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INVESTIGATIONS OF ALPHA RADIOACTIVITY USING NUCLEAR EMULSIONS

Dean Carl Dunlavey
(Thesis)

August, 1952

Berkeley, California

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Investigations of Alpha Radioactivity Using Nuclear Emulsions

Dean Carl Dunlavey
Radiation Laboratory and Department of Chemistry
University of California, Berkeley, California

August 8, 1952

ABSTRACT

An alpha emitter of particle energy 4.85 Mev has been found in the bismuth fraction produced by the bombardment of lead with 60 Mev protons. Half-life observations and alpha systematics indicate that the emitting isotope is Bi^{203}. The assignment of a curium isotope having an alpha energy of 6.5 Mev has been corroborated as Cm^{238} by showing it to be the parent of Pu^{234}. Investigations have been made of complex structure in the alpha particle emission of Sm^{147}, Gd^{148}, Bi^{210}, Po^{208}, Th^{232}, U^{232}, U^{233}, U^{234}, U^{236}, U^{238}, Np^{237}, Pu^{236}, Pu^{238}, Pu^{239}, Am^{241}, Cm^{242}.

Investigations of Alpha Radioactivity Using Nuclear Emulsions

INTRODUCTION

Nuclear emulsions are finding a continually increasing number of uses in laboratories for investigations of radioactivity. The ability to detect, as for many instruments, results from the ionization produced as the activity traverses matter.

Becquerel[1] and Crookes[2] were among the first to perceive the fogging effect of uranium and thorium salts on photographic emulsions and to attribute the effect to radioactivity. Reinganum,[3] in 1911, is regarded as the first to associate a linear series of developed silver grains as recording the trajectory of a discrete particle. Although many early scientists employed the optical photographic emulsions for various radioactivity studies, the current acceptance of emulsions as an accurate tool of research began with the fairly recent availability of fine-grained 'nuclear emulsions' of especially high silver bromide content, with reproducible stopping power, and particularly suited for the detection of densely ionizing particles.

The use of emulsions is particularly suited for low counting rates, due either to a long half-life or to a scarcity of activity, and for coincidence work. In addition, the Ilford C-2 (alpha sensitive) and D-1 (primarily for fission fragments) emulsions have been found advantageous in locating alpha activity accompanied by high beta or electron capture activity which interfered with mechanical pulse analysis. Since each radioactive atom is completely surrounded by emulsion sensitive to charged particles and since most emitted particles come to rest before

leaving the emulsion, the method offers 100 percent geometry with no particle energy loss such as is found in the use of thick samples prepared for mechanical counters or caused by particles leaving the ionization chamber before expending their energy completely.

The emulsion can be loaded with a radioactive isotope and left for weeks or months, after which the accumulated activity can be counted. Radioactive decay rates too low for mechanical counters are easily identified by this method. Since each emitted charged particle leaves a track originating from the point where the parent atom is located, the complete decay of each such atom is uniquely recorded.

Observation of the grain density of a track allows easy identification of the particle as an electron, proton, alpha particle or fission fragment. Measurements of particle ranges permit quite accurate energy determinations; i.e. to within 0.1 Mev of alpha particles under 10 Mev and to within 5 kev for electrons of 15-75 kev.

The methods described herein were developed primarily for work involving alpha particle counting and for the observation of alpha-electron coincidences which are useful in determining the decay schemes of alpha emitting isotopes. The results obtained through the use of emulsions for alpha-electron coincidence studies in the investigation of complex alpha structure in certain isotopes have been previously described by other workers.[4,5,6]

EMULSIONS

The emulsions now used are composed of a layer of gelatin matrix, bearing a high content of small spherical grains of silver halides

(mostly the bromide, but with a small percentage of iodide), which is generally mounted on a 1 x 3 inch glass microscope slide. Table I shows a comparison of nuclear and ordinary optical emulsions.[7]

Table I

Property	Optical Emulsion	Nuclear Emulsion
AgBr:gelatin (mass)	47:53	83:17
AgBr:gelatin (volume)	15:85	45:55
Grain diameter, μ	1-3.5	0.1-0.6
Grain separation	interlocking	isolated by gelatin
Thickness, μ	2-3	25-400
Mass/unit area, mg/cm^2	1-4	40 mg per 100 μ thickness
Sensitivity to light	very high	high but not panchromatic (insensitive to safelight)

The passage of a charged particle through a grain renders the entire grain developable (provided the total ionization loss in the grain has been adequate). If this were not so, the tracks would not be visible. The observed path of the particle is, of course, that continuous accumulation of developed grains rendered developable by passage of the particle. By varying the size of the grains, emulsions can be made partially selective as to particles recorded since the grain size determines the minimum rate of ionization loss which a particle must have in order to sensitize the grains. Emulsions manufactured by Ilford are selective in respect to recording single events as listed in Table II (see Figure 1). It should be recognized that a large excess of any activity will fog any of the plates listed.

Table II

Film	Fission Fragments	Alpha Particles	Protons	Electrons	Gamma Rays
D-1 (fine grain)	heavy tracks	moderate tracks at low energies only	x	x	x
E-1	heavy tracks	heavy tracks at low energies	moderate tracks at end of range	x	x
C-2	heavy tracks	heavy tracks	moderate tracks up to 50 Mev	x	x
B-2	heavy tracks	heavy tracks	heavy tracks	x	x
G-5 (coarse grain)	heavy tracks	heavy tracks	heavy tracks	moderate tracks improving with lower energies	x

ERADICATION

Because of inevitable periods of delay between the manufacture of the emulsion and its use, all plates used were found to contain background tracks (i.e., alpha particles in C-2, alpha particles and electrons in G-5) formed either by radioactive impurities contained in the emulsion and glass backing or by cosmic radiation. In doing precise work with tracks of the same kind as those of the background, it was always desirable, and sometimes mandatory, to eradicate this background. In the work of interest to the author, attention was concentrated on alpha counting in C-2 and D-1 emulsion plates and alpha-electron counting in

G-5 plates; the eradication procedures used for removal of background of these types is described.

The generally described methods for eradicating alpha particles in C-2 plates involve placing the plate in an atmosphere of water vapor or dilute hydrogen peroxide vapor for varying periods of time and at various temperatures up to 40° C or occasionally higher. These procedures were found to reduce the density of the alpha tracks to varying degrees but never enough to preclude their recognition. The method for C-2 and D-1 eradication described below was developed by the author and gave complete and absolute eradication without any observable loss of sensitivity in respect to subsequent alpha recording.

Immerse the plate in 0.1 percent hydrogen peroxide solution at 20° C for 30 minutes.
Dry rapidly over sodium hydroxide (about 30 minutes if air is circulated).
Keep over anhydrous sodium carbonate in quiet air for 36 hours.

At the end of the 36 hour drying period the plates were thoroughly 'clean' and subject only to that background created subsequently. Effectiveness of the method in respect to eradication without desensitization was checked repeatedly by suspending an alpha emitting source above different portions of the emulsion surface before and after eradication; this produced a high density of tracks without incorporating any activity within the gelatin.

No comparably effective method of eradicating electrons from a G-5 plate was found. It was observed that any effective eradication involving heat or humidity appreciably decreased the electron sensitivity, particularly at energies over 40 kev. Therefore, the G-5 plates were ordered from the manufacturer frequently and in limited quantities so that

they could be used without eradication within 1-6 weeks after manufacture. Cosmic ionization of electrons from atoms composing the emulsion appeared to be the primary source of the background in the G-5 plates. Lead shielding was provided during storage and exposure to keep the background to a minimum.

The extent of latent image fading in the plates was determined by exposing a small section of the emulsion to an alpha active disc for a brief period at the beginning of each exposure time. The fading of the alpha tracks created by the disc then represented the maximum fading experienced by any other track in the film. The plates used in the current work were kept in a cool and dry atmosphere during exposure periods, never longer than two months, and no appreciable fading was observed.

LOADING

The emulsion forms a hard layer on the glass plate when dry but swells to several times its volume and becomes very spongy and tacky when wet. If a solute is present in the wetting solution, it diffuses into the gelatin until equilibrium is established. When the wetted emulsion is then dried, these solute atoms are trapped more or less uniformly throughout its volume. In the case of those trapped atoms which are radioactive, a complete record is left of all charged particles subsequently emitted.

The solutions introduced had to have a pH of 3-9. If the pH was lower, the emulsion was desensitized or even dissolved; if higher, most solutes of interest became hydrolyzed or colloidal and would not penetrate into the emulsion. Frequently, in solutions of pH as low as 4 or 5,

it was necessary to have a complexing agent present to obtain satisfactory penetration. Dilute ammonium citrate or ammonium acetate ($\leq$0.01 M) solutions were commonly used because of their buffer and complexing properties. No oxidants were included in solution because of their destructive action on the latent images of the tracks. When ample activity was available, solutions were prepared having ~10 absolute disintegrations per minute per 100 λ. Saturation of the emulsion with the solution, followed by a 66 hour exposure period, provided a concentration of tracks in the emulsion (5-10 per field of view) adequate for rapid counting without interference arising from an excessive number of tracks. Using the well technique of loading (described below), 500 λ of such solution was ample. If the activity was not available in this concentration, longer exposure periods and extended microscopic counting could lower the counting rate of the sample to a magnitude of one count a day. The overall solute concentration of solutions used could be as high as 0.1 to 1 N without noticeably affecting the sensitivity or range-energy relations of the plates.

If soaked in a large volume of solution, the emulsion was effectively saturated after about 60 minutes. In its swelled condition, it contained about the same concentration of the radioactive solute as the solution, volume for volume. Thus, when dried and shrunk to its original volume, its volume for volume solute concentration was 3-5 times that of the solution. This ratio is generally referred to as the penetration factor. By knowing the volume of emulsion searched with the microscope for evidence of radioactivity, a fair estimate could be made of the total number of solute atoms which acted as the parent of the observed activity

and, if desired, the specific activity could thus be calculated. The following formula relates the amount of activity observed, its half-life and the abundance of the parent:[8]

$$dN = \lambda N dt = \frac{\ln 2 \cdot N \cdot dt}{T}$$

$$dN = \frac{\ln 2 \cdot n \cdot \pi \cdot r^2 \cdot d \cdot f \cdot c \cdot A \cdot dt}{T}$$

dN = number of events found in all.
n = fields of view searched.
r = radius of field of view.
d = dry film thickness.
f = penetration factor.
c = concentration of the solution.
A = Avogadro's number.
dt = exposure time.
T = half-life of the radioactivity.

If a small aliquot of the solution was allowed to dry completely into the emulsion, the amount of solute absorbed was again known. However, much better penetration was achieved by having excess solution in contact with the emulsion.

When the amount of radioactive solution was limited, a small well was constructed above the emulsion by using a one-fourth inch thick block of plastic cut to the dimensions of the plate. A hole of appropriate size was milled through the block and when the block was placed flush against the emulsion, a well was formed enabling a column of liquid to be placed above the emulsion. A watertight seal was produced by lightly greasing the bottom of the block before placing it on the plate (see Figure 2). This column gave penetration equivalent to that obtained by immersing the entire plate. When saturation had been reached (1-3 hours),

the well was emptied with a pipet, the block removed from the plate, the wetted area allowed to dry and the residual grease removed by careful scraping so as not to spread it onto the exposed area. Because the sensitivity of the emulsion and its stopping power were both much decreased when wet, the time taken for loading and drying had to be short relative to the total time of exposure (recording of activity). Drying of 100 μ C-2 and D-1 emulsions could be accomplished in 20-30 minutes by placing in a sodium hydroxide charged dessicator containing a circulation fan (see Figure 3). When dried, the emulsion was promptly removed to prevent peeling caused by lowering the water content of the gelatin excessively. G-5 plates (100 μ) had to be allowed to dry normally while standing in air (~3 hours) or over anhydrous sodium carbonate since rapid drying over sodium hydroxide highly increased the grain background of the area after development.

Following loading and prior to processing, the plates were kept in a relatively cool (20° C) and dry (over anhydrous sodium carbonate) location to minimize fading. In case of G-5 emulsions, two inch lead shielding against cosmic irradiation was also provided.

PROCESSING

Since the emulsions used were solely of 100 μ thickness, no complex developing procedures were required. The developer used predominantly was D-19 stock solution diluted 1:6 with water (pH about 11). In cases where a somewhat acidic complexing agent (i.e. citrate) had been retained in the emulsion with the activity, an acid developer (diaminophenol hydrochloride) of pH about 6 was occasionally used to give uniform track

densities throughout the emulsion. In most cases, however, presoaking of the emulsion in dilute sodium carbonate solution at 15° C for 20 minutes was used to prepare such acidic emulsions for satisfactory D-19 development. The fixer used was ordinary photographic acid fixer. To avoid radioactive contamination from batch to batch, no chemicals were used more than once.

The exact procedure was as follows (see Figure 3).

Presoak in dilute sodium carbonate solution at 20° C if necessary.

Develop 30 minutes at 20° C (45-60 minutes for G-5) with constant but mild agitation. The combination of weak developer and long development time gave a more nearly uniform track density throughout the emulsion.

Stop bath of water or 1 percent acetic acid for 5 minutes at 20° C without agitation.

Fix for 4.5 hours with constant, mild agitation. Plates must not be exposed to any light except safelight until fixation is completed.

Rinse 90 minutes in running water; allow to dry overnight.

Following processing and drying, any coating of metallic silver left on the emulsion surface was removed, losing less than 0.25-0.50 μ of emulsion, by careful scrubbing with alcohol dampened cotton. Because of the high silver halide content which is removed by fixation, the emulsions decrease in thickness after processing and this shrinkage must be corrected for in track measurements (see COUNTING).

When properly processed, the plates were clear and transparent with no distortion visible in the emulsion. Ilford plates, excepting G-5, had a slight green or brown tint from colloidal silver left in the gelatin.

COUNTING

Following processing and drying, the emulsions were best examined with a binocular microscope having a rectangular stage. If every event in a given area of the plate had to be located (i.e. determining the specific alpha activity of an aliquot), scanning was done using a 5x eyepiece and a 90x oil-immersion objective. This gave the minimum magnification (450x) and maximum area coverage per field of view commensurate with the accuracy and detail required to assure location of every event. Upon location of an event, measurement of each particle range or energy was accomplished by changing to a 15x eyepiece and using the procedure outlined below. If it was only required to count the relative frequencies of different events (i.e., alpha particles vs alpha particles with coincident conversion electrons), then a 10x eyepiece was convenient for both scanning and counting.

Scanning was most simply done by searching from the top to the bottom of the emulsion in one field of view and then moving on to an adjacent field of view somewhat overlapping the first and repeating the top to bottom search, etc. A field of view of 450x magnification covered about 0.03 mm^2 of emulsion area; hence, it took a minimum of 3300 separate fields of view to cover 1 cm^2 of emulsion area, discounting the necessary overlapping. It was therefore of obvious interest to minimize the area requiring scanning.

In determining the range, and so the energy, of a particle in the emulsion, it was generally necessary to record the horizontal and vertical components of the track and then to compute the true path. The horizontal component was measured by use of a reticle contained in the 15x

eyepiece, the graduations of the reticle having been previously calibrated in terms of microns by use of a horizontal stage micrometer. The vertical component of the track was measured using the graduations of the fine adjustment knob of the microscope, focusing first on one end of the track and then on the other. Through prior calibration with a vertical stage micrometer, the knob reading differences were also translated into microns. Since the focal plane of the field of view was slightly concave upward, the ends of a measured track had to be at the same distance from the circumference of the field of view.

The depth of the emulsion decreased about 50 percent between the time the track was formed and the time it was measured (see emulsion processing); therefore, the measured vertical component had to be multiplied by a shrinkage factor to correct it to the original depth. It was determined in this laboratory, using uranium tracks as a standard, that the factor was 2.2 for C-2 emulsions (and essentially the same for other Ilford emulsions). Hence, a track measured at 12 μ horizontal component and 5 μ depth component actually was formed with a 12 μ horizontal component and a 11 μ vertical component, with a true length of $\sqrt{144 + 121} = 16.3\ \mu$. Because this depth correction introduced the major uncertainty of the range measurement, the best energy determinations were made on those tracks lying most completely in the horizontal plane.

IDENTIFICATION OF PARTICLES AND THEIR ENERGIES

Fission fragments, alpha particles, and low energy protons, because of their high mass and rate of ionization, all leave dense, straight tracks in the C-2 and G-5 emulsions. Electrons, in G-5 emulsions, in

contrast, leave uniquely irregular, sketchy tracks. Differentiation of alpha tracks from proton or fission fragment tracks was always made easy merely by considering the range and grain density of the tracks.

Besides identifying the kind of particle, it was generally of interest to determine its energy. As in any medium, the particle range was used as the best measure of its energy. The stopping power of emulsions of the same kind (i.e. C-2) is now sufficiently uniform so that range-energy relationships prepared for one manufactured batch could be considered valid for all others. Accurate range-energy relationships for alpha particles in C-2 emulsions have been prepared by Rotblat[9] (see Figure 4). These were checked by the author from the 8.8 Mev alpha of Po^{212} to the 2.18 Mev alpha particle of Sm^{147} and they have also been corroborated to within a few percent by other workers.[10] This range vs energy curve also offered a very close approximation when using either other Ilford emulsions or Eastman NTA or NTB plates.

Less accurate range-energy determinations were available for electrons in G-5 emulsions and could only be used to approximate the energy[11] (see Figure 5).

PHOTOMICROGRAPHY

Clear and detailed photographs of the field of view seen through the microscope could be obtained by use of only an intense light source and a monocular tube. The light was transmitted from the sub-stage mirror through the condenser, plate and objective and out through the eyepiece of the monocular tube. The image, exactly as seen in the microscope, was in sharp focus on a screen held from 1 inch to 1 foot above

the eyepiece. Substitution of film for the screen provided a photo of any desired magnification.

Bi^{203}

In plotting the alpha decay energies vs mass numbers for the isotopes of bismuth and heavier elements, it is seen that the energy of disintegration for any element increases as the number of included neutrons of the daughter isotopes decreases to the especially stable number of 126. Past this energy maximum the energy drops abruptly to a minimum and then begins to rise again.[12] For bismuth, the isotope 211 (decaying to a daughter with 126 neutrons) lies at the top of the energy peak and the isotope 210 is known to lie much below it. However, the next known lighter alpha emitting isotope is 201 and this lies well up on that part of the energy curve (past the 126 neutron closed shell) which is again increasing with decreasing neutron number. Alpha particle emission from the intermediate isotopes 202-208, presumably because of the lower alpha particle energies and subsequently longer half-lives, has not been observed due to the short overall half-lives resulting from electron capture instability.

The most favorable alpha/electron capture ratio within this region was expected for the lighter isotopes where the alpha particle energy is highest; 202, 203 or possibly 204. To produce the maximum relative concentration of these isotopes, a target of natural, highly pure lead foil was bombarded for 50 minutes with a 60 Mev proton beam. A period of 12 hours after bombardment was allowed for chemistry and for decay of the light isotopes, the longest lived of which was 60 minute Bi^{201}. The

chemistry was designed to give carrier-free bismuth, separated from all other radioactive elements, in a solution of pH ~4..

The activity of the Bi^{201} alpha particle (5.15 Mev) could no longer be detected with the pulse analyzer following the 12 hour period. From the Geiger decay curves it was found that, during the last 36 hours of the first 48 hours following bombardment, ~90 percent of the total activity was of about a 12 hour half-life, assumed equally from Bi^{203} and Bi^{204} (see Figure 6). This 36 hour span, occurring after Bi^{201} and lighter isotopes had decayed out and before longer lived isotopes could contribute appreciably to the overall activity and covering three half-lives of the appropriate isotopes (Bi^{203}, Bi^{204}), was the period decided upon for investigation.

Twelve hours after the first bombardment, 100 λ aliquots of the original solution and solutions prepared by systematic dilution were impregnated into separated areas of about 2 cm^2 each of Ilford C-2 emulsions, rapidly dried in circulating air over sodium hydroxide and allowed to stand for 36 hours. The emulsions were then developed and inspected under the microscope to find the maximum electron capture activity tolerated by the emulsion before being fogged. The highest tolerable activity was found to be that from a sample having an absolute counting rate of 1×10^6 c/m per 100 λ (windowless proportional counter) at the beginning of the exposure period. This represented about 1×10^9 total disintegrations during the 36 hours.

In all subsequent bombardments, the bismuth solutions were prepared approximately equal to this activity, impregnated into eradicated C-2 emulsions and rapidly dried over sodium hydroxide. The alpha activities

found during the 36 hour decay periods, as well as those found in 'background' checks, are recorded in Table III.

Table III

Bombardment	Exposure Time* (hours)	Total Counts (Absolute) (x 10^8)	Alpha Particles (Mev)** 4.0-5.0	Alpha Particles (Mev)** Other	Background Alpha Particles (Mev)† 4.0-5.0	Background Alpha Particles (Mev)† Other	Overall α/EC Ratio†† (10^{-8})
5	36	2	3	5	1	7	1.5
6	36	1.5	4	6	1	2	2.7
	36	1.5	2	3			1.3
9	36	8.4	9	5	0	1	1.1
	36	8.4	13	2			1.5
10	36	11.2	19	11	2	8	1.7
11	13	11.0	9	2	1	6	1.5
	13	(first 39	6	5			(for
	13	hours)	2	3			first 3
	26		1	7			periods)

*Beginning 12-14 hours post bombardment.
**Background alpha particles deducted (see Figure 7 for alpha particle energy histogram).
†Taken from 5-7 days post bombardment with a 48 hour exposure.
††Considering only those alpha particles in the 4.0-5.0 Mev range.

A plot of the alpha particles observed between 4.0 and 5.0 Mev is shown in Figure 7. Because of the colloidal nature of the bismuth in the nearly neutral solution, most of the alpha particles originated from within 'activity spots' caused by the concentration of associated gamma-electron activity (see Figure 8). It was considered of interest and significance that neither the alpha particles outside the 4.0-5.0 Mev

energy range nor uranium activity, added as a sensitivity check during each run to other plates containing bismuth aliquots and developed separately, originated from such spots.

By considering the time lapse before, and the duration of, the exposure period, it seemed certain that the alpha particles were originating from a parent whose half-life was of the order of a day. The final bombardment indicated this half-life to be of the order of one-half day. From either consideration, and from the fact that no other alpha emitter of comparable energy and half-life is known, it was apparent that the parent must be either Bi^{203} or Bi^{204}. Comparing this energy on the plot of energy vs mass number of the other bismuth isotopes, it was found that it closely fitted the energy expected from Bi^{203} but was too high for that of Bi^{204}. Therefore, the alpha particle emitting isotope was assigned as Bi^{203}.

The alpha/electron capture ratio for Bi^{203} was then about 3.3×10^{-8}. Because of the quick drying of the impregnated solution, the penetration into the emulsion was poor, of the order of 1-5 μ. This meant that approximately one-half of the alpha particles emitted passed out the surface of the emulsion and could not be measured. Hence, the total alpha particle count was assumed to be effectively twice the number of measured tracks and the final alpha/electron capture ratio for Bi^{203} was assigned as 6.6×10^{-8}.

Using this branching ratio, the alpha partial half-life was easily calculated.

$$\frac{\text{alpha half-life}}{\text{EC half-life}} = \frac{\text{EC counts}}{\text{alpha counts}}$$

$$\text{alpha half-life} = \frac{13 \text{ hours}}{6.6 \cdot 10^{-8}} = \frac{13}{24 \cdot 365 \cdot 6.6 \cdot 10^{-8}} \text{ years} \approx 2 \cdot 10^{4} \text{ years}$$

This energy and corresponding alpha particle half-life are in good agreement with the curve of alpha particle energy vs half-life for other bismuth isotopes. From the results of this experiment, it seems certain that isotopes 204-208 will have increasingly unfavorable alpha/electron capture ratios and hence cannot be examined by this method.

MASS ASSIGNMENT OF Cm^{238}

An interesting and unusual application of nuclear emulsions was employed to confirm the mass assignment (238) of a curium isotope having an alpha particle energy of 6.5 Mev and a half-life of 2.3 hours.

Following preparation by G. H. Higgins, a 30 λ aliquot of the curium activity, having 0.7 c/m of the 6.5 Mev alpha particle, was diluted to reduce the citrate concentration to 0.1 M and impregnated into an Ilford C-2 nuclear plate. One hour was allowed for penetration after which the plate was rapidly dried in the sodium hydroxide dessicator.

Considerable additional alpha activity as well as $\sim 10^{4}$ c/m electron capture activity was present in the aliquot from contaminating americium and curium isotopes. The daughter of Cm^{238}, Pu^{234}, has a half-life of 8 hours, decaying with an electron capture/alpha ratio of 20/1. Therefore, if the alpha emitter under investigation was indeed Cm^{238}, a limited number of events showing two alpha particles of appropriate energies and coincident in origin were expected to be found in the emulsion following a 42 hour exposure.

Investigation of approximately one-fourth of the impregnated area revealed five events having two coincident alpha particles; these were of energy 6.5 ± 0.1 Mev (Cm^{238}) and 6.15 ± 0.1 Mev (Pu^{234}) (see Figure 9).

In view of the abundance of the events and the fact that two and only two successive alpha particles of such energies cannot be placed in any other known decay chain, the mass assignment was considered confirmed.

Of special interest were the following points:

An activity of ~2 hours half-life was successfully impregnated into a nuclear emulsion and dried in time to observe the bulk of the decay.

A concentration of 0.1 M citrate did not affect the sensitivity or development of the C-2 emulsion to an observable extent.

The activity penetrated throughout the emulsion uniformly within one hour's time.

Five events of the desired kind were effectively segregated and identified from among several thousand contaminant alpha tracks and ~10^4 c/m electron capture disintegrations.

INVESTIGATIONS OF COMPLEX STRUCTURE IN ALPHA PARTICLE EMISSION WITH NUCLEAR EMULSIONS

A study has been made, utilizing the method of photographic emulsions, of the conversion electrons accompanying the complex alpha group structure in a number of nuclides. These conversion electrons, coincident in origin with the alpha particles, were identified in Ilford G-5 nuclear emulsions (see Figure 10). Energies of the electrons were approximated from their range and this, together with postulation of the shell of conversion (or binding energy) of the electrons, allowed calculation of the gamma ray energy or nuclear energy level separation. The percentage of alpha particles having such conversion electrons associated with them

represent a lower limit to the alpha decay leading to the corresponding nuclear energy level since the unconverted gamma rays are not measured by this technique.

Fresh plates were ordered frequently and used within one to eight weeks after the time of manufacture in order that no eradication of electron background (with associated desensitization of the emulsion) would be required. Because of residual acidity left in the emulsion with the radioactivity through use of citrate or acetate complexing agents (to aid penetration of the radioactivity into the emulsion), a 30 minute presoaking of the plates in dilute sodium carbonate solution preceded normal development with Eastman D-19 solution. The customary period of exposure was 66 hours; where the daughter was a beta emitter of sufficiently short half-life, appropriate corrections were made in the percentages of electron abundance. Alpha particle emitting contaminants were negligible (0-1 percent) in all cases except U^{238} for which an isotopically enriched sample was used and appropriate corrections made for U^{234} and U^{235}. Close agreement was found between the present results and those of Asaro and co-workers in earlier work on the two nuclides Cm^{242} and Pu^{238} using the alpha magnetic spectrometer; this was regarded as indicating satisfactory efficiency of the emulsion method in estimating both electron abundances and energies.

The results are summarized in Tables IV and V where the gamma ray energies are probably accurate to about 5 kev. As can be seen, in the case of the even-even emitters, the alpha decay observed in every case divides between the ground state and one higher energy level of the daughter nucleus. Since the conversion coefficient for the gamma ray

Table IV

Conversion Electrons Accompanying Alpha Decay of Even-Even Nuclides

Isotope	Alpha Events Observed	% Having Conversion Electrons	Electron Energies (kev)	Shell of Conversion	Gamma Energy (kev)
Cm^{242}	5250	23 ± 3	25 ± 5 (19%)	L	~45
			40 ± 5 (4%)	M	~45
Pu^{238}	5700	23 ± 3	20 ± 5	L	~40
			35 ± 5	M	~40
Pu^{236}	7000	20 ± 3	25 ± 5 (17%)	L	~45
			40 ± 5 (3%)	M	~45
U^{238}	6300	22 ± 3	25 ± 5	L	~45
			40 ± 5	M	~45
U^{236}	9700	27 ± 3	30 ± 5 (23%)	L	~50
			45 ± 5 (4%)	M	~50
U^{234}	6700	25 ± 3	30 ± 5	L	~50
			45 ± 5	M	~50
U^{232}	8000	30 ± 3	40 ± 5 (26%)	L	~60
			55 ± 5 (4%)	M	~60
Th^{232}	5100	24 ± 3	35 ± 5	L	~55
			50 ± 5	M	~55
Po^{208}	6500	0			
Gd^{148}	6200	0			

Table V

Conversion Electrons Accompanying Alpha Decay of Odd Nucleon Nuclides

Isotope	Alpha Events Observed	% Having Conversion Electrons	Electron Energies (kev)	Shell of Conversion	Gamma Energy (kev)
Am^{241}	2600	56 ± 5	≤20 (12%) 20-35 (31%) 35-60 (9%) 2 electron coincidences (4%)		several levels, highest at least 65 kev above the ground level
Pu^{239}	8100	12 ± 2	30 ± 5 45 ± 5 20 ± 5 30 ± 5 100 ± 20 (0.5%)	L M L M	~50 ~50 ~35 ~35 origin uncertain
Np^{237}	3500	80 ± 5	20 (10%) 20-40 (38%) 40-65 (20%) 2 electron coincidences (11%) 3 electron coincidences (1%)		several levels, highest at least 65 kev above the ground level
U^{233}	4700	9 ± 3 ~0.4	20 ± 5 35 ± 5 70 ± 10	L M L	~40 ~40 90 ± 10
Bi^{210}	1800	0			
Sm^{147}	7500	0			

seems to be high for all such daughter nuclei of the even-even type where this has been measured, it seems reasonable to assume that the percentages of conversion electrons listed in Table IV correspond essentially to the percentages of the alpha decays to the level above the ground state. In the case of Po^{208} and Gd^{148} the absence of conversion electrons indicating decay to an excited level is attributed to the fact that the first excited level of even-even nuclides in a magic number region lies sufficiently above the ground state to preclude any readily observable competition with decay to the ground state.

It is seen that each of the nuclides with odd nucleons exhibits alpha decay to several energy levels with widely varying percentages; for these, the decay to one of the excited states is in each case less hindered than the corresponding decay to the ground state, in agreement with the view that the conditions for the assembly of the outgoing alpha particle can be more favorable in certain instances when the highest lying odd nucleon need not be included.

For the even-even isotopes, it was of interest to determine the quantitative agreement with alpha systematics by observing the consistency of their nuclear radii, assuming the validity of the one-body theory for alpha decay. For the first treatment, the expression of Perlman and Ypsilantis[13] was selected because of its relative simplicity and relates the decay constant, decay energy, atomic number and nuclear radius as follows (see Bethe):[9]

$$\log_{10}\lambda = 21.843 + 0.5\log_{10}E - \log_{10}r + 0.217\,\frac{4}{A-4} - \frac{1.104\,(Z-2)}{\left\{E\left[1+(4/A-4)\right]\right\}^{1/2}}\,(\alpha_o - \sin\alpha_o\cos\alpha_o)$$

with

$$\cos\alpha_o = 0.5893\,[Er/(Z-2)]^{1/2}$$

The numerical coefficients were calculated to give λ in units of $sec.^{-1}$ with E (Mev) the alpha decay energy, r (cm x 10^{13}) the radius of the product nucleus, while Z and A are the charge and mass number, respectively, of the emitting nucleus.

For a second treatment, the more exact expression of Preston was used:[14]

$$\lambda = \frac{2v}{r_o}\,\frac{\mu^2 \tan\alpha_o}{\mu^2 + \tan^2\alpha_o}\,\exp\left[-\frac{8\epsilon^2 Z'}{\hbar v}(\alpha_o - \sin\alpha_o\cos\alpha_o)\right]$$

λ in units of $sec.^{-1}$.
v = alpha velocity = velocity of alpha particle $\left(1 + \frac{m_\alpha}{m_n}\right) = \left(\frac{2E}{m}\right)^{1/2}$.
r_o = radius of product nucleus.
m = reduced mass of system; m_α = mass of alpha particle; m_n = mass of recoil nucleus.
$\mu^2 = 0.52/E$.
α_o = arc cos $(mv^2 r_o/4\epsilon^2 Z')^{1/2}$.
Z' = charge of product nucleus.
$\epsilon = 4.8025 \times 10^{-10}$ esu.
E = total alpha disintegration energy = energy of alpha particle + energy of recoil nucleus + energy of orbital electron rearrangement.

This formula reduces to the more simple form:

$$\log\lambda = 22.143 + 0.5\log E - \log r_o + 0.5\log\left(\frac{m_n + m_\alpha}{m_n}\right) + \log\frac{\frac{0.52}{E}\tan\alpha_o}{\frac{0.52}{E} + \tan^2\alpha_o} - \left[\frac{Z' r_o m_n}{m_n + m_\alpha}\right]^{1/2}[\text{anti-log } 9.80956 - 10][\gamma]$$

λ in units of $sec.^{-1}$.
E = total alpha disintegration energy in Mev.
r_o = radius of the product nucleus (cm x 10^{13}).
Z' = charge of product nucleus.
m_n = mass of recoil nucleus.
m_α = mass of the alpha particle.
α_o = arc cos $[Er_o/Z']^{1/2}$ [anti-log 9.77035 - 10].
$\gamma = \left[\frac{\alpha_o}{\cos\alpha_o} - \sin\alpha_o\right]$, tabulated as $f(\sin\alpha_o)$ in UCRL-1473.[15]

Calculations of the nuclear radius were made for nine even-even nuclides, each having two recognized alpha groups. Table VI shows the calculated radii for these nuclides together with the alpha particle energies and partial alpha half-lives used. The values of r_o, progressively decreasing as the region Z = 96, A - Z = 148 is approached, may be indicative of a closed shell structure attained at Cm^{244}. Further investigation of the complex alpha structure in nuclides of higher Z should clarify this point.

For all the even-even nuclides, the decay to the excited level appears somewhat hindered. The hindrance factors listed in Table VI were calculated using the Preston formula,[14,15,16] assuming zero spin change for decay to both the ground and excited levels and assuming that the decay to the ground state is not hindered. However, Goldhaber and Sunyar assert that, for even-even nuclides, alpha decay to the first excited level involves a spin change of two.[17]

If the Preston formula is applied rigorously, taking this spin change into consideration, then the half-life so calculated for decay to the first excited level is shortened approximately 60 percent from that calculated without regard for the spin change. If this consideration had been made in using the Preston formula, the hindrance factors of Table VI would have been further increased by approximately 60 percent.

In examining all results, one should consider the possible errors in the two experimentally determined parameters, the decay constants (dependent upon the abundance of each group), and the decay energy. A 10 percent error in the experimental half-life determination would change the radius of both alpha groups by about 0.25 percent. An increase of the

abundance of the lower energy group of any isotope by 3 percent would increase the radius calculated for this group by ~0.03 while decreasing the radius of the higher energy group by ~0.01. An increase of 0.02 Mev in decay energy would decrease the radius of any group by nearly 1 percent.

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Table VI

Nuclide	Experimental Half-Life	Alpha Decay Energy (Mev)	Partial Alpha Half-Life	Calculated Radii (cm x 10^{13}) Perlman-Ypsilantis	Preston	$r_o = r/A$	Hindrance Factor = $\frac{T_{1/2 exp}}{T_{1/2 calc}}$
Cm^{244}*	~20 y	5.939	26.7 y	8.86	9.29	1.495	
		5.898	80 y	8.72	9.15		1.81
Cm^{242}	162.5 d	6.253	211 d	8.86	9.32	1.504	
		6.208	706 d	8.70	9.16		1.97
Pu^{238}	89.6 y	5.625	116.5 y	9.89	9.33	1.514	
		5.585	390 y	8.73	9.17		1.96
Pu^{236}	2.7 y	5.89	3.38 y	8.93	9.38	1.527	
		5.84	13.5 y	8.75	9.20		2.11
U^{238}	4.49×10^9 y	4.311	5.75×10^9 y	9.07	9.43	1.530	
		4.266	20.4×10^9 y	8.98	9.34		1.43
U^{236}	2.39×10^7 y	4.615	3.28×10^7 y	8.96	9.34	1.520	
		4.565	8.85×10^7 y	8.93	9.31		1.11
U^{234}	2.48×10^5 y	4.888	3.31×10^5 y	8.96	9.36	1.528	
		4.838	9.92×10^5 y	8.89	9.29		1.31
U^{232}	~70 y	5.44	100 y	8.95	9.39	1.537	
		5.38	233 y	8.94	9.38		1.05
Th^{232}	1.39×10^{10} y	4.134	1.83×10^{10} y	8.94	9.31	1.524	
		4.079	5.79×10^{10} y	8.94	9.29		1.07

*Spectrometer data of Asaro.[18]

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LIST OF ILLUSTRATIONS

1. Pu^{236} alpha activity illustrating varying sensitivities of D-1, C-2, G-5 emulsions.
2. Loading activities in emulsion.
3. Desicating and developing nuclear plates.
4. Graph of range vs energy for alpha particles in nuclear emulsions.
5. Graph of range vs energy for electrons in nuclear emulsions.
6. Bismuth fraction decay curve.
7. Bi^{203} alpha histogram.
8. Bi^{203} alpha particles originating from areas of high electron capture activity.
9. $Cm^{238} \xrightarrow{\alpha} Pu^{234} \xrightarrow{\alpha} U^{230}$ as recorded within nuclear emulsion.
10. Photomicrograph illustrating conversion electrons in coincidence with alpha particles.

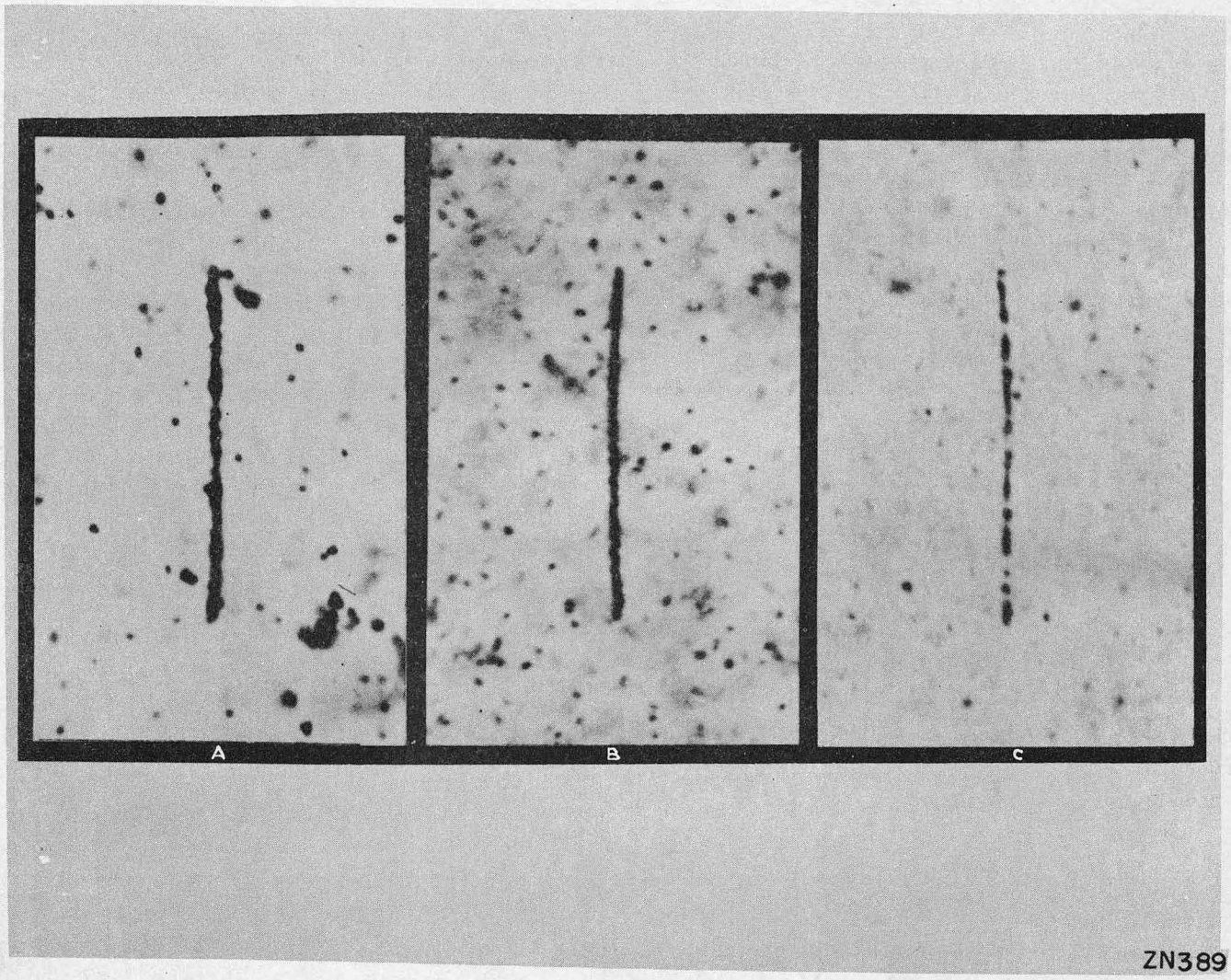


Fig. 1

Pu_{236} alpha activity as recorded in Ilford
A. G-5 (observe 25 kev coincident conversion electron)
B. C-2
C. D-1
nuclear emulsions. All developed with 1:6 D-19 at
20° C. for 45 minutes.

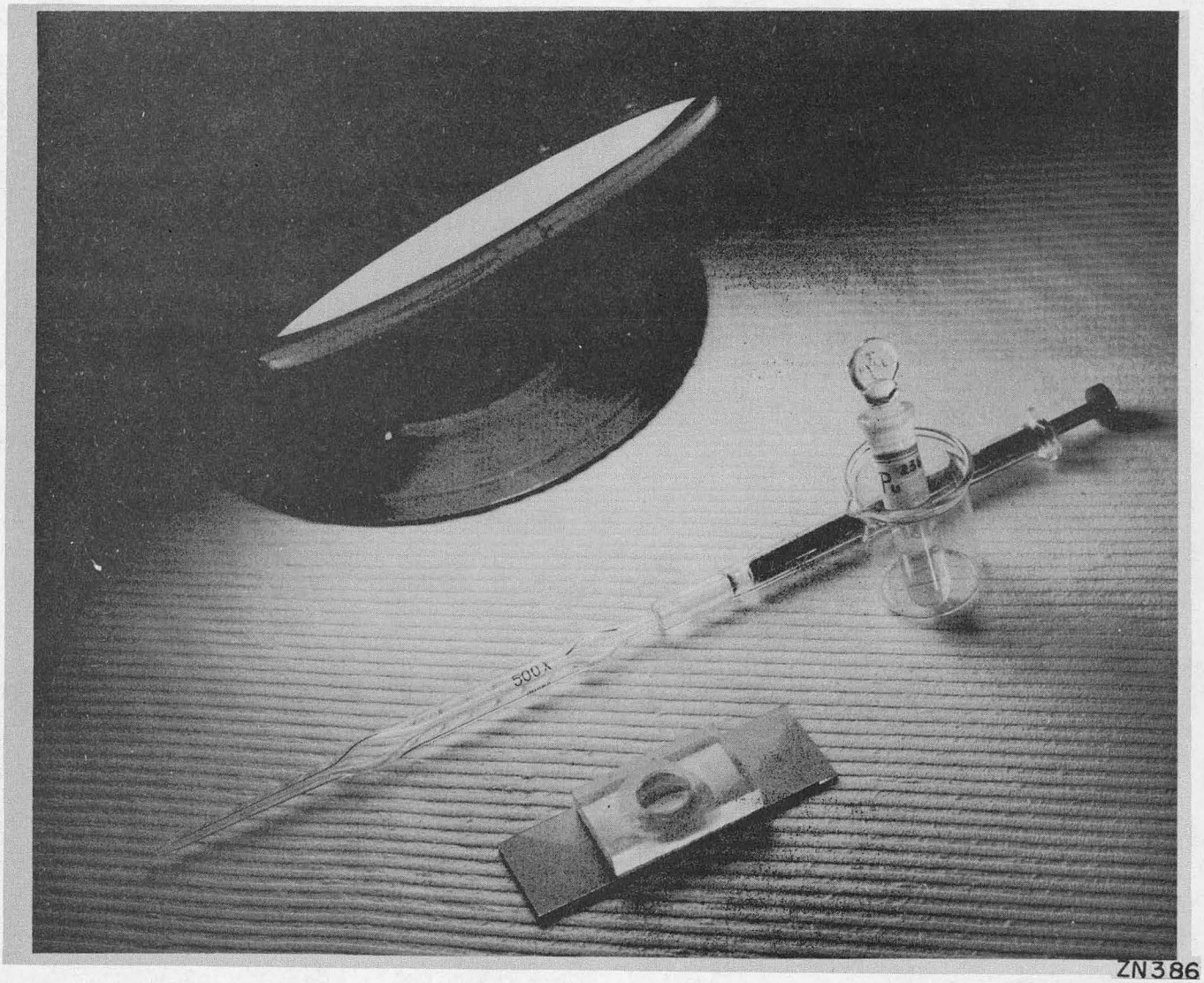


Fig. 2

Radioactive solution column supported over emulsion provides good impregnation of limited activities.

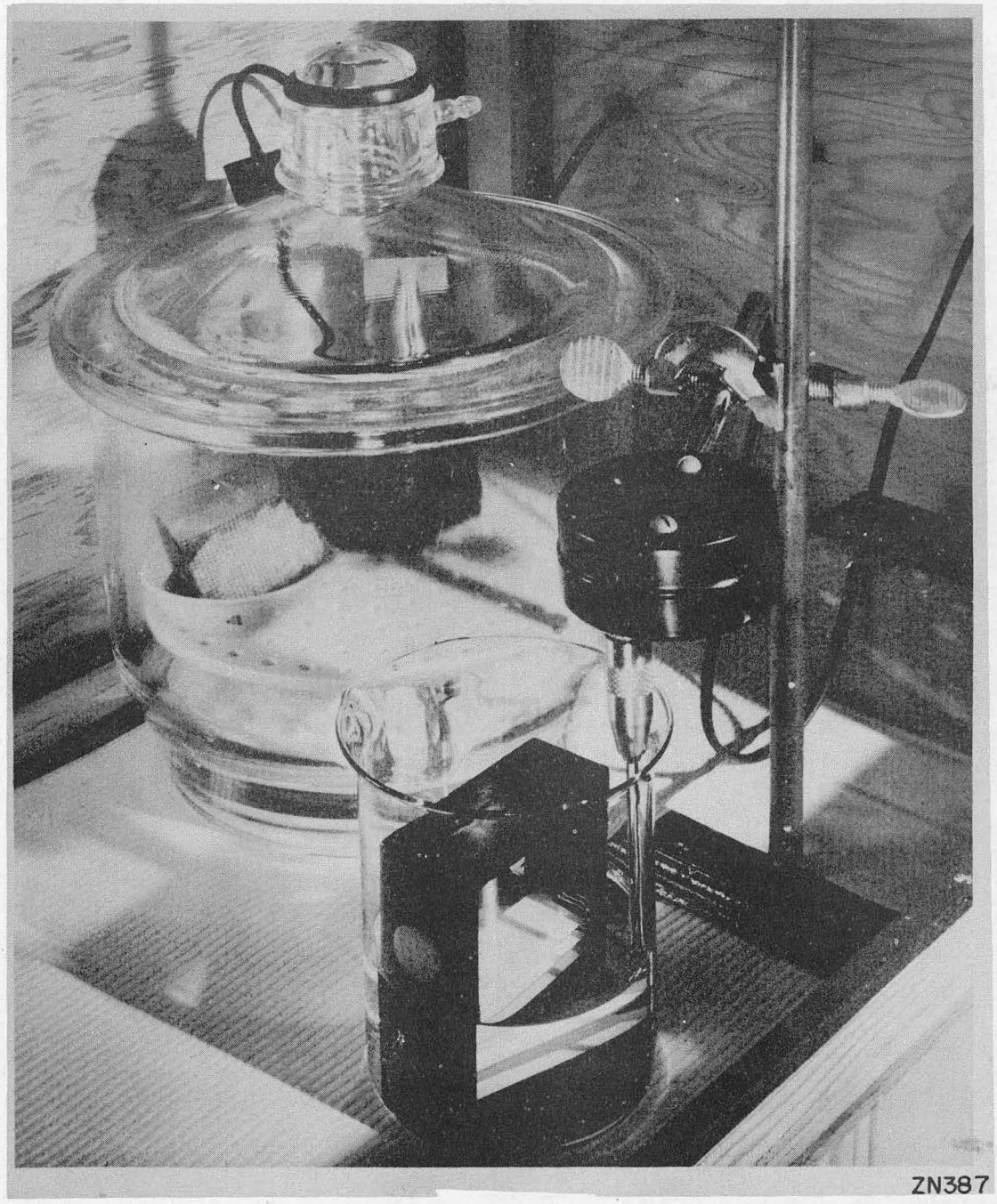
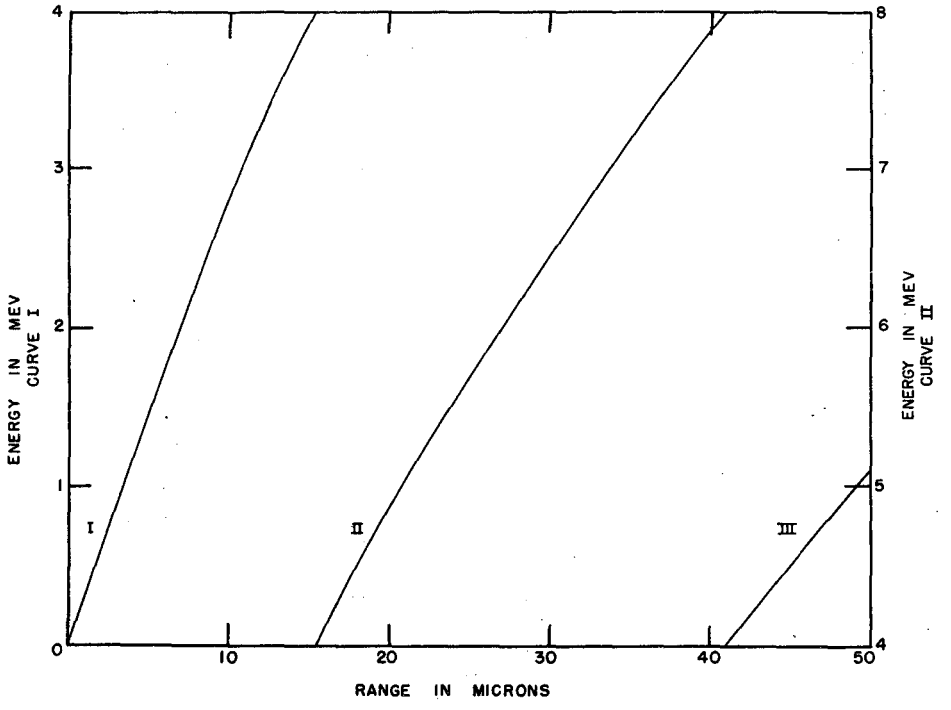


Fig. 3

Rear - NaOH desiccator with circulation for rapid drying of plates.

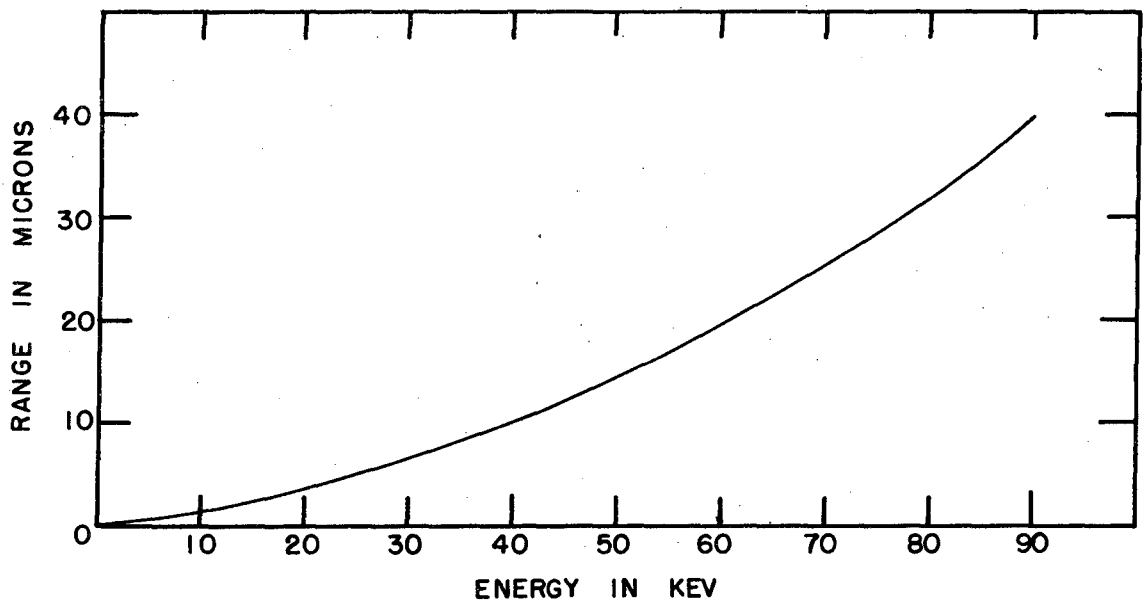
Front - Transfer of plate holder between beakers containing developing and fixing solutions provides simple but excellent processing.



RANGE vs. ENERGY
ALPHA PARTICLES IN ILFORD C-2
EMULSIONS

MU4041

Fig. 4



RANGE vs. ENERGY

ELECTRONS IN
ILFORD G-5 EMULSIONS

MU 40 40

Fig. 5

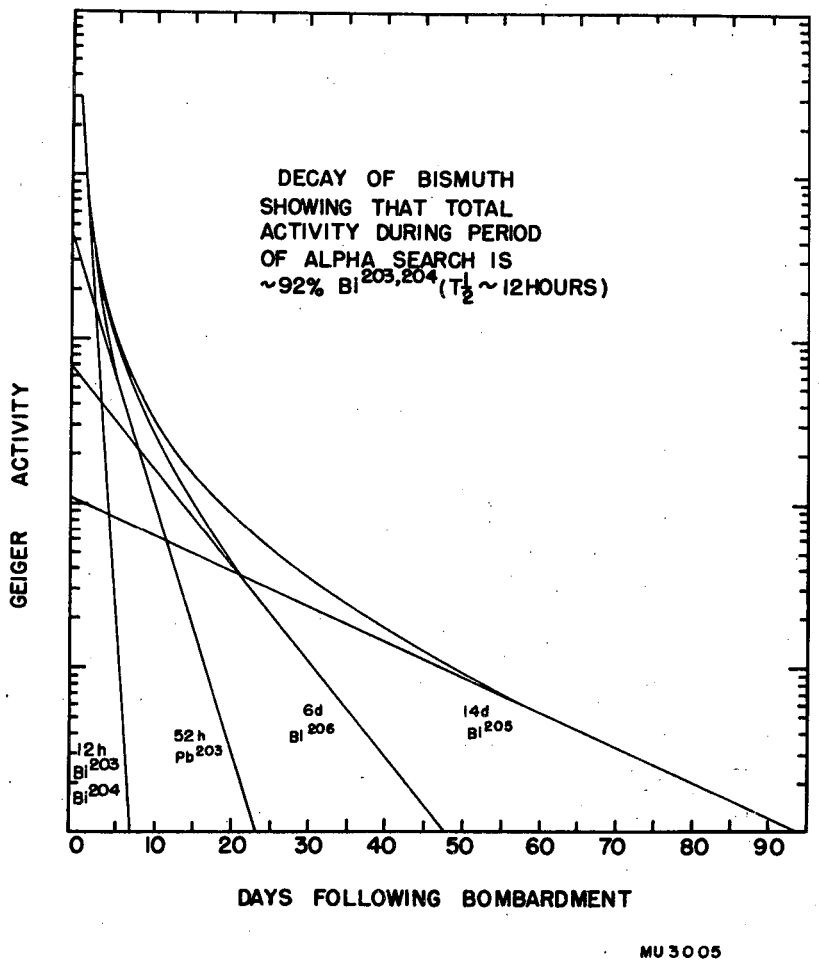

DECAY OF BISMUTH
SHOWING THAT TOTAL
ACTIVITY DURING PERIOD
OF ALPHA SEARCH IS
~92% Bi203,204 (T½ ~ 12 HOURS)
GEIGER ACTIVITY
12h
Bi203
Bi204
52h
Pb203
6d
Bi206
14d
Bi205
0
10
20
30
40
50
60
70
80
90
DAYS FOLLOWING BOMBARDMENT
MU3005

Fig. 6

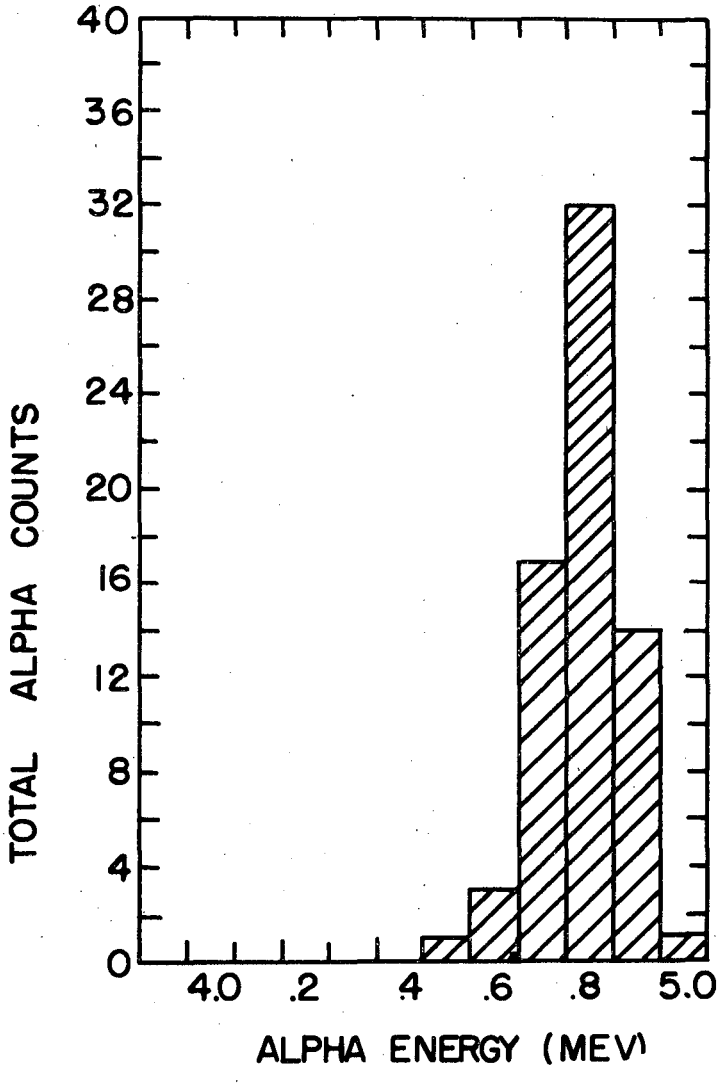

40
36
32
28
24
20
16
12
8
4
0
TOTAL ALPHA COUNTS
4.0
.2
.4
.6
.8
5.0
ALPHA ENERGY (MEV)
MU3004

Fig. 7

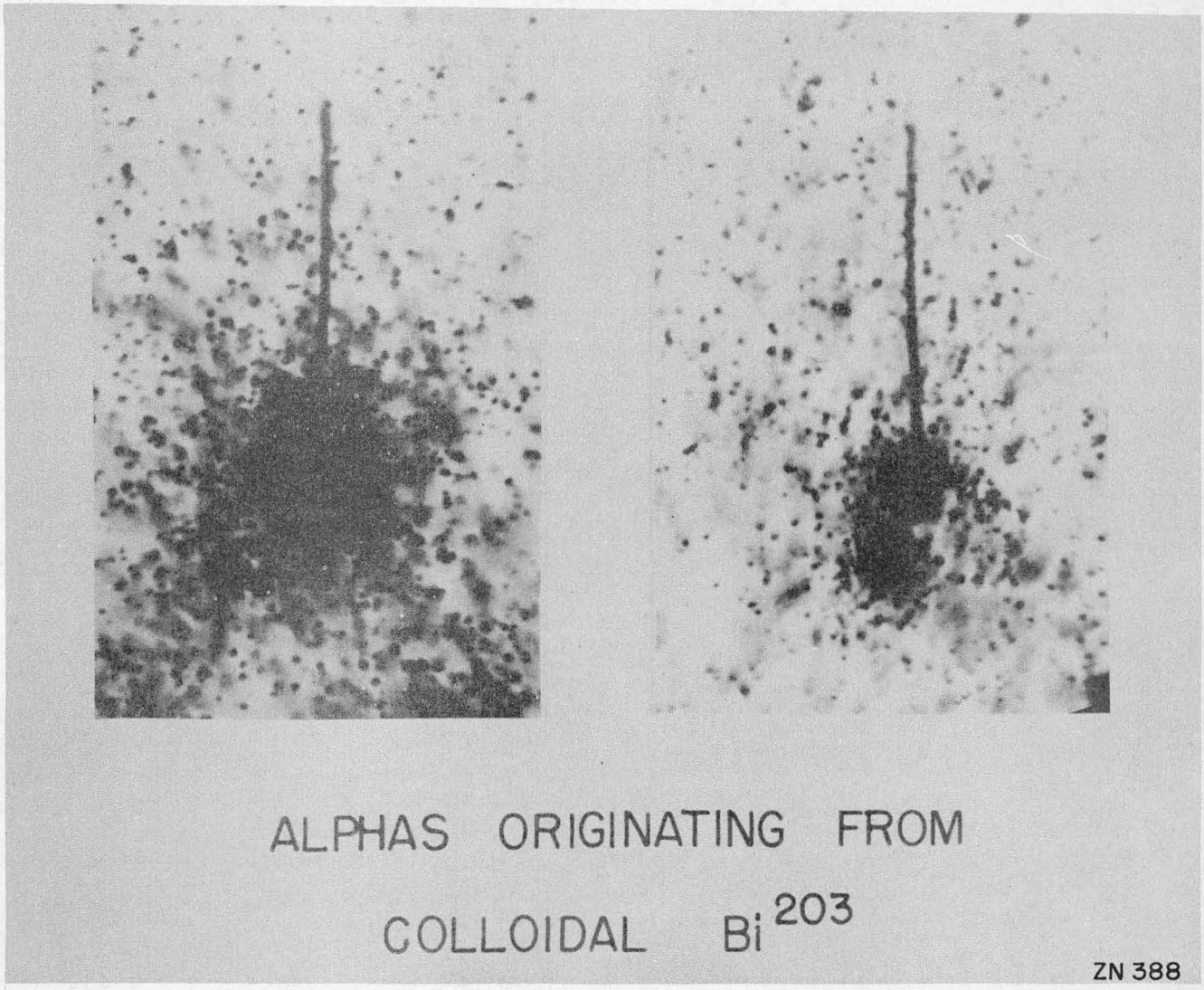


Fig. 8

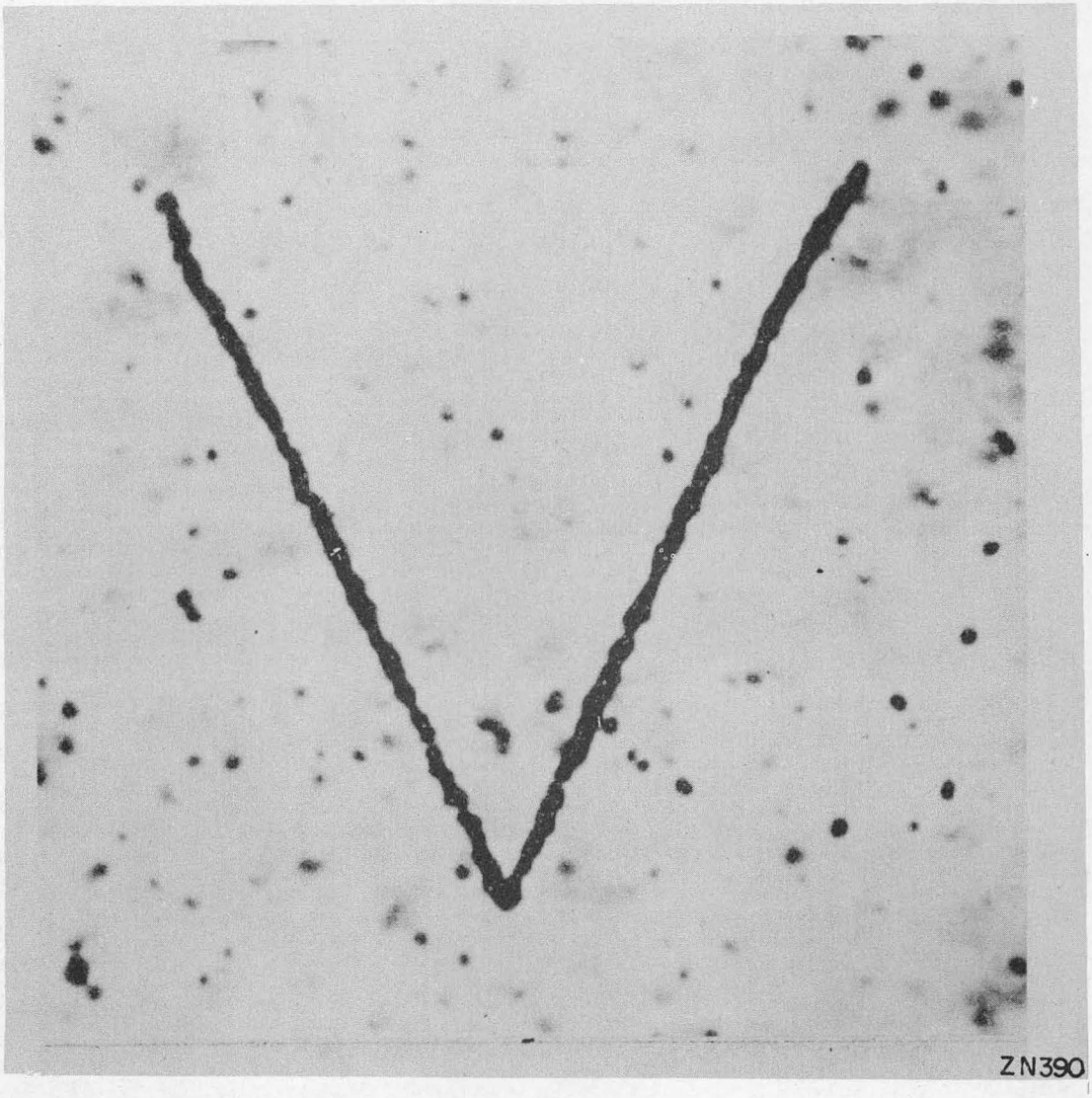


Fig. 9

$$Cm^{238} \xrightarrow{\alpha} Pu^{234} \xrightarrow{\alpha} U^{230}$$

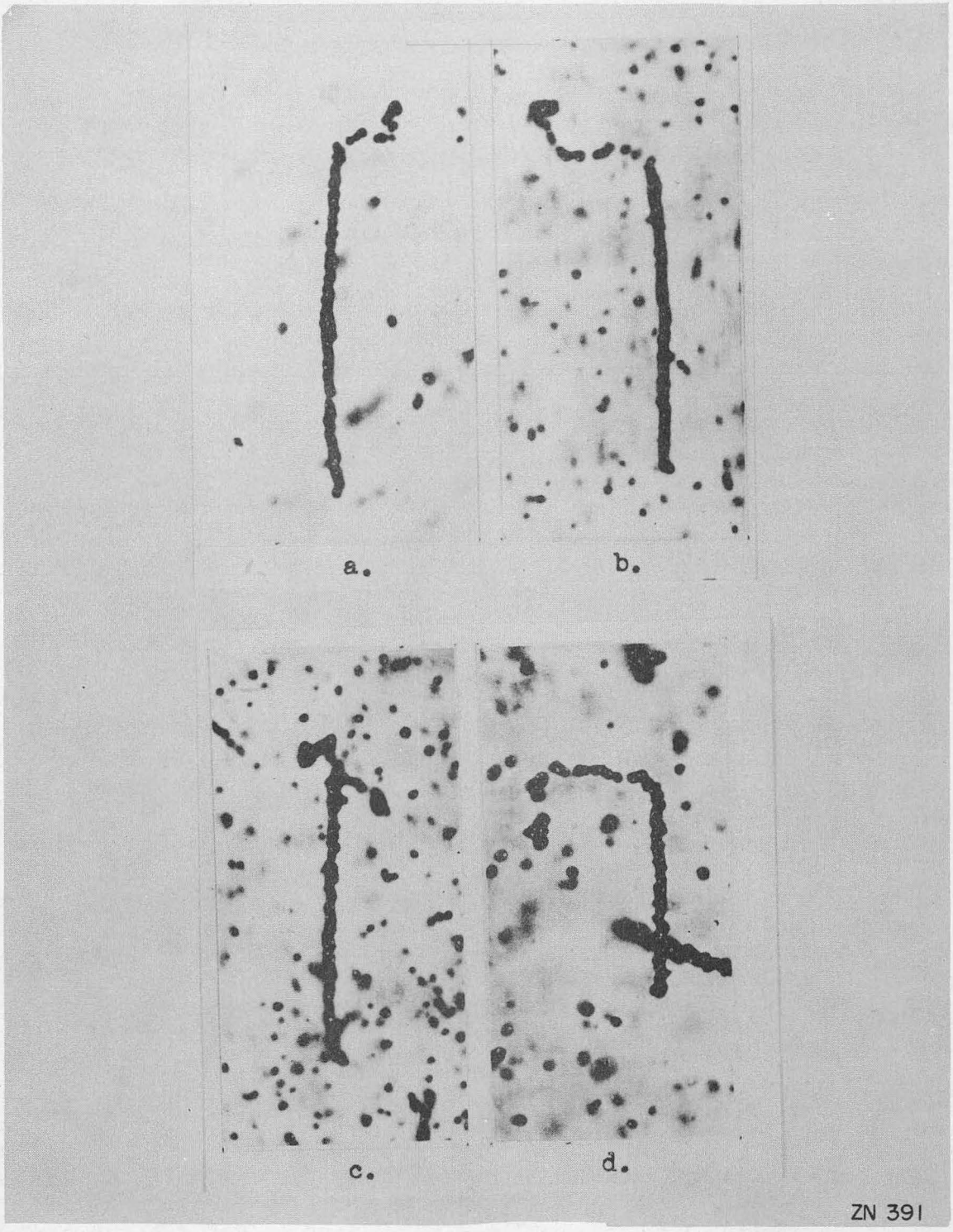


Fig. 10

Conversion electrons from

a. Cm^{242}

b. Am^{241}

c. Am^{241} (two electron coincidence)

d. Th^{232}