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ABSTRACT

Several isotopes of the new element 95 have been produced and their radiations characterized. The chemical properties of this tripositive element are similar to those of the typical tripositive lanthanide rare-earth elements. Element 95 is different from the latter in the degree and rate of formation of certain compounds of the complex ion type, which makes possible the separation of element 95 from the lanthanide rare-earths.

The name americium (after the Americas) and the symbol Am are suggested for the element on the basis of its position as the sixth member of an actinide rare-earth series, analogous to europium, Eu, of the lanthanide series.

The isotopes found and studied in this work are: (1) Am^{241}, which decays by the emission of alpha-particles (energy-5.45 Mev) with a 510 ± 20 year half-life and is produced by the beta-decay of Pu^{241}, which, in turn, is produced by the (α,n) reaction on U^{238}; (2) Am^{242}, which decays by the emission of beta particles (ca. 0.8 Mev maximum energy) with a 17-hour half-life or, in another isomeric form, by branching decay with the emission of alpha-particles (energy unknown) and beta-particles (ca. 0.5 Mev maximum energy) in the ratio ca. 0.002 alpha-particles per beta-particle; both isomers are produced by neutron capture in Am^{241}; (3) Am^{239}, which undergoes branching decay, decaying (a) by orbital electron capture with a 12 hour half-life and emitting 0.285 Mev gamma rays and conversion electrons in addition to the characteristic x-rays and (b) by alpha particle emission (energy unknown) in the ratio ca. 0.001 alpha-particles per electron capture. This isotope is produced by the (d,2n) reaction on Pu^{239} and by the (α,2n) reaction on Np^{237}; (4) Am^{238}, which decays by orbital electron capture with a 50-hour half-life emitting 1.3-1.4 Mev gamma rays and conversion electrons in addition to the characteristic x-rays. Am^{238} is produced by the (d,3n) reaction on Pu^{239} and by the (α,3n) reaction on Np^{237}.

1. Introduction

Isotopes of the element with atomic number 95 have been produced and identified in experiments carried out with material activated in sixty-inch cyclotron of the University of California. The target materials which have been successfully used in the production of these isotopes are U^{238}, Np^{237} and Pu^{239} The energetic helium ion bombardment of U^{238} leads to the formation of plutonium isotopes of mass numbers

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236 to 241, of which Pu^{236}, Pu^{238} and Pu^{239} are known to be beta-stable. Pu^{241} is shown in this work to be unstable to the emission of beta-particles, leading to the production of 95^{241}. The helium ion bombardment of Np^{237} and the deuteron bombardment of Pu^{239} are capable of forming isotopes of element 95 directly, the expected mass numbers being 235 to 240. Of these, 95^{238} and 95^{239} should be formed in the highest yields.

At the beginning, in the search for activities due to isotopes of element 95, it was assumed that the chemical properties would be similar to those of the lanthanide elements in the (III) oxidation state. It has been pointed out by Seaborg(1) that the chemical properties of the elements following actinium (element 89) in the periodic system may be explained on the assumption that they constitute a rare-earth-like series (actinide series) in which the 5f shell of electrons is in the process of completion. On this basis it was predicted that the increasing stability of the (III) state of the actinide elements should culminate in very stable (III) states in elements 95 and 96.

The first positive evidence for the existence of element 95 was found in the late fall of 1944, in the form of nuclear and chemical data pertaining to the isotope 95^{241}. It is suggested that the new element be named americium (in honor of the Americas) and have the symbol Am. This name is based on the strong analogy between element 95 and europium (after Europe), Eu, of the lanthanide rare-earth series.

II. Am^{241} and Related Isotopes

A. Helium-ion Bombardment of U^{238}.

Uranium in which the isotopic content of U^{235} was reduced by the electromagnetic process(2) was bombarded, in the sixty-inch cyclotron at Berkeley, with helium ions of ca. 38 Mev energy. Such bombardments are discussed in more detail in another paper(3). The activated uranium was milled from the target plates in layers of ca. 100 mg.cm.$^{-2}$. Each of the layers was processed separately to yield radiochemically pure plutonium fractions. Standard alpha and beta-particle measurements with the purified plutonium samples revealed the presence of no radiations which could not be accounted for on the basis of the radiations from previously known plutonium isotopes. Investigation of the very low energy beta spectrum, however, indicated the presence of beta-particles with ca. 20 kev maximum energy. The measurements were made in an apparatus designed by Raynor(4), in which window and gas absorption of the particles to be counted is reduced to ca. 300 micrograms per cm^2. In this case provision was made for absorption measurements using a limited number of absorbers made from thin calibrated films of cellulose nitrate. The beta-particle range was estimated visually on a semilogarithmic plot of the counting data to be 600-800 micrograms per cm^2, which corresponds to 20 kev maximum energy as obtained from the range-energy data of Schonland(5) for low energy electrons. Absorption curves for the 20 kev beta-particle component of the plutonium activities from the first and third layers of the activated uranium target are given in Figure 1, in which beta-particle intensities are normalized to the Pu^{239} alpha-particle activities in each sample. Thus, the lower intensities shown for the first layer indicate that the yield of the isotope responsible for the beta-particle activity increases relative to the yield of Pu^{239} as the depth of penetration of the helium ion increases. From the data compiled by Livingston and Bethe(6) on the relative stopping power of various elements for helium ions, the maximum energy of the ions in the first and third uranium layers was calculated to be 38 Mev and 28 Mev, respectively. At the lower energy, the ratio of yields, (α, 3n) to (α,3n) should be greater than at a higher energy.

Fig. 1

On this basis, the decrease in 20 kev beta-particle activity relative to Pu^{239} alpha-particle activity is an indication that it is due to an isotope resulting from the (α,n) or (α,2n) reaction:

$$U^{238}(\alpha,n)Pu^{241}$$

$$U^{238}(\alpha,2n)Pu^{240}$$

Of these, Pu^{241}, with an odd number of neutrons, is the most probable source of beta-particle activity.

An estimate of the half-life for Pu^{241} beta-particle emission may be made with the following observations: (1) the beta activity of curve I, Figure 1, extrapolated to zero absorption as shown, is 1550 counts per minute, (2) in the same sample and at the same counting geometry there are 54 counts per minute of alpha-particles due to Pu^{239}, (3) the half-life for alpha particle emission of Pu^{239} is 24300 years, and (4) the yield from the (α,n) reaction relative to that from the (α,3n) reaction is usually ca. 0.01 in the 38 Mev helium ion bombardment of heavy isotopes. From these considerations the half-life of Pu^{241} for beta particle emission is ca. 10 years.

The rare earth fraction from a similarly activated uranium sample contained an alpha-particle activity (energy - 5.45 Mev) of long half-life. Numerous tracer chemical experiments which were carried out with such activity are reported in another paper.(7) The evidence obtained shows conclusively that the activity is due to a previously unknown element. The occurrence of relatively energetic alpha-particle emission in a rare-earth fraction (lanthanum fluoride carriable) may be considered sufficient evidence, of itself, for the presence of an isotope of a rare-earth-like heavy element, since alpha-particle emission is an extremely rare property in isotopes of atomic number less than 81. The direct formation of americium isotopes by the helium ion bombardment of U^{238} is not possible; therefore, the presence of the alpha-particle activity must be considered evidence for the formation of Am^{241} as the product of beta-particle emission by Pu^{241}.

Several samples known to contain appreciable amounts of Pu^{241} were carefully purified to remove all traces of rare-earth, and rare-earth-like, activities, then were allowed to stand for long periods of time. When "rare-earth" fractions were again removed from the samples, the alpha activity previously observed was again found, having grown into the samples from the plutonium source. Standard samples of the previously known plutonium isotopes were treated in a similar manner, but failed to yield a comparable alpha activity. Samples of the alpha activity were removed again and again from the plutonium samples, resulting in the observations: (1) the rate of formation of the alpha particle activity was constant over a period of several years, due to the long half-life of the parent isotope, (2) the yield from a given sample was a linear function of the time allowed for growth, and (3) the amount of growth in similar periods of time was directly dependent upon the intensity of 20 kev beta-particles, i.e., the amount of Pu^{241}, in the plutonium samples. This evidence proves that the alpha-activity is due to Am^{241} arising from the beta-particle emission of Pu^{241}.

Several samples of Am^{241} were irradiated with thermal neutrons over a long period of time. The principal product observed was an isotope of curium (element 96), Cm^{242} (8) as determined by the half-life (ca. 5 months) and the alpha-particle energy (6.1 Mev). Separation of the curium and americium activities was later achieved by the use of a Nalcite (Dowex 50) resin column with selective elution in ammonium citrate solution.(9) The Cm^{242} was formed by the reaction:

$$Am^{241}(n,\gamma)Am^{242}$$

followed by rapid negative beta-particle decay of Am^{242}. In a later irradiation, carried out in the Argonne heavy water pile, Manning and Asprey[10] detected the beta-particles from Am^{242} and found the half-life to be ca. 17 hours. They also demonstrated the growth of Cm^{242} alpha-activity with the same 17-hour half-life. A further discussion of Am^{242} is given in the next section.

All of the experiments with the 5.45 Mev alpha-activity are consistent with its assignment to Am^{241}. The isotope results from the beta-decay of Pu^{241}; thermal neutron irradiation of the material results in the formation of a beta-active isotope which decays to Cm^{242}, which in turn decays to Pu^{238}, a well known isotope of plutonium[8].

Samples of plutonium analogous to those in which Am^{241} growth was observed were processed to yield radiochemically pure uranium fractions by an oxidation-reduction method employing nitric acid oxidation of uranium in sulfuric acid solution, precipitation of PuF_4 and carrier LaF_3, then titanous chloride reduction and LaF_3 precipitation to remove uranium from the solution. The uranium fractions were found to contain a beta-activity of 6.8-day half-life, corresponding to U^{237}, which could be formed as a result of alpha-decay of Pu^{241}. The yield of the activity (which was present in the plutonium at its equilibrium value) was compared with the yield of Am^{241} from the same plutonium sample to give a value for the branching ratio of Pu^{241}(alpha disintegrations per beta disintegration) of ca. 2×10^{-5}.

B. Chemical Properties of Americium.

A large number of tracer chemical experiments were carried out with the 5.45 Mev alpha activity and are described in detail in another paper.[7] It is of considerable interest, however, that the unique chemical nature of americium may be shown by a consideration of a relatively few experiments:

(1) The activity coprecipitates with rare-earth fluorides from strongly oxidizing solutions, such as 0.1 M $K_2Cr_2O_7$ in 1 M HNO_3 solution, and Ag^{++} with $(NH_4)_2S_2O_8$ in 2 M HNO_3 solution. Those elements, without regard to the plausibility of their formation in the nuclear processes described, which are not eliminated from consideration on the basis of this are: scandium, yttrium, indium, lanthanum, the rare-earth elements, actinium, thorium and possibly protactinium and thallium.

(2) Among the more logical possible alpha particle emitters to be considered, we may eliminate thallium, lead, bismuth and polonium by consideration of the fact that the activity does not coprecipitate with bismuth sulfide from 0.25 N HCl solution.

(3) Thorium peroxide does not carry the activity, under conditions in which thorium precipitates quantitatively, thus eliminating thorium.

(4) Actinium tracer activity and the activity in question may be fractionated by coprecipitation with zirconium or ceric iodate from 0.035 M potassium iodate - 1 N HNO_3 solution, the actinium tracer carrying to a greater extent.

(5) The activity may be separated from tracer or macro amounts of the rare-earth elements by the precipitation of a lanthanum compound of undetermined composition from 1 M ammonium fluosilicate - 5 M HNO_3 solution. The alpha activity remains largely in solution while the rare-earth elements are almost completely precipitated under these conditions.

(6) The activity may be separated from curium activity by selective elution with ammonium citrate solution from columns of resin, such as Amberlite IR-1, or Nalcite (Dowex-50). Curium is removed more easily.

This chemical evidence, with the nuclear evidence previously given, establishes beyond any reasonable doubt that the activity is due to an actinium-like transplutonium element, americium.

C. Nuclear Radiations of Am^{241}.

The disintegration of Am^{241} is accompanied by the emission of alpha-particles and some associated electromagnetic radiation. The energy of the alpha-particles has been measured by two methods: (1) absorption in thin mica sheets of known thickness and (2) differential analysis, by electronic means, of the pulses produced by the alpha-particles in an ionization chamber[11] (Figure 2). In both methods the energy was obtained from direct comparison with standard samples of other alpha activities of known energies. The values from the two methods check closely at 5.45 ± 0.05 Mev. In the mica absorption method the observed value is in terms of the range in air which is converted to energy in Mev by use of the range-energy relation given by Holloway and Livingston[12]. <Fig. 2

Absorption measurements on the electromagnetic radiations are shown graphically in Figures 3, 4, and 5. The intensities shown are for a sample containing 10^6 disintegrations per minute of alpha-particle activity. The apparatus used to detect the radiations was a bell jar type Geiger counting tube, 1-1/8 inches in diameter and 2-5/8 inches long. The window was of mica, ca. 3 $mg.cm.^{-2}$ thick and the body of the tube was copper. The tube was filled with a 90% argon-10% ethanol mixture to 10 cm. Hg pressure. The cathode was a 5 mil tungsten wire terminating in a small glass bead 3/16 inch from the mica window. Samples were counted in a position to have a 10% geometry factor. <Fig. 3, 4, and 5

The electromagnetic radiations observed are seen to be of two classes: (1) a 62 kev component, the counting efficiency for which is about 0.5 percent under the above conditions, and (2) a complex mixture of radiations apparently covering the range 10 to 20 kev which have the absorption characteristics of neptunium L x-radiation. The counting efficiency for the latter is not readily calculated, but is probably ca. 1.5 percent under the above conditions. The aluminum absorption curve expected for the mixture of neptunium L x-rays was constructed from an extrapolation of known L x-ray energies tabulated by Compton and Allison[13] and the relative intensities given for uranium by Allison.[14]

Using the counting efficiencies assumed in the above discussion the neptunium L x-rays are emitted in ca. 100 percent of the alpha disintegrations and the 62 kev gamma rays, in ca. 60 percent.

Internal conversion of a gamma-ray whose energy is insufficient to excite the K electrons (binding energy, ca. 120 kev) may take place in the L shell where the binding energy is much less (ca. 24 kev), if enough energy is available. The subsequent occupation of the vacant L electron state by another electron is accompanied by the characteristic L x-radiation of the product element. The energy of the electrons in the case of the conversion of 62 kev gamma rays is about 35 kev. Such electrons are not energetic enough to pass through the 3 $mg.cm.^{-2}$ mica window of the Geiger tube described above and would not have been detected.

The half-life of americium has been determined by Cunningham[15] to be 510 $\pm$ 20 years. The value is based on specific activity measurements carried out on the ultra-microchemical scale.

III. Nuclear Properties of Am^{242}

The production of Am^{242} by the thermal neutron irradiation of Am^{241} has been mentioned in Section II. The existence of the isotope was first indicated by the formation of an isotope of curium (at. no. 96), Cm^{242}, under conditions expected to result in the formation of Am^{242}, i.e., the radiative capture of thermal neutrons in Am^{241}. The emission of negative beta particles, in Am^{242}, with a half-life short compared to the total time of irradiation, results in the observation given above.

The first detection and half-life measurement of the actual process of beta particle decay of Am^{242} was achieved by Manning and Asprey in samples investigated soon after termination of irradiation in the Argonne pile.(10) A decay curve obtained in recent experiments at this laboratory is given in Figure 6 for the Am^{242} beta particles. A long-lived background activity is present due to the electromagnetic radiations from Am^{241} and the radiations arising in a long-lived isomer of Am^{242} to be described later. The predominant activity due to 17-hour Am^{242} consists of the beta particles whose aluminum absorption characteristics are shown graphically in Figure 7. <Fig. 6 <Fig. 7

A sample of thermal-neutron-activated Am^{241} was carefully purified and allowed to stand for several months. At the time of purification the 17-hour Am^{242} should have been completely gone from the sample. A combined plutonium and neptunium fraction was then removed from the sample and purified by oxidation-reduction cycles. Samples of the neptunium-plutonium fraction were observed to contain beta-particle activity which decayed with a 2.0 day half-life. The aluminum absorption characteristics of the beta particles were entirely consistent with those of Np^{238} (16) whose half-life is 2.0 days (Figure 8). <Fig. 8

Np^{238} is a "shielded" isotope in the sense that it is not produced by either negative beta particle emission or by orbital electron capture, the hypothetical parents of Np^{238} by these processes being Pu^{238} and U^{238}, respectively. Each of these is stable with reference to the process by which it would produce Np^{238}. Since the Np^{238} was observed to grow in the irradiated americium sample, a long-lived Am^{242}, decaying by the emission of alpha particles must be responsible:

$$Am^{242} \xrightarrow[\text{long}]{\alpha} Np^{238}.$$

After allowing time for restoration of the Am^{242}-Np^{238} equilibrium a second separation was made in which the results were the same as in the first. The 2.0-day half-life of Np^{238} is evidently short compared to that of the long-lived Am^{242} and the equilibrium value of the Np^{238} activity is a measure of the alpha activity in Am^{242}. If it is assumed, entirely for the sake of discussion with no thought that this need be true, that the cross-section for the formation of the long-lived Am^{242} is the same as that for the 17-hour isomer, the half-life for alpha-particle emission is calculated to be ca. 3×10^5 years on the basis of the observed Np^{238} activity.

An aluminum absorption curve of the radiations from the activated americium sample reveals the presence of beta particles of ca. 0.5 Mev maximum energy (Figure 9). The other radiations in the sample are essentially those due to Am^{241}. In view of the beta instability of the 17-hour isomer of Am^{242}, beta particle emission in the long-lived isomer is not surprising. Again arbitrarily assuming equal cross-sections for the formation of both isomers the half-life of the long-lived Am^{242} for the emission of beta particles is ca. 600 years. This half-life and that calculated for the emission of alpha particles are directly proportional to the assumed cross-section and a reduction of the latter would decrease the calculated half-lives.

< Fig. 9

IV. Other Isotopes of Americium

Americium isotopes of mass number equal to or less than 239 are expected to decay by the capture of orbital electrons, since their nuclei are deficient in neutrons. Am^{240}, by virtue of its position close to the region of maximum stability might decay either by the emission of negative beta-particles or by orbital electron capture, or by both processes. Branching decay with alpha-particle emission is possible in any of the isotopes and may be found if the half-lives of the competing processes are appropriate.

A. Deuteron Bombardment of Pu^{239}.

Several targets of Pu^{239} were bombarded with 19 Mev deuterons in the sixty-inch cyclotron in order to produce americium isotopes of mass numbers 237 to 240. The targets were prepared by the evaporation of slurries of plutonium(IV) fluoride on rectangular platinum plates of one cm.^{2} area (interceptor targets). After ignition in air, the samples were placed in the target chamber of the cyclotron in such a way as to intercept the most intense portion of the ion beam. As soon as possible after the end of bombardment, the samples were dissolved and processed chemically to yield rare-earth fractions by the successive precipitation of lanthanum(III) fluoride carrier from strongly oxidizing solutions. The actinide elements were separated from lanthanide elements by the precipitation of lanthanum(III) from 3 M HNO_3 - 1 M H_2SiF_6 solution.[7] Such precipitates are known to carry 90 percent of rare-earth activities and only 10 to 30 percent of actinide activities. Recovery of the actinides (in this case americium) was effected by the addition of sufficient concentrated HF to precipitate the remaining lanthanum which under these conditions carries the transplutonium activities. Cycles such as this were repeated until the activity had a constant composition with respect to all types of radiation and until the chemical yield per cycle was that expected of americium isotopes.

Decay measurements were made under four sets of counting conditions, using a thin window (ca. 3 mg.cm.$^{-2}$ mica) Geiger tube detector:

(1) 7 mg.cm.$^{-2}$ aluminum filter, to prevent the detection of alpha particles,

(2) 1500 mg.cm.$^{-2}$ beryllium filter, to allow passage of most electromagnetic radiation, but stop all except the most energetic beta particles,

(3) 1500 mg.cm.$^{-2}$ beryllium + 150 mg.cm.$^{-2}$ lead combined filter, to detect electromagnetic radiation of greater than about 200 kev energy, but not that of less than about 200 kev energy, and,

(4) 5 g.cm.$^{-2}$ lead filter, to present detection of all but energetic gamma radiation.

The difference in intensity observed under condition (2) and (3), the "differential count", was a measure of the low energy electromagnetic radiation (probably x-rays).

In rare-earth activities obtained in the thermal neutron irradiation of uranium or plutonium the activity measured by a "differential count" is only a few tenths percent of the activity measured through 7 mg.cm.$^{-2}$ aluminum. The activity observed in the rare-earth fractions of deuteron bombarded Pu^{239} often gave a "differential count" as high as one percent of the total. In general, this increased to 5 - 10 percent in the americium fractions resulting from the fluosilicate cycles.

Two half-life periods, 12 hour and 50 hour, were observed in the decay of the total activity and the activity measured by the "differential count" (Figures 10 and 11). Only the 50 hour period was found in the decay measured with a 5 g.cm.$^{-2}$ lead filter(Figure 12). At several times during the decay of the observed activities, absorption data were taken in order to identify and characterize the radiations due to each activity. Measurements of the electromagnetic radiations were made with a 1500 mg.cm.$^{-2}$ beryllium filter, to eliminate the beta or electron activity. Examples of the data, for the 50-hour activity are shown in Figures 13 to 15 (taken after complete decay of the 12 hour activity), and for the 12 hour activity, in Figures 16 to 18 (after subtraction of the radiations due to the 50-hour activity).

< Figs. 10 and 11

< Fig. 12

< Figs. 13, 14, 15

< Figs. 16, 17, 18

The curves were quite complex and the resolutions can only be approximate, especially in the case of the 12-hour activity. However, the electromagnetic radiations of each of the activities correspond rather closely to the expected L and K x-radiation of plutonium, with some additional gamma radiation. The electron activities in each of the isotopes are best explained as due to internal conversion of the gamma rays, and the respective energies are consistent with this interpretation. Relative intensities are limited in their accuracy to that of the values of the counting efficiency for each of the components. These values are not too well known for low energy electromagnetic radiation. The results obtained from several bombardments are given in Table 1. < Table 1

Measurements of the electron components were made using a strong, variable magnetic field to enable determination of values of $H\rho$. The device used was designed for high geometry and consequently the resolution was poor. However, the distribution observed for the low energy electrons of the 12 hour isotope was that characteristic of monoenergetic electrons of *ca.* 200 kev average energy. In all of the samples measured, the 1.2-1.3 Mev electrons were just detectable over the counter background, hence could not be studied with any degree of accuracy with the magnetic device.

Alpha particle decay measurements showed the presence of an alpha activity of 12-hour half-life. The branching ratio (alpha disintegrations per orbital electron capture) was found to be *ca.* 0.1 percent, corresponding to a partial alpha half-life of *ca.* 500 days. The alpha activity was always present in low intensity in the observed samples and no energy determinations were made.

Observed Radiations in Americium Fractions

	12-hour activity			50-hour activity		
Component	Observed relative activity	Assumed counting efficiency (percent)	Relative No. of events	Observed relative activity	Assumed counting efficiency (percent)	Relative No. of events
L x-radiation	2.7	1.5	0.9	2.8	1.5	0.9
K x-radiation	1.0	0.5	1.0	1.0	0.5	1.0
285 kev gamma radiation	0.4	0.5	0.4			
1.3-1.4 Mev gamma radiation				2.3	1.2	0.95
200-240 kev electrons	38.3	100	0.2			
1.2-1.3 Mev electrons				11.5	100	0.06

B. Helium ion Bombardment of Np^{237}.

Samples of Np^{237} were prepared and bombardments were carried out in a manner analogous to the methods of Section IV-A for deuteron bombardments of Pu^{239}.. The activated samples were chemically processed in the same way and fractions similar to those in the previous part were obtained. Analysis of the radiations from americium samples resulted in decay and absorption data which provided ample evidence for the presence of 12-hour and 50-hour activities with the same nuclear properties as before. The possible products of the helium ion bombardment of Np^{237} are just those expected in the deuteron bombardment of Pu^{239}, since the same compound nucleus is formed in each case.

In comparable bombardments, the relative yield of the 12-hour isotope compared to the 50-hour isotope was somewhat higher for the case of helium ions on Np^{237}. The kinetic energy of the helium ions was reduced from 38 Mev to 32 Mev in one case with a consequent change in relative yield. These results and the corresponding results from the Pu^{239}- deuteron experiments are discussed in Section IV-D.

C. Helium ion Bombardment of Pu^{239}.

In the helium ion bombardment of Pu^{239} (5) the particle and electromagnetic radiations of the 50-hour activity could be observed in the combined americium-curium fraction and in the americium fraction after separation of the americium and curium by means of their selective elution from a resin (Dowex 50) adsorption column with ammonium citrate solution. The 12-hour activity may well have been present initially but the time involved in this separation was so long that none remained after the separation. The formation of the 50-hour activity may have been due to any or all of the following mechanisms:

(1) Deuteron contamination of the ion beam, $Pu^{239}(d,3n)\ Am^{238}$

(2) $Pu^{239}\ (\alpha,p4n)Am^{238}$

(3) $Pu^{239}\ (\alpha,5n)\ Cm^{238} \xrightarrow[\text{short}]{K} Am^{238}$

D. Mass Assignments.

The isotopes of americium discussed in the previous parts of this section should, on the basis of their method of formation, have mass numbers in the range from 237 to 240. There is no genetic relationship between the two as might have been possible since the 12-hour activity could decay by isomeric transition. If the 50-hour activity were produced by decay of the 12-hour isotope, the logarithmic decay of the gamma activity, shown in Figure 12, would not be observed, but rather the curve would be convex due to the superposition of a 12-hour growth component for the 50-hour activity upon the decay of the initial independently formed 50-hour activity.

Neptunium and plutonium fractions were removed from portions of the americium fractions from the deuteron bombardment of Pu^{239}, after allowing time for the growth of daughter activities. The methods were sensitive enough to have detected Np^{236} (17 hour; beta particle emission) as the daughter of the 12-hour alpha activity if the latter were due to Am^{240}, but it has not been observed. The only other daughter isotope capable of being formed with sufficient yield to be detected as a result of growth from the americium activities, is Pu^{238} (ca. 90 year; alpha particle emission) if formed from Am^{238} by electron capture. The amounts of this isotope expected from the most radioactive samples of americium obtained were just on the limit of detection and positive evidence of its formation is lacking.

A quantitative evaluation of relative yields of different isotopes as a function of bombardment energy may often give an idea of their mass assignments especially if the mass number of one of the isotopes is known. In this case both are unknown. Since the half-lives of the isotopes are different, the calculation must be based upon an accurate knowledge of the cyclotron beam intensity at each time in the bombardment, if the latter is longer than, or comparable to, the half-lives of the isotopes. The beam intensity may, in some cases, vary widely from time to time, making summation methods necessary.

The number of atoms of a given isotope formed during a bombardment interval is given by

$$N = \frac{N_0 \sigma \varphi}{\lambda} (1 - e^{-\lambda t})$$

in which N is equal to the number of atoms formed; N_0, the number of target atoms per unit area; σ, the cross section for the reaction involved; λ, the decay constant of the product nucleus; φ, the intensity of incident particles; and t, the length of the bombardment interval. If observations are made at a time, t', after the end of the bombardment interval the equation must be modified:

$$N = \frac{N_0 \sigma \varphi}{\lambda} (1 - e^{-\lambda t}) e^{-\lambda t'}$$

Now, the number of disintegrations of the product nucleus, A, is related to the number of atoms, N, by the constant of proportionality, :

$$A = \lambda N$$

so that:

$$A = N_0 \sigma \varphi (1 - e^{-\lambda t}) e^{-\lambda t'}$$

If a given bombardment be considered as a series of intervals in each of which the intensity is relatively constant:

$$A = N_0 \sigma \sum_n \varphi_n (1 - e^{-\lambda t_n}) e^{-\lambda t'_n}$$

where t'_n is the time from the end of each bombardment interval to the end of the total bombardment (or to any other time of observation desired). It follows that the ratio of activities due to isotopes "a" and "b" is:

$$\frac{A_a}{A_b} = \frac{\sigma_a}{\sigma_b} \frac{\sum n(1 - e^{-\lambda_a t_n})e^{-\lambda_a t'_n}}{\sum n(1 - e^{-\lambda_b t_n})e^{-\lambda_b t'_n}} = \frac{\sigma_a}{\sigma_b} Y$$

Any observable quantity, A', which is proportional to the true rate of disintegrations through a constant, k, may be used in comparing the relative cross sections for the two reactions, the only modification being the introduction of a constant, K, into the observed ratio:

$$\frac{A_a}{A_b} = \frac{k_a A'_a}{k_b A'_b} = \frac{\sigma_a}{\sigma_b} Y \quad \text{and} \quad \frac{1}{Y} \frac{A'_a}{A'_b} = \frac{k_b}{k_a} \frac{\sigma_a}{\sigma_b} = K \frac{\sigma_a}{\sigma_b}.$$

By using the counting rates observed in the differential method described in Section IV-A, which are closely related to the x-radiation from each isotope, values of $\left(\frac{A' \ 12 \text{ hour}}{A' \ 50 \text{ hour}}\right)$ have been obtained for 19 Mev, 16 Mev and 13 Mev deuteron bombardments of Pu^{239}, as well as for 38 Mev and 32 Mev helium ion bombardments of Np^{237} (Table 2). The corresponding values of Y then enable the calculation of $K\left(\frac{12 \text{ hour}}{50 \text{ hour}}\right)$ for each particle and energy to be made. <Table 2

From these data certain facts become apparent: (1) the yield of the 12-hour isotope increases relative to the 50-hour isotope with decrease in kinetic energy of the incident particle, whether helium ions or deuterons are considered, (2) the variation in relative yields is not as great as is normally expected for the reaction (α,n) or (α,4n), compared to (α,3n) or (α, 2n)(or, similarly, for the (d,n) or (d,4n) reactions, compared to the (d,3n) or (d,2n)), and (3) the actual yield of 12-hour activity appears to be greater than that of 50-hour activity, since K is probably not very far from unity and for 38 Mev helium ions the value of $K\left(\frac{\sigma 12 \text{ hour}}{\sigma 50 \text{ hour}}\right)$ is quite large. At 38 Mev the (α,2n) and (α,3n) reactions are known to be predominant (similarly, the (d,2n) and (d,3n) reactions at 19 Mev), and at 13 Mev the (d,4n) reaction is expected to have a very low yield. Mass assignments which are entirely consistent with the observed data are:

Table 2

Relative Yields of Americium Activities in Charged Particle Bombardments

Projectile	Kinetic Energy	Y	$\left(\frac{A' \text{ 12 hour}}{A' \text{ 50 hour}}\right)$	$K\left(\frac{\sigma \text{ 12 hour}}{\sigma \text{ 50 hour}}\right)$
He^{++}	38 Mev	2.94	11.72	4.0
	32 Mev	3.74	21.80	5.8
d	19 Mev	1.19	0.95	0.8
	16 Mev	2.41	2.67	1.1
	13 Mev	2.83	3.48	1.2

(a) Am^{238}, for the 50-hour isotope and (b) Am^{239}, for the 12-hour isotope.

Another fact emerging from the data of Table 2 is that the compound nucleus in the helium ion bombardment of Np^{237} is different in some respect from that formed in the deuteron bombardment of Pu^{239}. If the expected excitation energy is corrected for the difference in binding energies per particle in the helium nucleus and the deuteron, the compound nucleus formed in the 32 Mev helium ion bombardment should correspond rather closely to that in the 19 Mev deuteron bombardment. The relative yields in the two cases are more different than any of those observed with a wide variation in kinetic energy of either particle. This may be due in part to large differences in binding energy of the nucleons or differences in angular momentum of Np^{237}, relative to Pu^{239}, and/or the failure of the compound nuclei to achieve uniform distribution of the excitation energy.

We wish to thank Professor J. G. Hamilton, Mr. T. M. Putnam and their associates in the Crocker Radiation Laboratory for their cooperation in providing bombardments with the 60-inch cyclotron. Mr. Albert Ghiorso and Mr. S. G. Thompson participated in many of the experiments. The cooperation of the groups at the Clinton Laboratories and the Hanford Engineer Works in making the neutron irradiations is also gratefully acknowledged.

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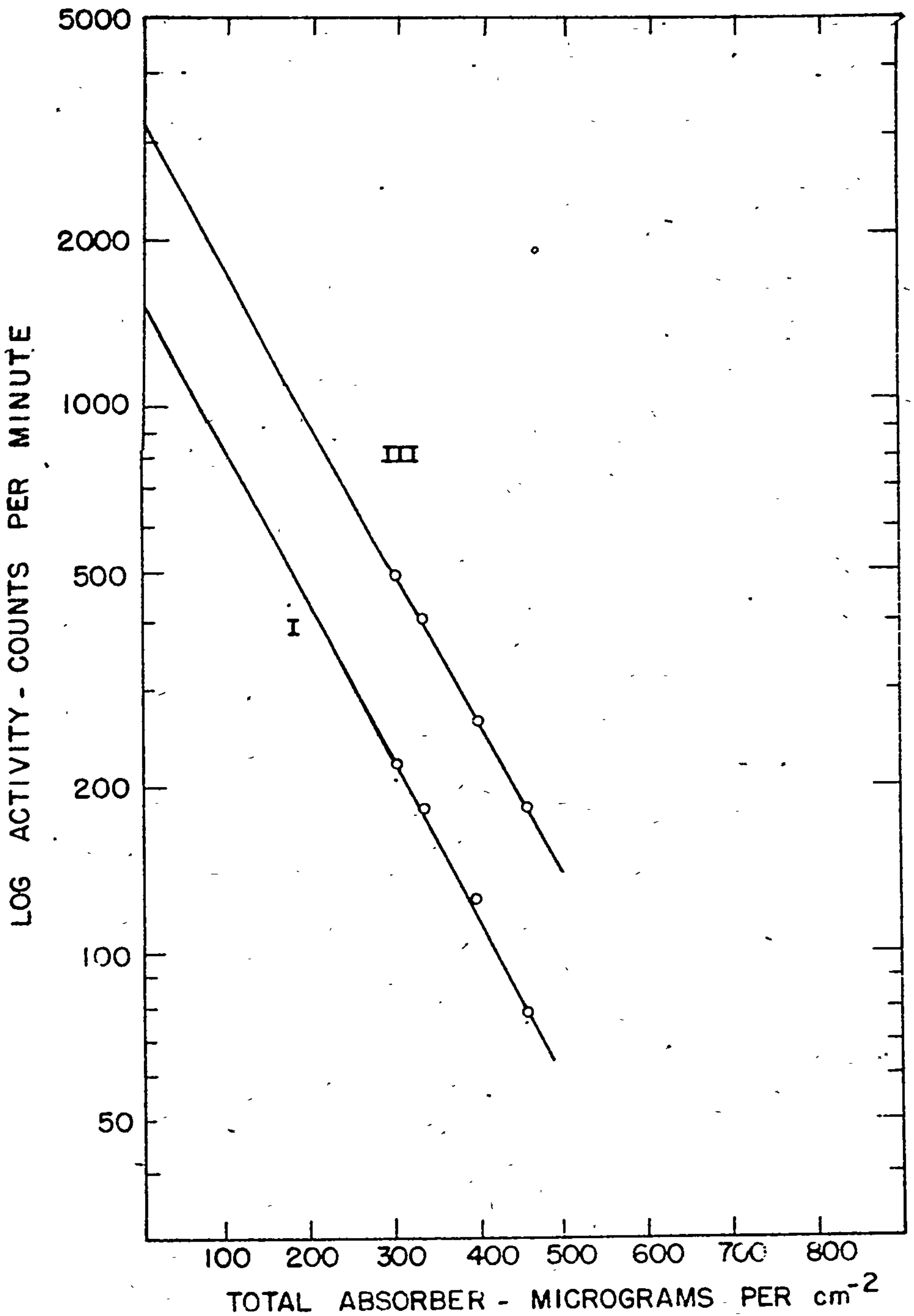


Figure 1
Cellulose nitrate absorption curves for beta particle activity due to Pu^{241} in the first (I) and third (III) 100 mg.cm.$^{-2}$ layers of a helium ion bombarded uranium target.

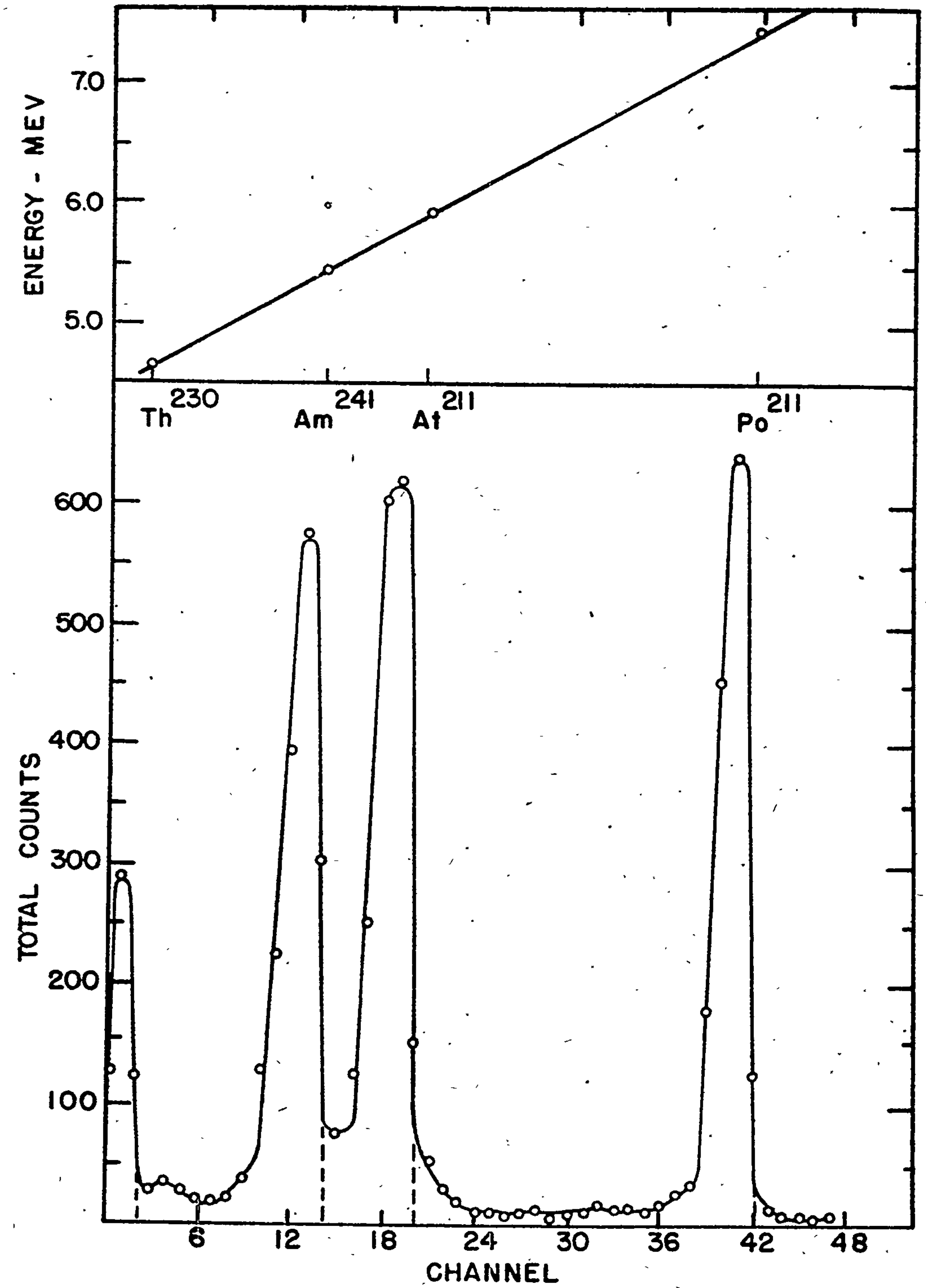


Figure 2.
Alpha particle pulse analysis curve for Am^{241} and alpha particle energy determination with Th^{230}(Io), At^{211} and Po^{211} alpha particle standards.

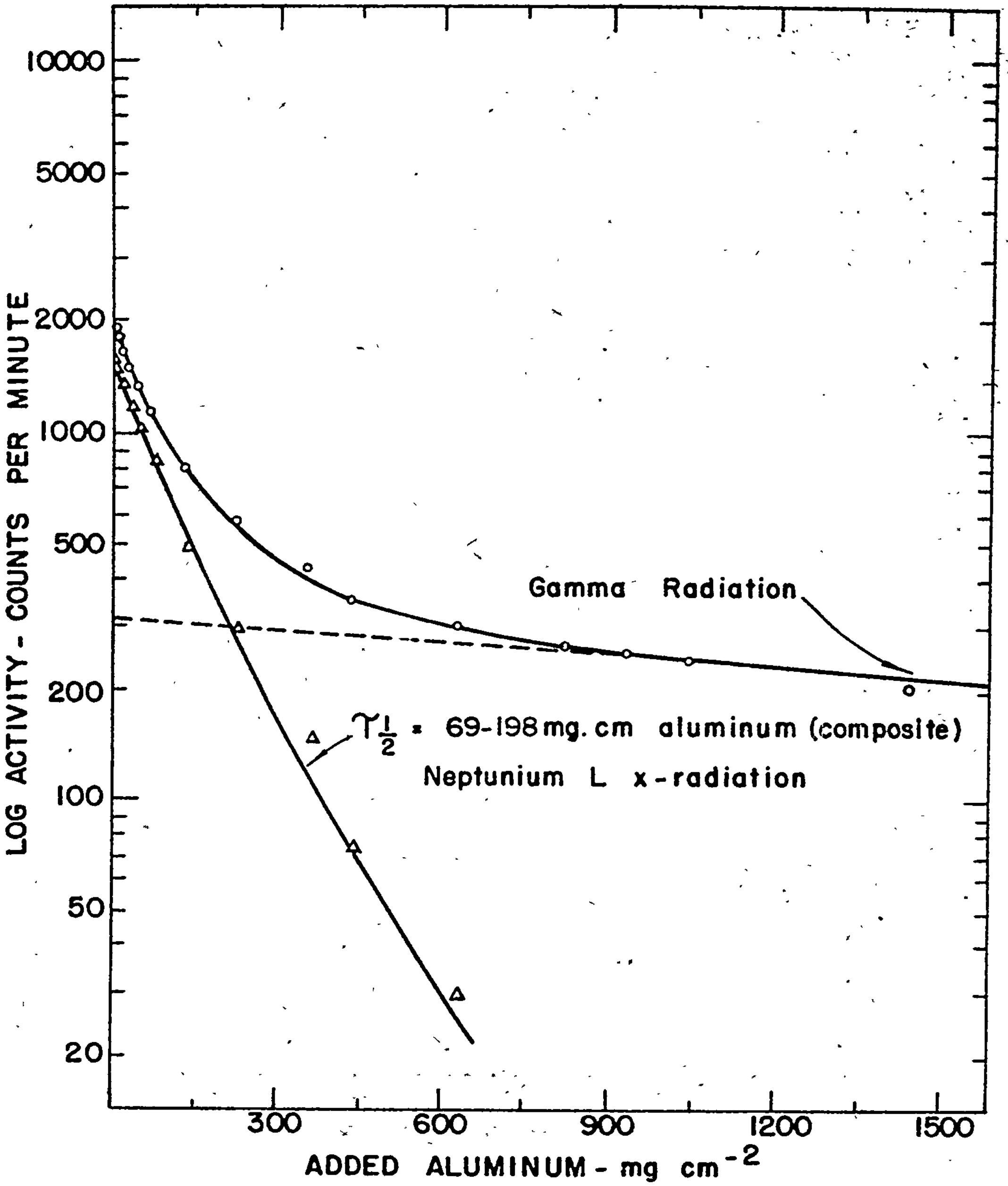


Figure 3
Aluminum absorption curve for Am^{241} electromagnetic radiation.

A - Observed absorption curve.
B - Gamma radiation.
C - L x-radiation (neptunium).

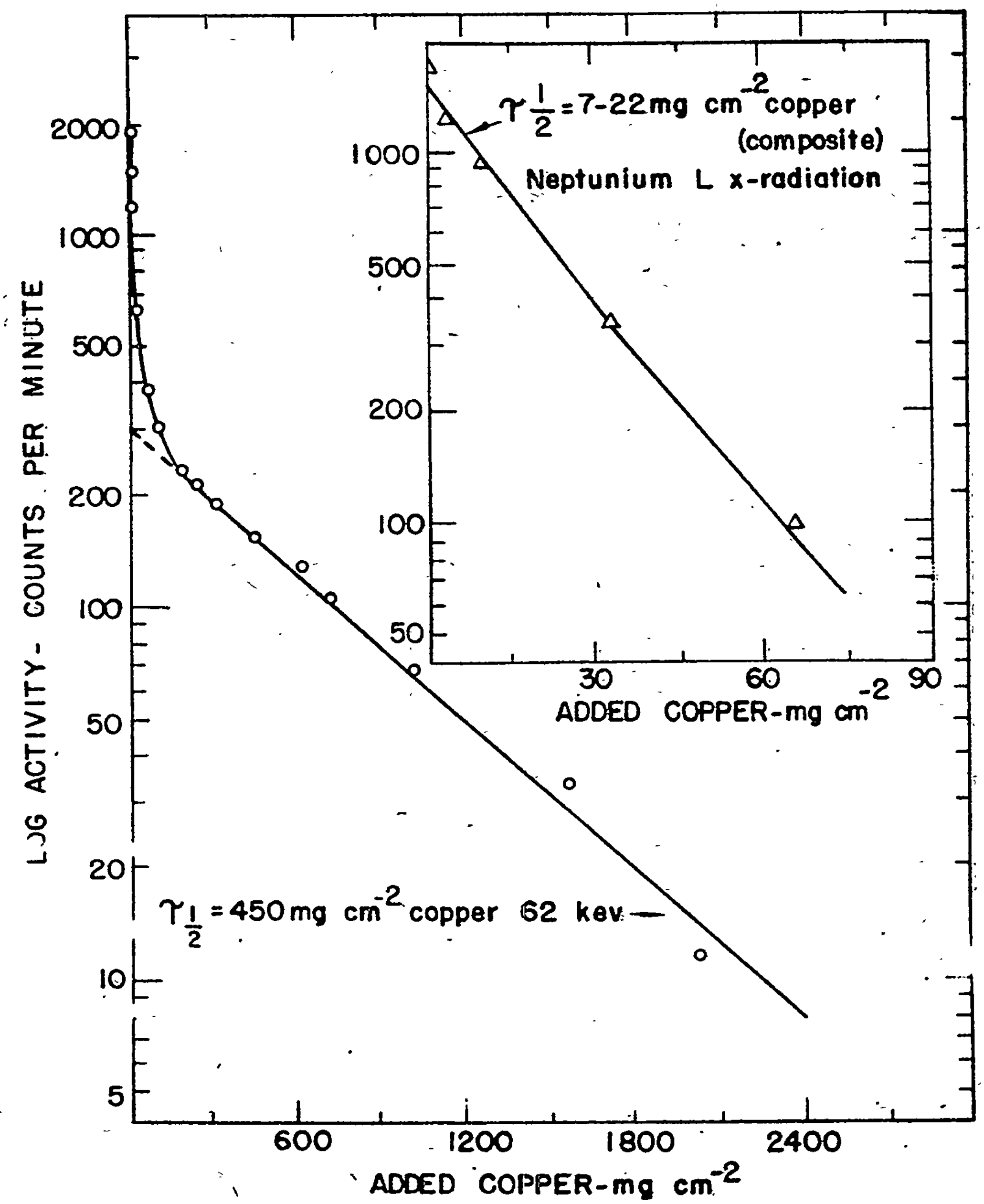


Figure 4
Copper absorption curve for Am^{241} electromagnetic radiations.

A - Observed absorption curve.
B - Gamma radiation; (62 kev).
C - L x-radiation (neptunium).

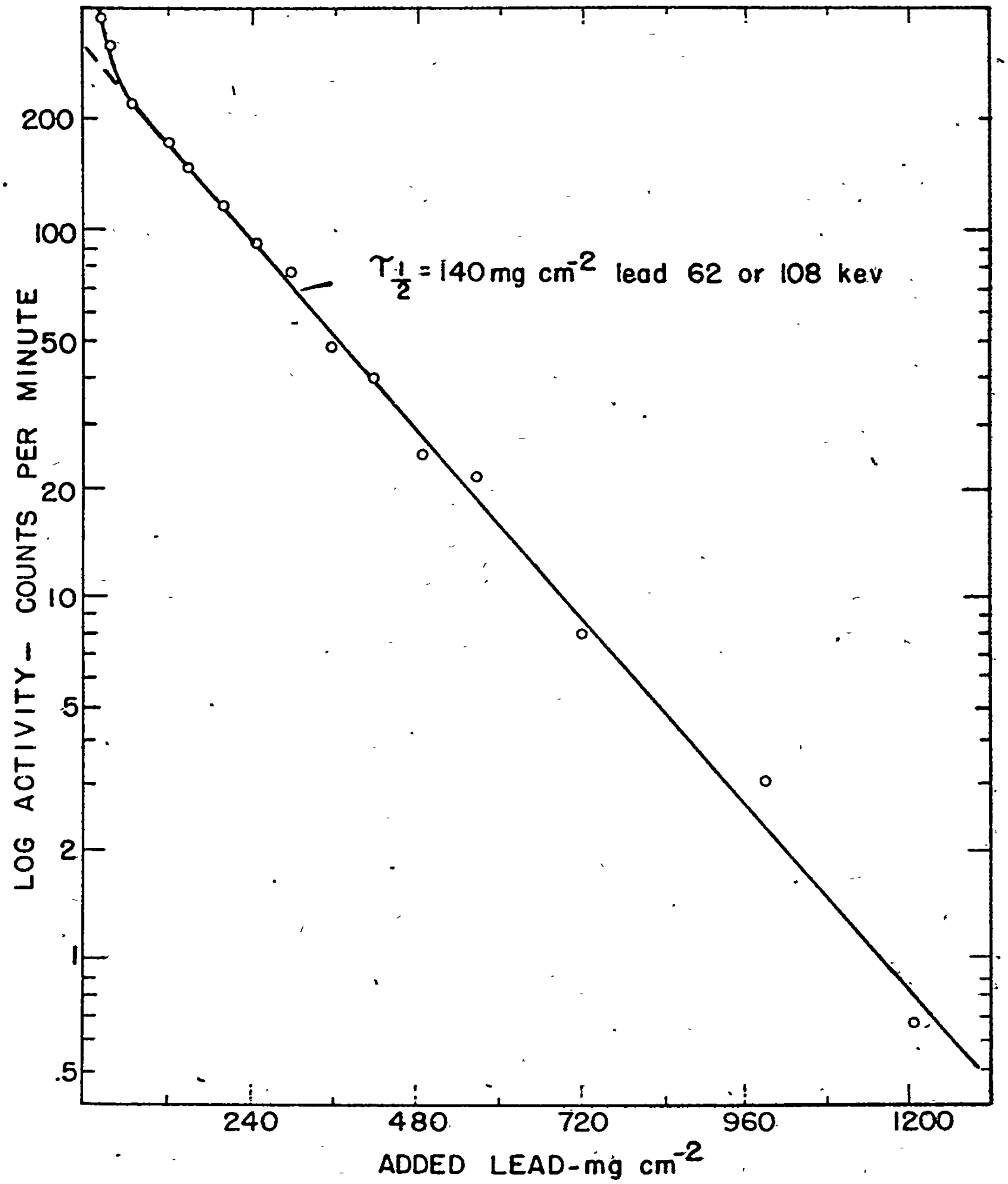
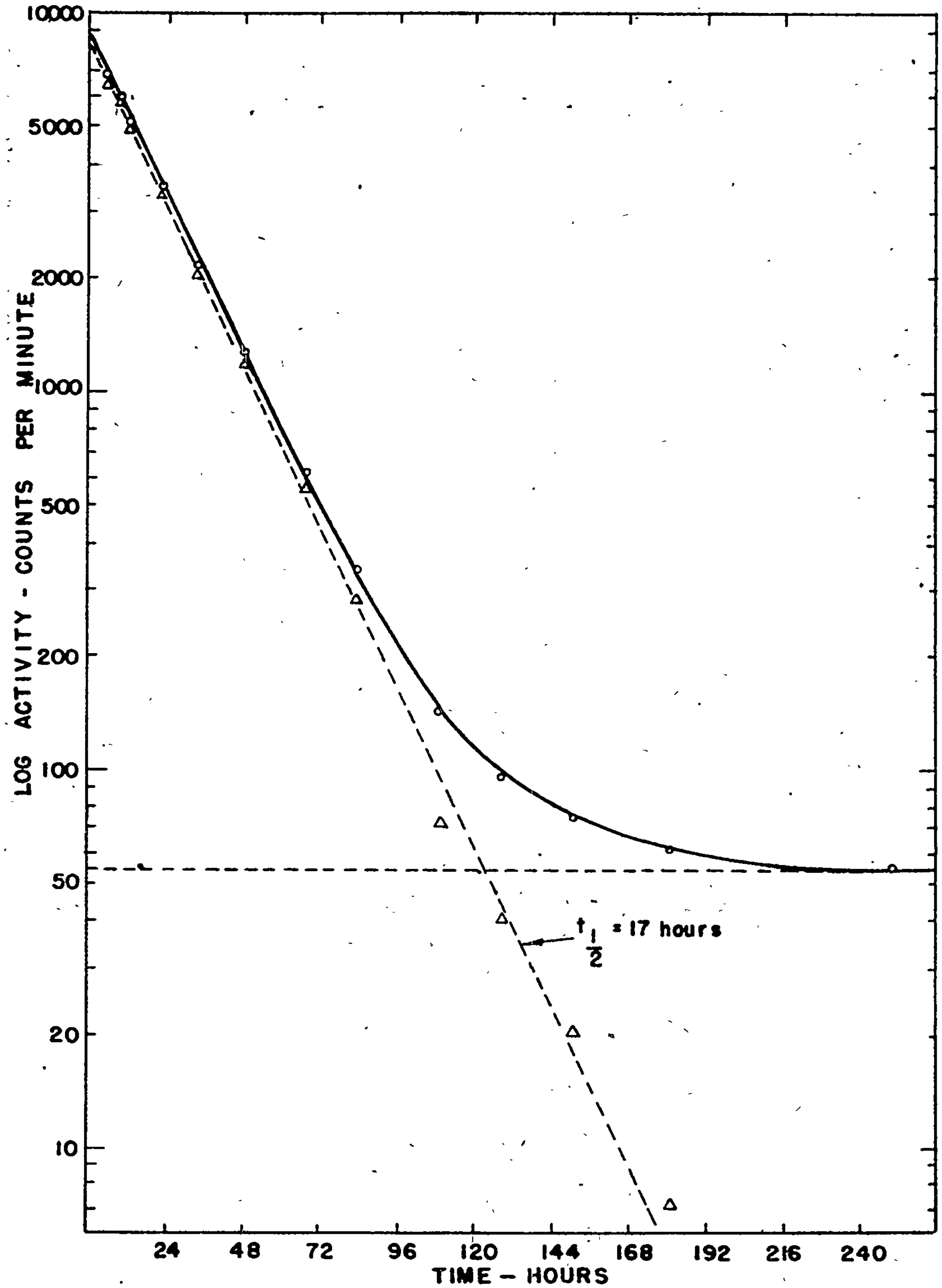


Figure 5
Lead absorption curve for Am^{241} showing the 62 kev gamma radiation.

Figure 6
Decay curve obtained for the americium fraction from neutron irradiated Am
A - Decay of activity measured through a 7 mg.cm.$^{-2}$ aluminum filter.
B - Long-lived activity.
C - Decay of beta particles of Am^{242} (17 hour).



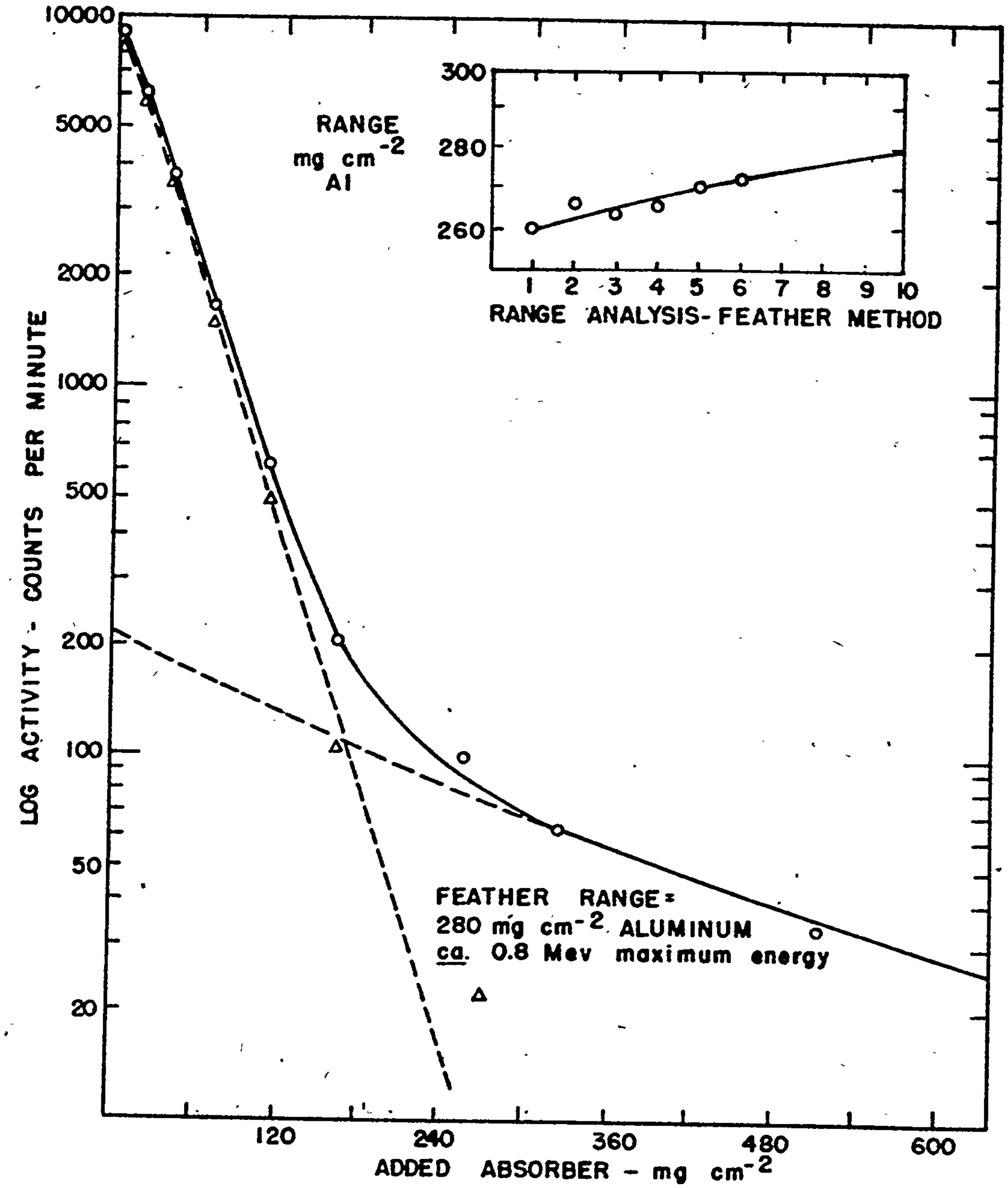


Figure 7
Aluminum absorption curve for the americium fraction from neutron irradiated Am^{241}.

A - Observed absorption curve.
B - Components due to long-lived activity.
C - Beta particles of Am^{242}

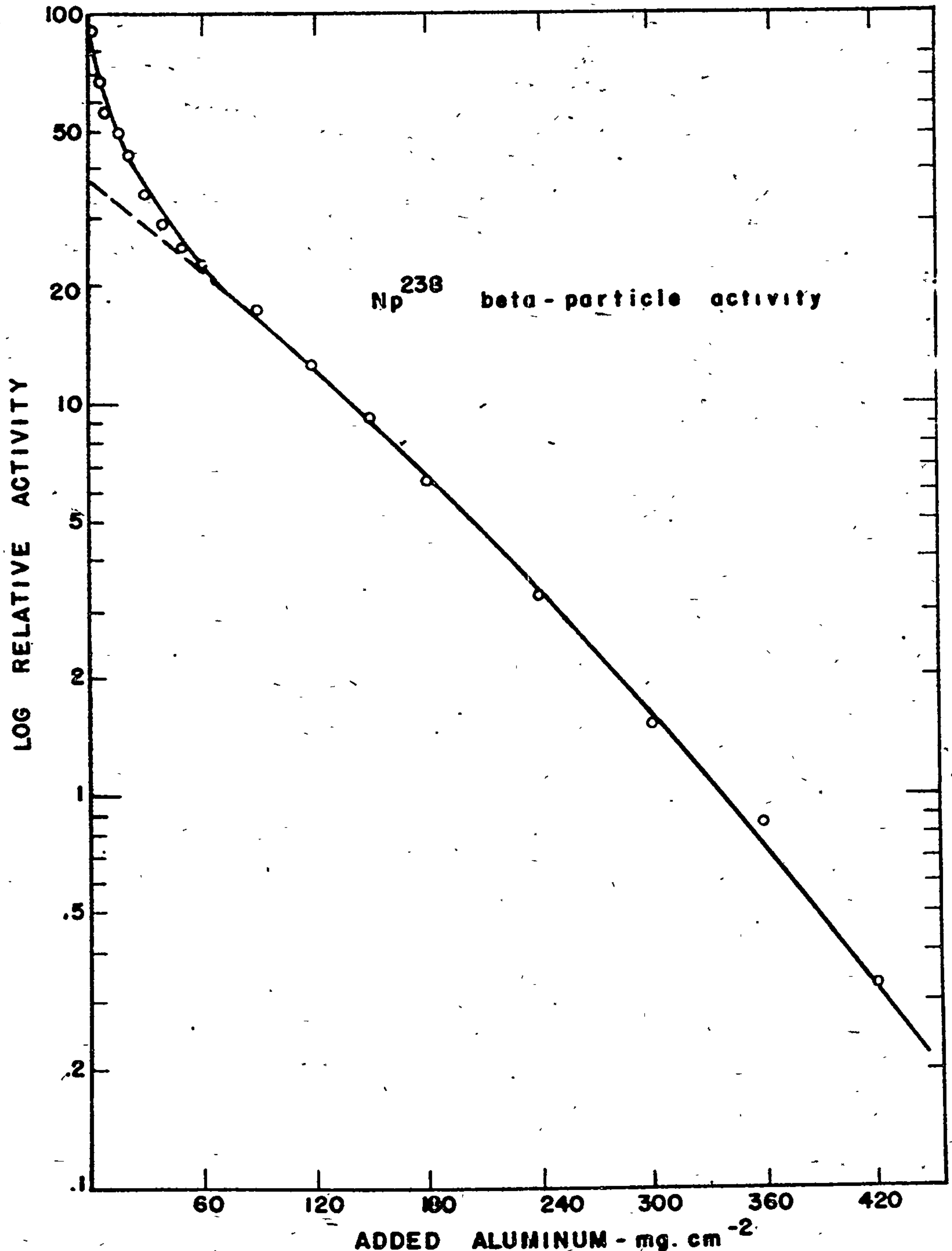


Figure 8
Aluminum absorption curve obtained for beta activity in the neptunium fraction removed from neutron irradiated Am^{241} after six months. The solid line is the absorption curve for known Np^{238}

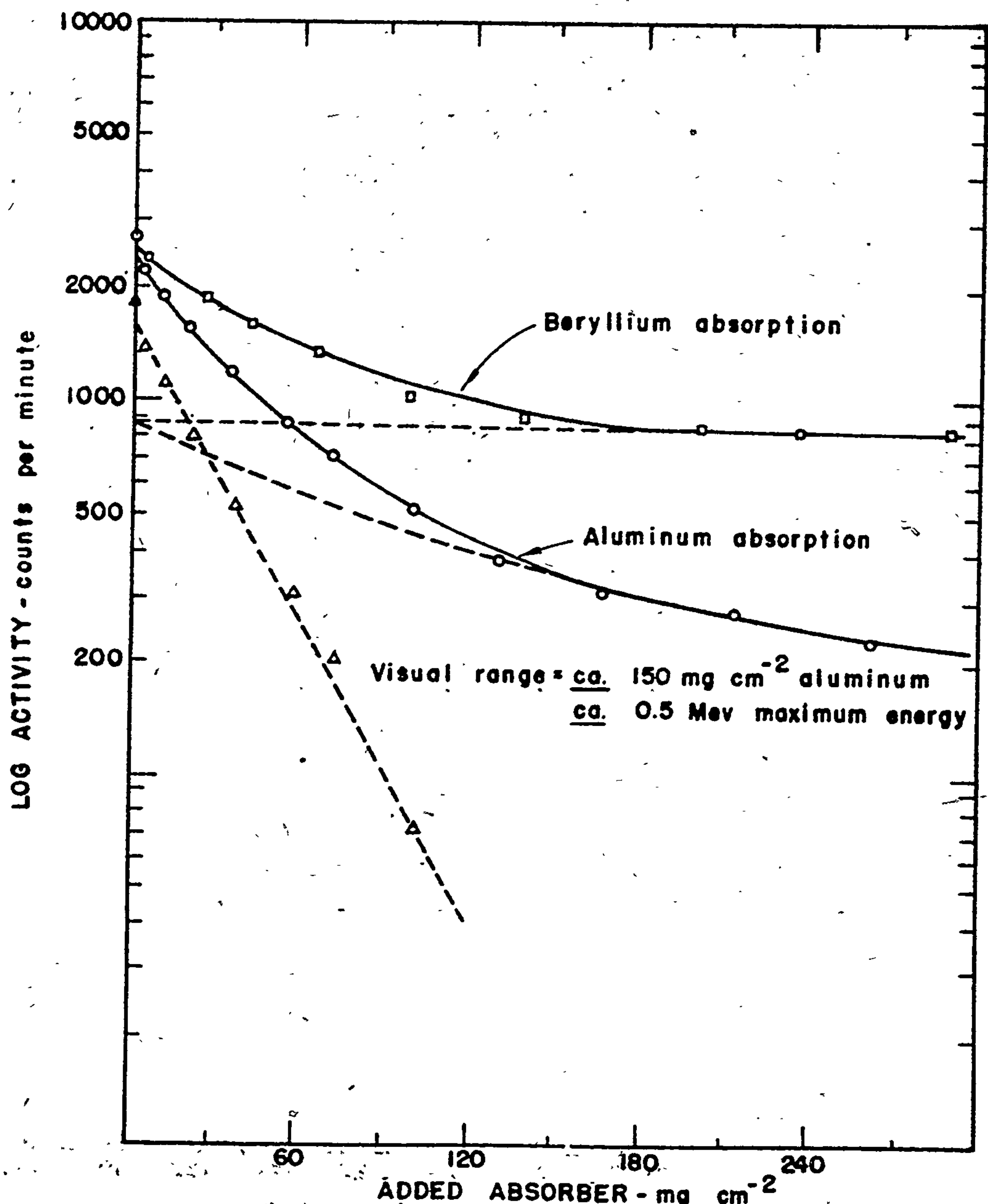


Figure 9
Absorption curves for the long-lived beta activity of Am^{242}. Counting geometry - 10 percent.

A - Aluminum absorption curve for beta and electromagnetic radiations in the americium fraction of neutron irradiated Am^{241} (after several months).

B - Electromagnetic components, due largely to Am^{241}.

C - Beta activity of Am^{242} (long-lived).

D - Beryllium absorption curve for beta and electromagnetic radiations in the americium fraction.

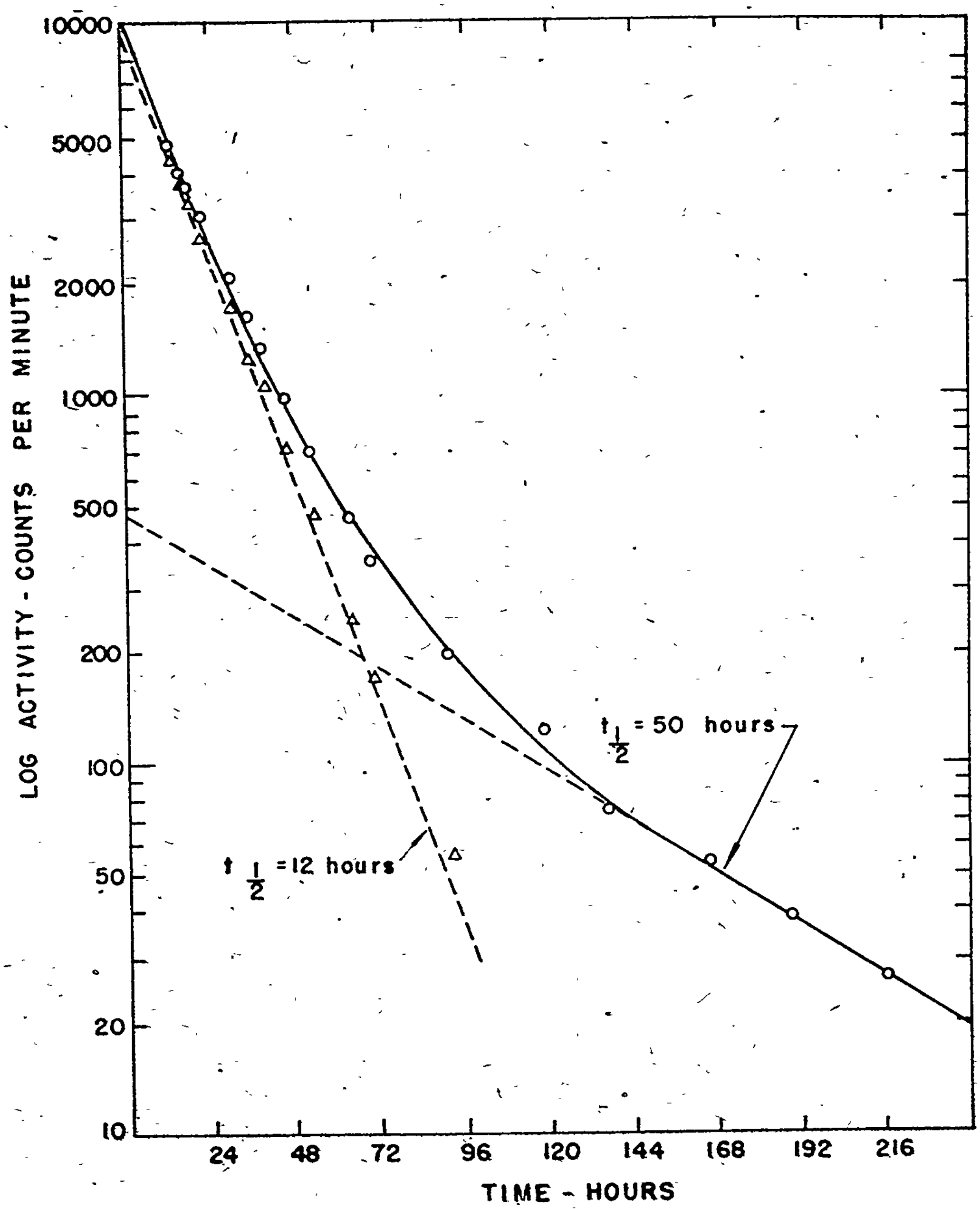


Figure 10

Decay of beta and electromagnetic radiations in the americium fraction from deuteron bombarded Pu^{239}, measured through a 7 mg.cm.$^{-2}$ aluminum filter. Counting geometry - 10 percent.

A - Observed decay.
B - 50 hour component.
C - 12 hour component.

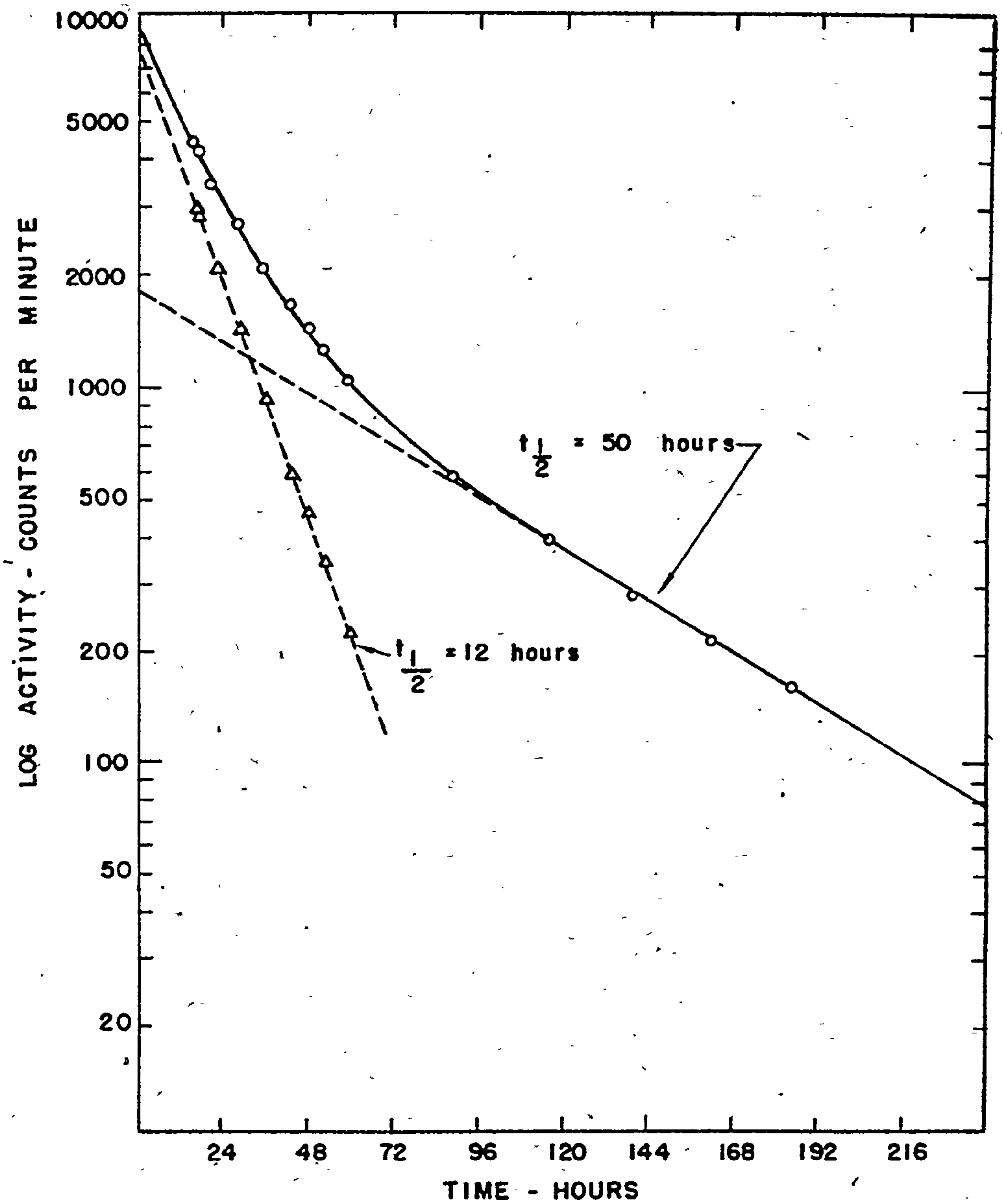


Figure 11
Decay of low energy electromagnetic radiations in the americium fraction from deuteron bombarded Pu^{239}. Data was obtained as the difference between the amount of activity passing through a 1500 mg.cm.$^{-2}$ beryllium filter and that passing through a combination of 1500 mg.cm.$^{-2}$ beryllium and 160 mg.cm.$^{-2}$ lead filters. Counting geometry - 10 percent.

A - Observed decay.
B - 50 hour component.
C - 12 hour component.

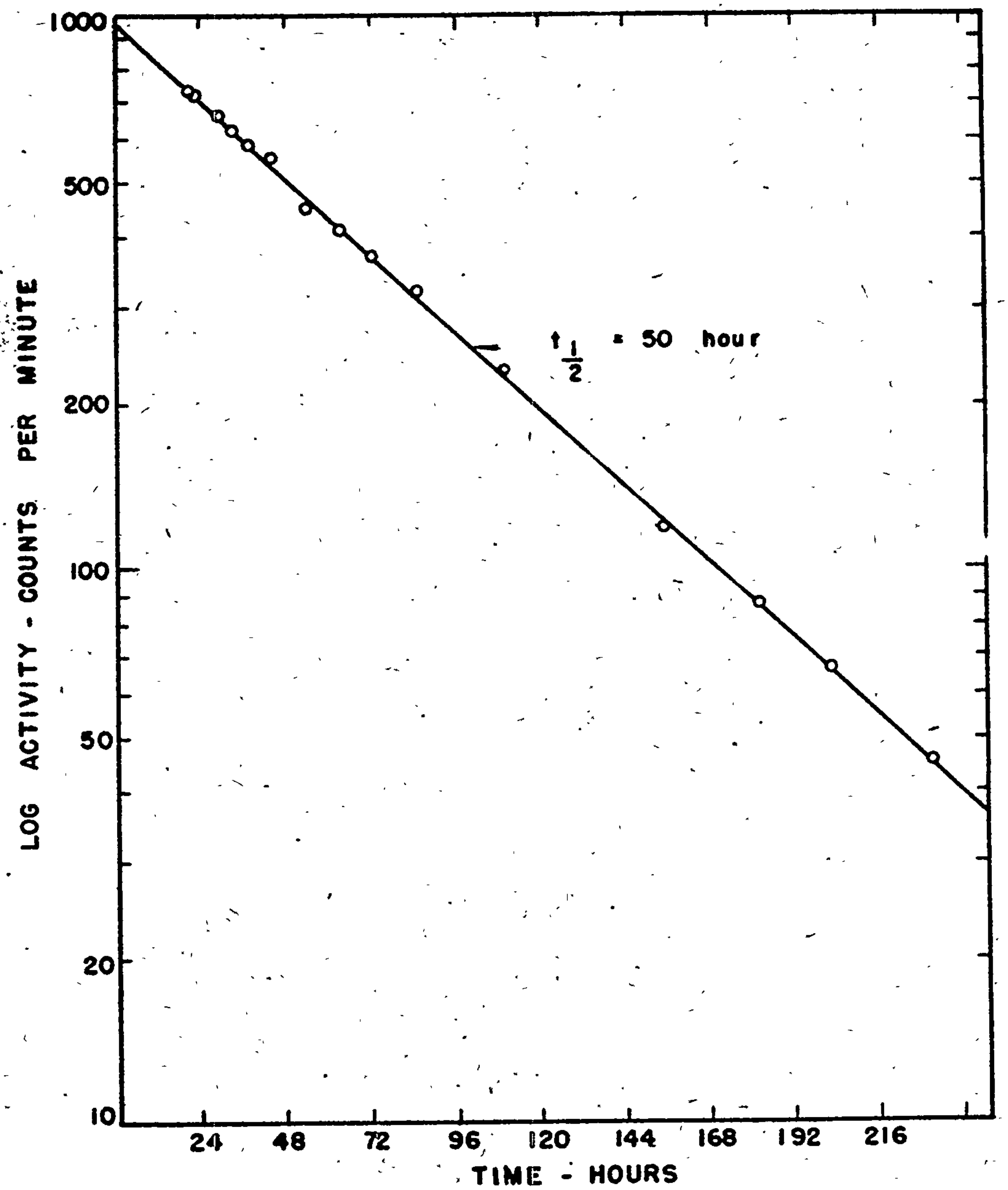


Figure 12
Decay of hard gamma radiation (5 g.cm.$^{-2}$ lead filter) in the americium fraction from deuteron bombarded Pu^{239}. Counting geometry - 10 percent.

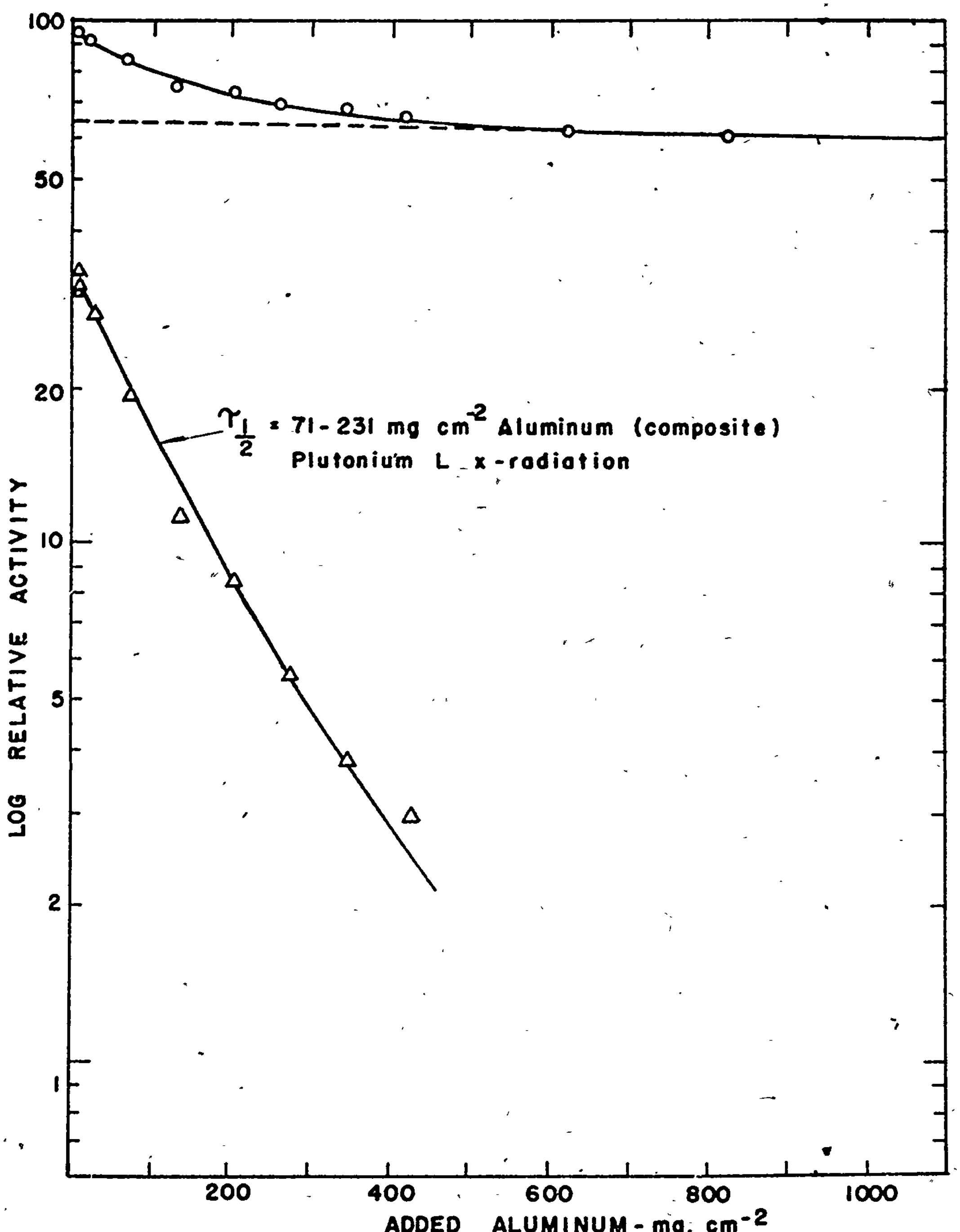


Figure 13
Aluminum absorption curve for electromagnetic radiations due to the 50 hour activity in the americium fraction of deuteron bombarded Pu^{239} (1500 mg.cm.$^{-2}$ beryllium filter to remove electrons). Counting geometry - 10 percent.

A - Observed absorption curve.
B - Gamma and K x-radiations.
C - L x-radiation (curve drawn for plutonium L x-rays).

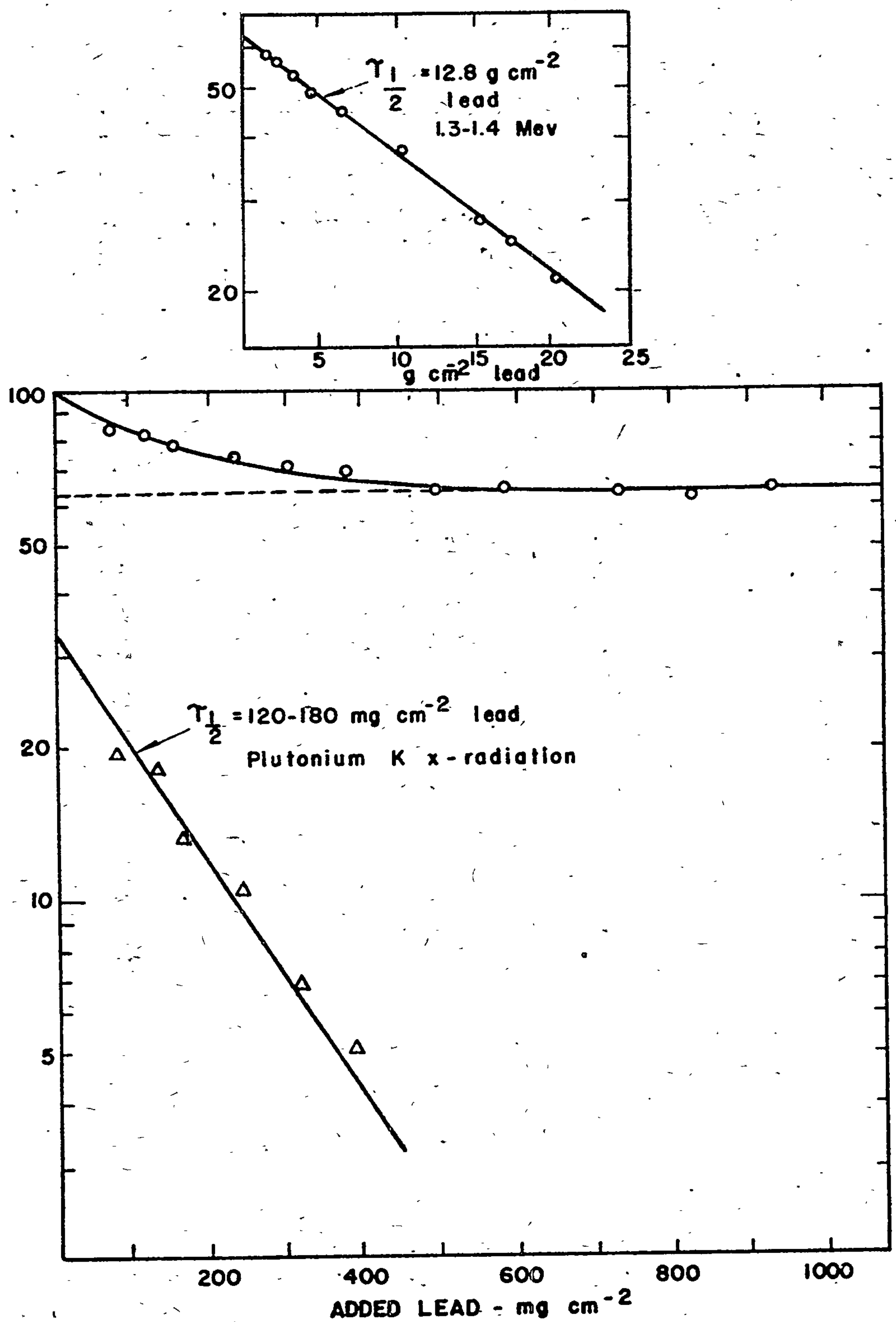


Figure 14
Lead absorption curve for electromagnetic radiations due to 50 hour activity in the americium fraction from deuteron bombarded Pu^{239} (1500 mg.cm.$^{-2}$ beryllium filter). Counting geometry - 10 percent.

A - Observed absorption curve.
B - Gamma radiation.
C - K x-radiation (curve drawn for plutonium K x-rays).

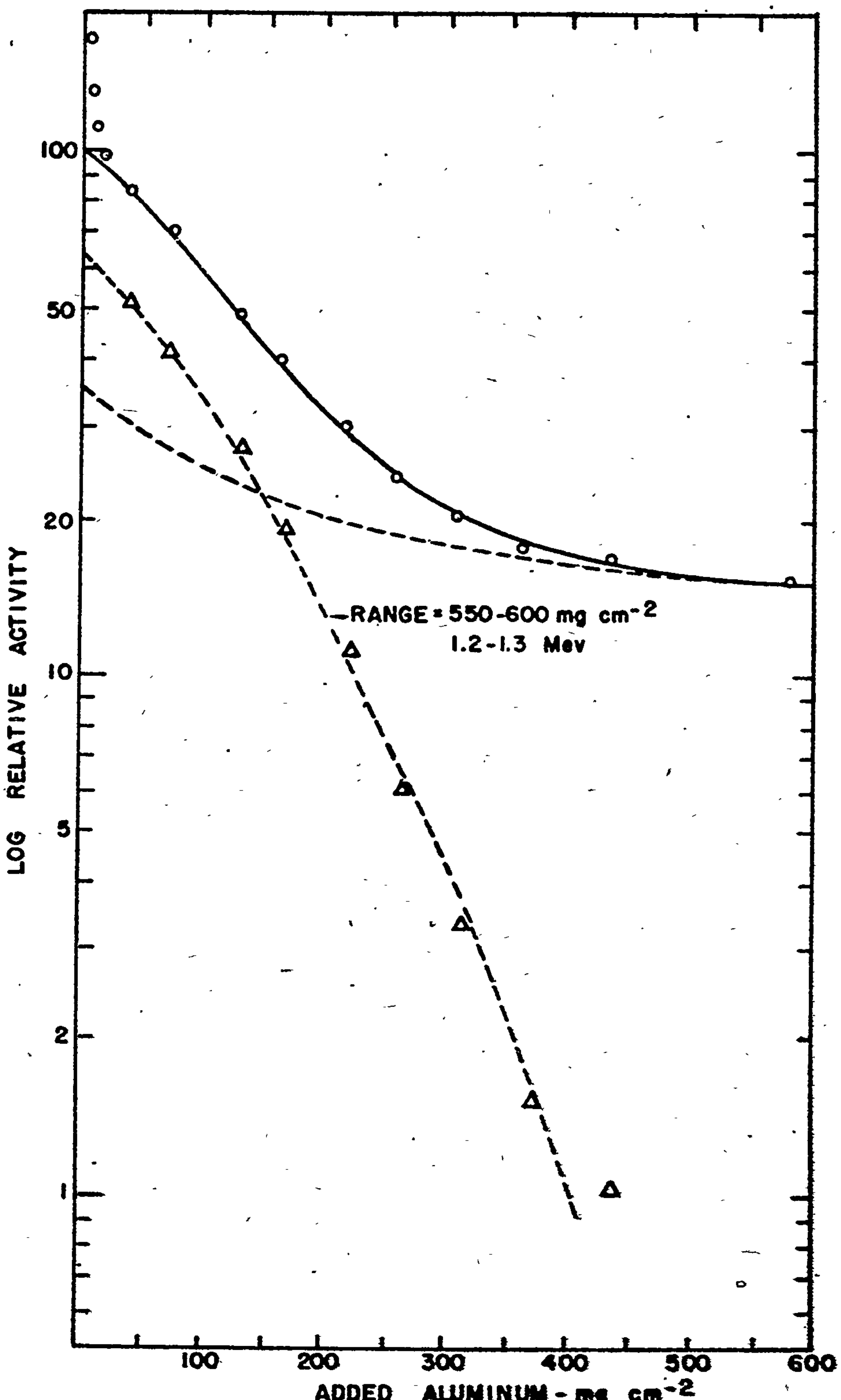


Figure 15
Aluminum absorption curve for electron activity associated with the 50 hour activity in the americium fraction of deuteron bombarded Pu^{239}. Electromagnetic radiations have been subtracted. Counting geometry - 10 percent.

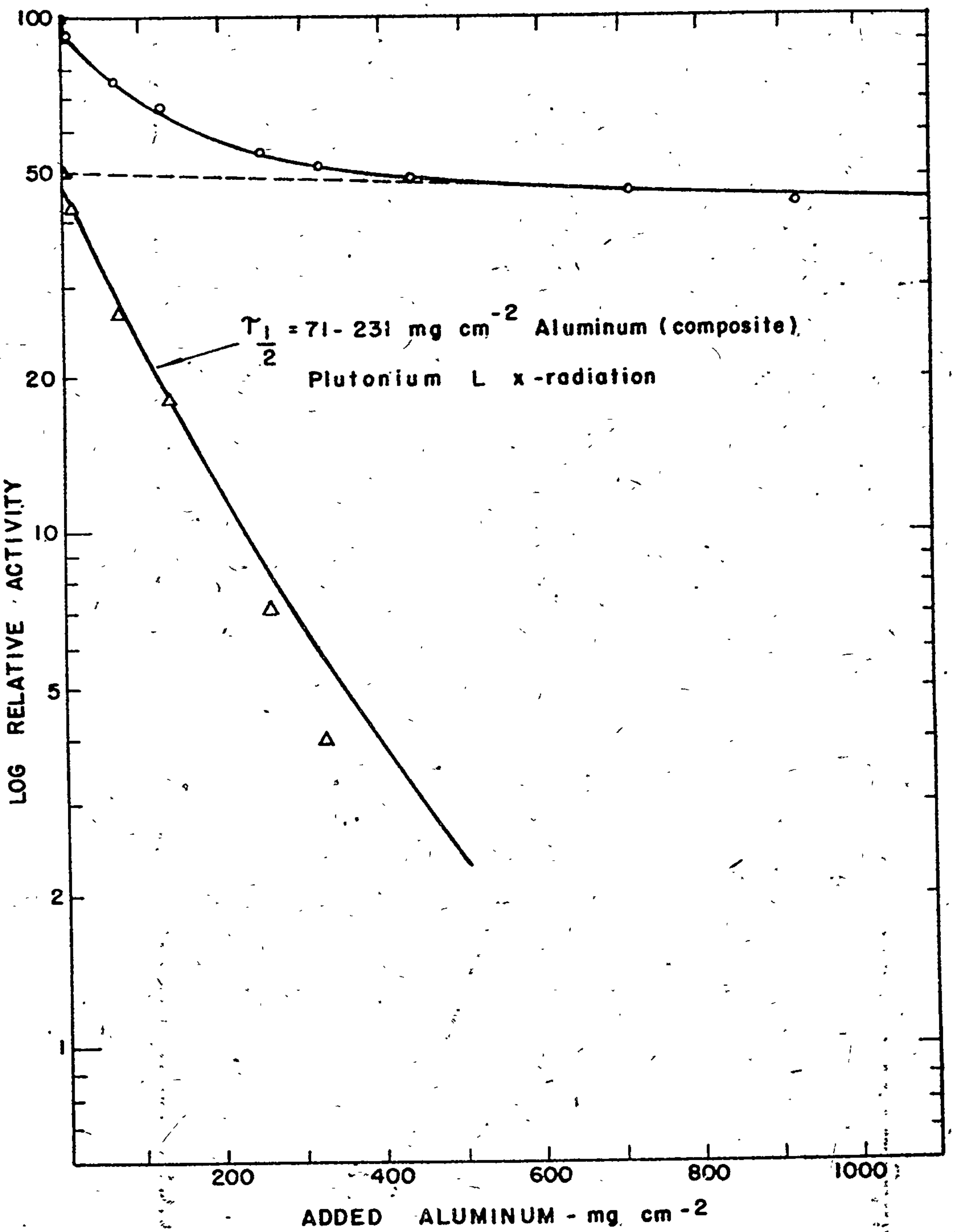


Figure 16
Aluminum absorption curve for electromagnetic radiations due to 12 hour activity in the americium fraction from deuteron bombarded Pu^{239}. (1500 mg.cm.$^{-2}$ beryllium filter). Counting geometry - 10 percent.
A - Absorption curve obtained by subtraction of 50 hour components.
B - Gamma and K x-radiations.
C - L x-radiation (curve drawn for plutonium L x-rays).

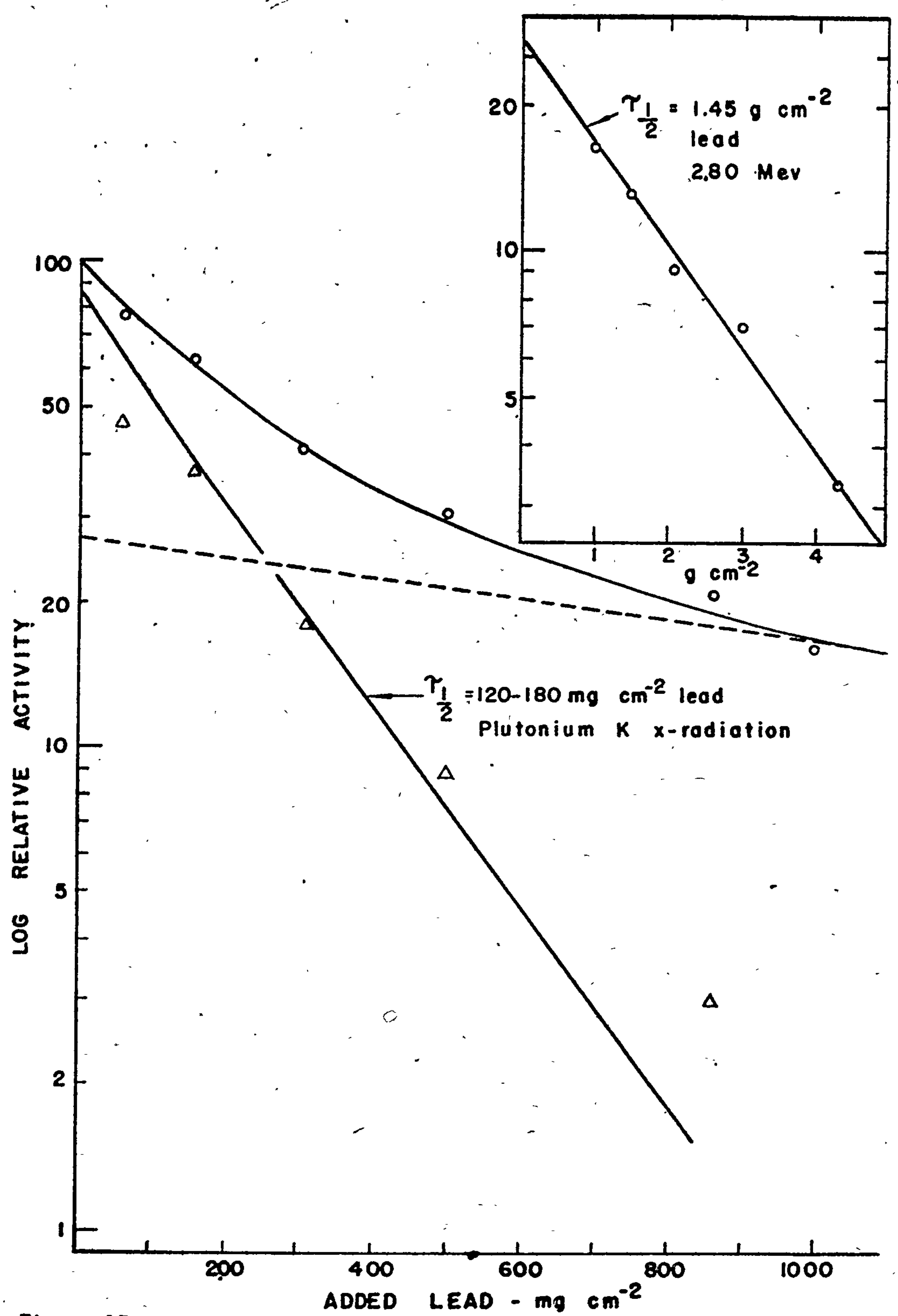


Figure 17
Lead absorption curve for electromagnetic radiations due to 12 hour activity in the americium fraction from deuteron bombarded Pu^{239} (1500 mg.cm.$^{-2}$ berylliu filter). Counting geometry-10 percent.

A.- Absorption curve obtained by subtraction of 50 hour components.
B.- Gamma radiation.
C.- K x-radiation (curve drawn for plutonium K x-rays).

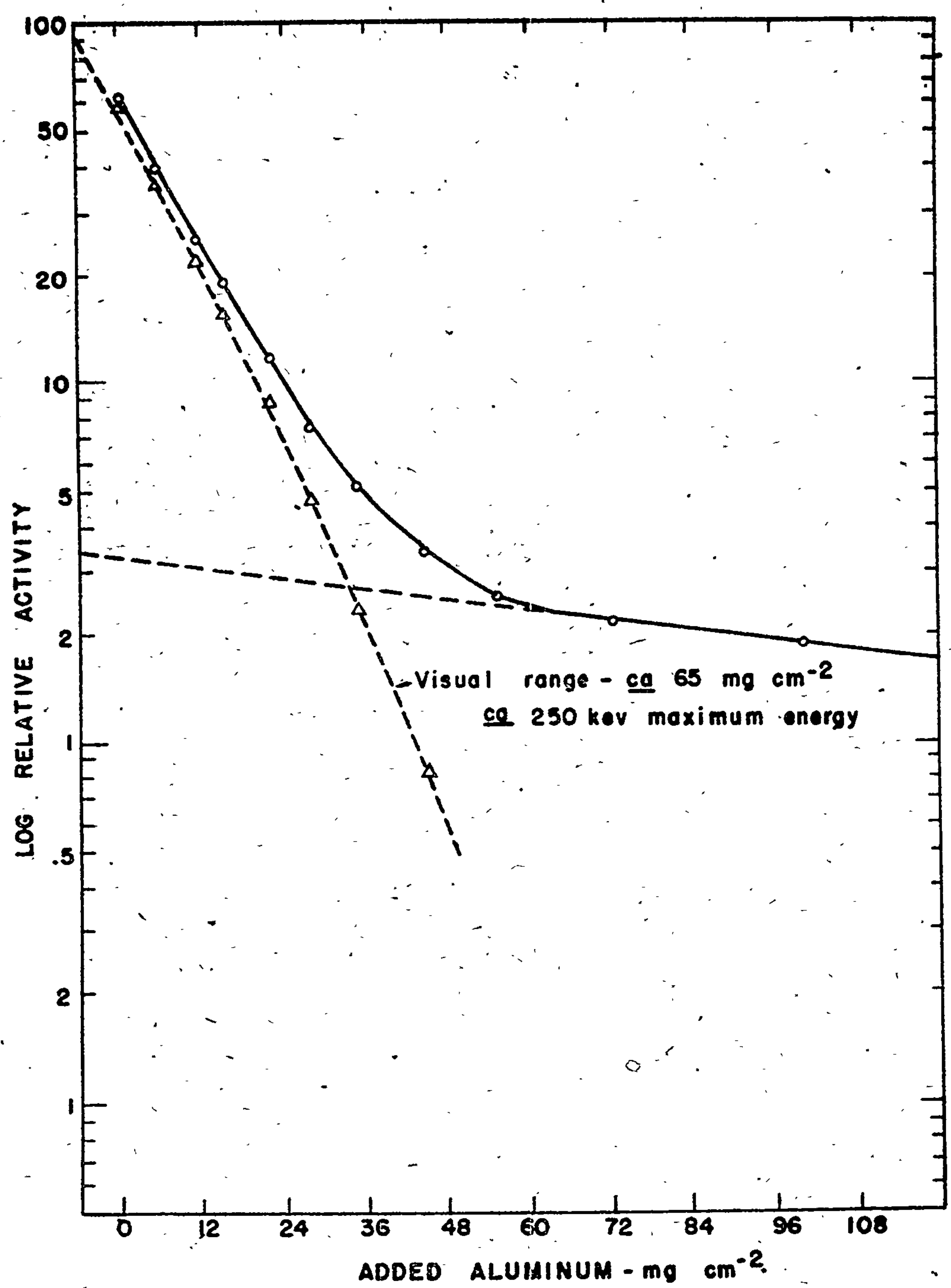


Figure 18

Aluminum absorption curve for electron activity associated with 12 hour activity in the americium fraction of deuteron bombarded Pu^{239}. Fifty hour electron activity and all electromagnetic radiations have been subtracted. Counting geometry - 10 percent.