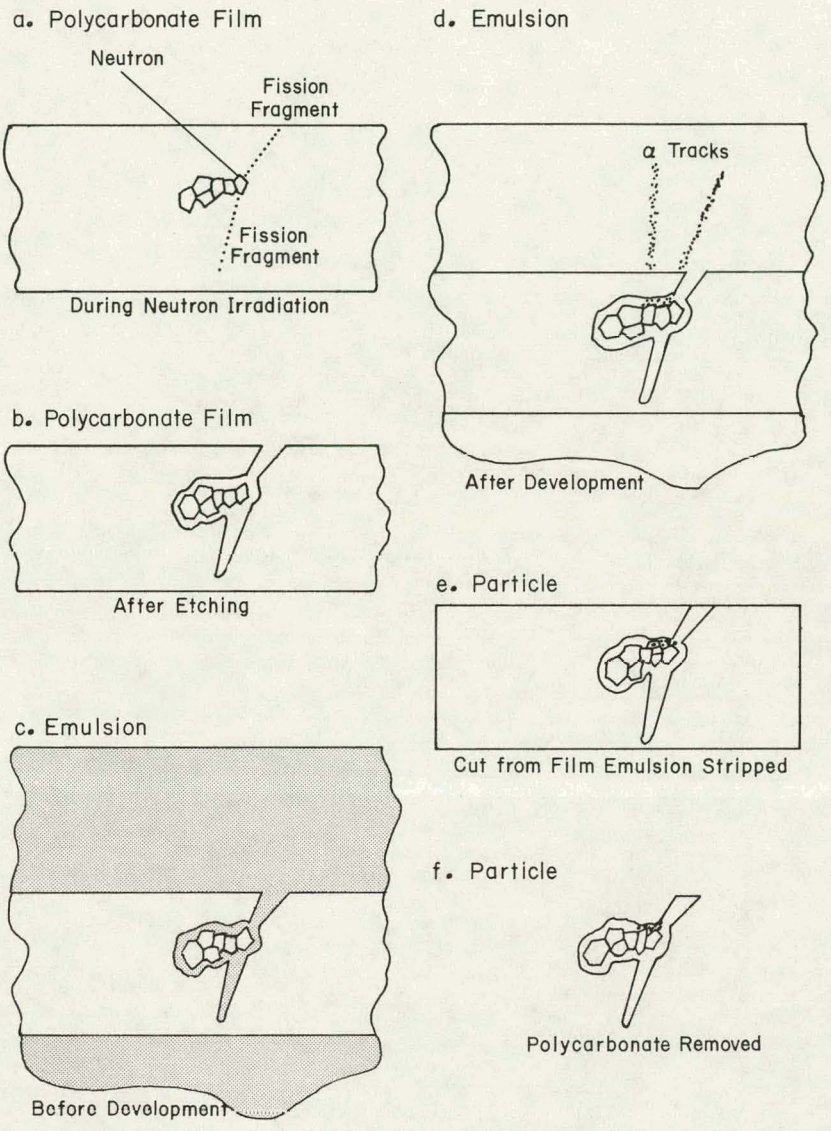
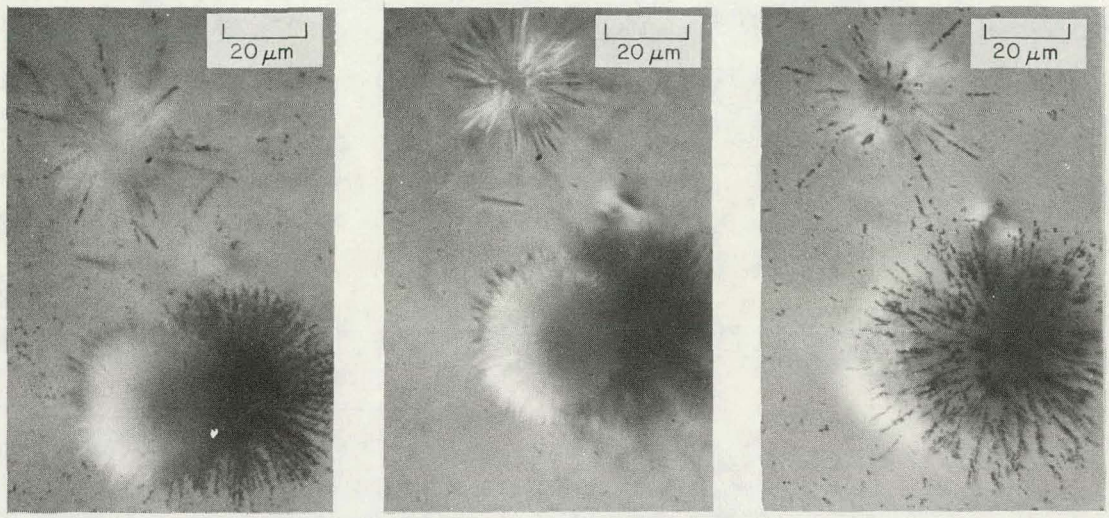


FIGURE 3. Graphic Representation of the Formation of Fission Fragment Tracks, Alpha Particle Tracks, and Gelatin Replicas



NTB Emulsion Below Two Particles Bearing ^{239}Pu

Two Particles Bearing ^{239}Pu in Polycarbonate

NTB Emulsion Above two Particles Bearing ^{239}Pu

FIGURE 4. Alpha Particle and Fission Fragment Tracks in Photographic Emulsion and Polycarbonate

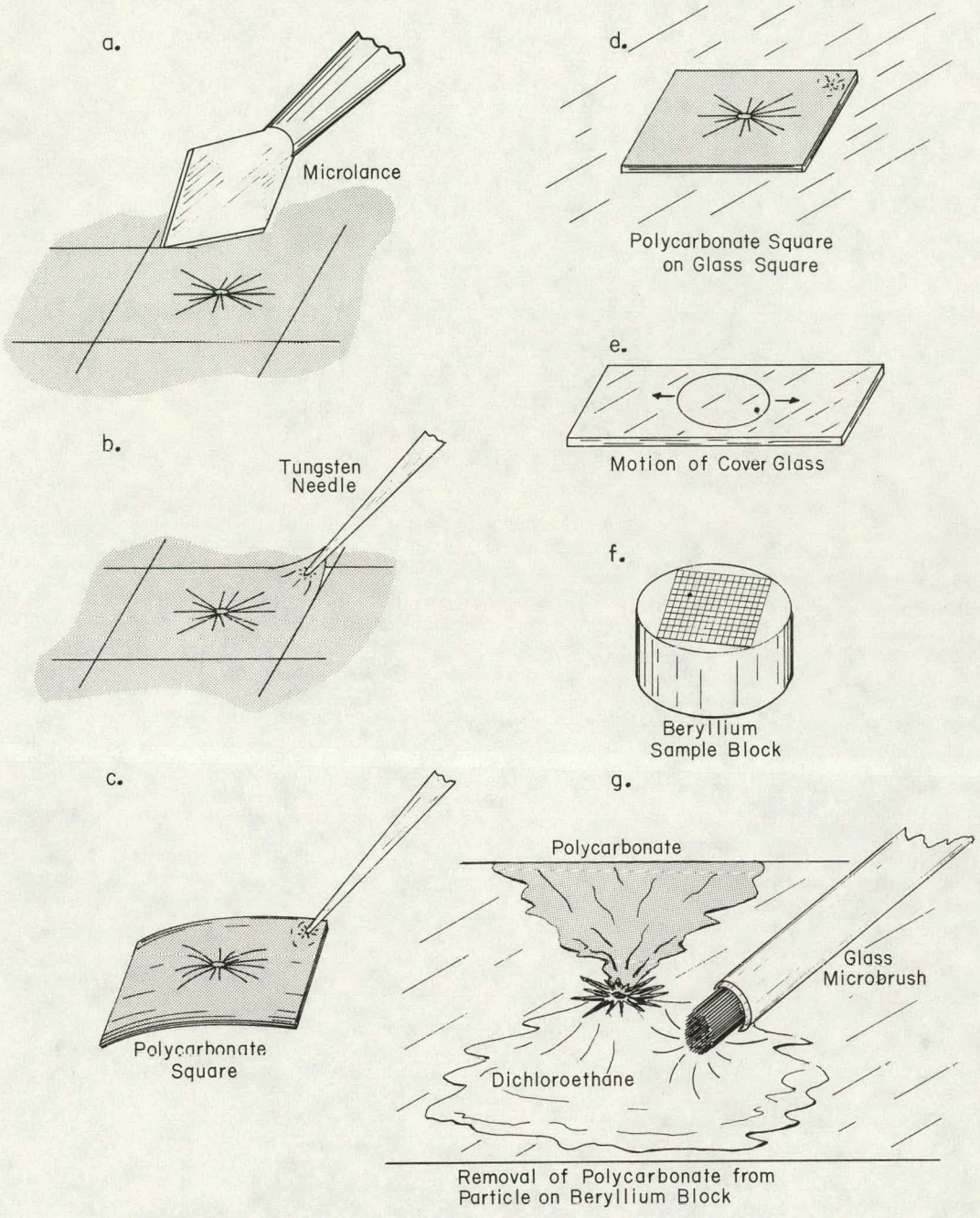


FIGURE 5. Procedure for Mounting Particle for Fissionable Material Identification

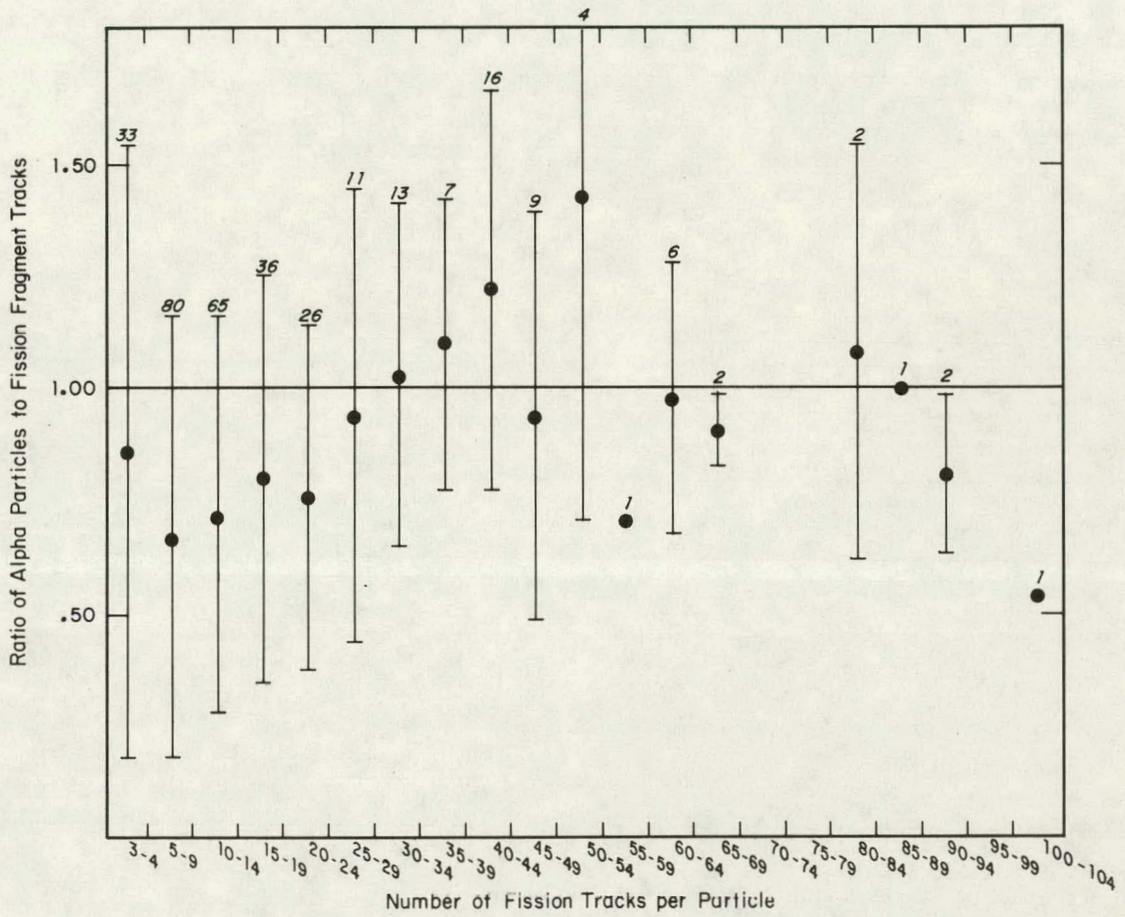
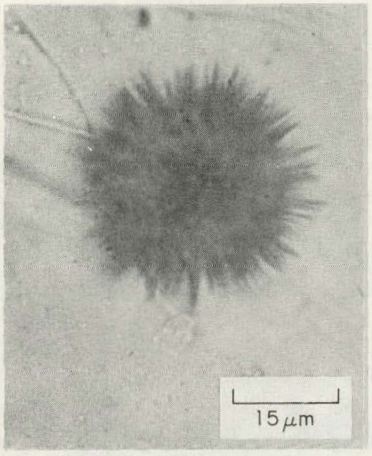
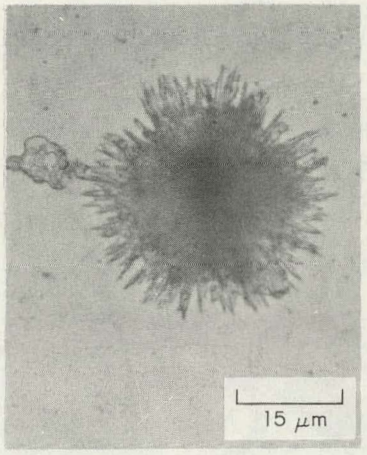


FIGURE 6. Effect of the Number of Fission Fragment Tracks on the Ratio of Fission Fragment Tracks to Alpha Particle Tracks



Particle No. 364 in Polycarbonate



Particle No. 364 with Polycarbonate Removed

Particle No. 364 with Emulsion Oxidized

FIGURE 7. Plutonium-Bearing Particle in Last Stages of Mounting