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ABSTRACT

The decay characteristics of the two isomers of Am^{242} have been investigated. The beta decay of the 16-hour Am^{242m} has a branching ratio of 51 ± 5 percent to the first excited state (42.3 kev) of Cm^{242} with the remainder of the decay going to the ground state. The electron capture decay of Am^{242m} has a branching ratio of approximately 60 percent to first excited state (44.8 kev) of Pu^{242}. An upper limit of 6 percent has been set for the fraction of Am^{242m} decay via isomeric transition. The beta decay of the 100-year Am^{242} has a branching ratio of 45 ± 5 percent to the first excited state of Cm^{242}, with the remainder of the decay going to the ground state. The beta spectrum end points for Am^{242m} and Am^{242} have been measured to be 620 ± 10 kev and 585 ± 10 kev respectively. A decay scheme for the two isomers has been proposed. Log ft values have been calculated for beta and electron capture decay of the isomers and are discussed in conjunction with spin and parity assignments.

RADIOACTIVE DECAY OF THE ISOMERS OF AMERICIUM-242*

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I. INTRODUCTION

The two isomers, 16-hour Am^{242m} and long-lived (approximately 100 years) Am^{242}, were first observed as neutron-capture products of Am^{241}.[1] The decay characteristics of these isomers, particularly Am^{242m}, have been studied by O'Kelley et al.[2] From the experimental data obtained by the authors, it is possible to formulate decay schemes for these isomers which differ in certain respects from those reported in previous work. The fraction of Am^{242m} decay which proceeds via the isomeric transition was found to be much smaller than was thought previously. Also, the energies of the levels of Pu^{242} and Cm^{242} populated by the decay of Am^{242m} have been determined from a reinterpretation of the conversion electron spectrum. The results of Church[3] on the decay of Am^{242m} essentially agree with the data reported here.

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II. EXPERIMENTAL METHODS AND RESULTS

The measurements described were performed on quantities of Am^{242m} and Am^{242} produced in three separate neutron irradiations of purified Am^{241}. The decay characteristics of Am^{242m} were studied using americium from two irradiations of one day and five days in duration.[4] A separate sample of americium which had been subjected to a much greater total neutron irradiation was used in the study of the long-lived Am^{242}. Mass analysis of this sample revealed the following isotopic abundances: Am^{241} -- 90.4 percent, Am^{242} -- 1.1 percent, and Am^{243} -- 8.5 percent. The americium was purified after irradiation using a combination of hydroxide and fluoride precipitations, ion exchange columns, and oxidation to the hexapositive oxidation state, techniques which have been described previously.[5] Since the Am^{242} isomers were produced by neutron irradiation, Am^{241} constituted a large fraction of the mass of each sample. The radiations of Am^{242m} were identified by their 16-hour decay, while the radiations of long-lived Am^{242} were resolved in a mixture of Am^{241} and Am^{242} by eliminating those which also appeared in a pure sample of Am^{241}.

The electromagnetic radiation of Am^{242m} in the region of 13-60 kev was examined with a bent crystal spectrometer of the Cauchois type.[6] A portion of this spectrum showing the L α lines is shown in Fig. 1. The L x-rays of neptunium observed in the spectrum arise from the conversion of gamma rays from the alpha decay of Am^{241}. The data on the L x-rays associated with the decay of Am^{242m} are listed in Table I. In addition to the bent crystal spectrometer study, a sodium-iodide-crystal spectrometer was employed to measure the intensity of the Pu K x-ray. Using fluorescence yields based on Kinsey's estimates,[7] the relative L shell vacancies have been calculated for curium and plutonium: Cm $L_2:L_3$ = 250:110 and Pu $K:L_2:L_3$ = 40:70:30. These figures were based upon the observed intensities and were corrected for the abundance of L lines not observed using data on Am^{241}.[8] The L_1 quantum yield has been omitted because of uncertainties in line assignments and low abundance. The fluorescence yield for the

plutonium K shell is estimated as 98 percent.[9] A correction has also been applied for the L vacancies in plutonium due to emission of K x-rays. No corrections have been made to the above intensity ratios for nonradiative transitions of the Coster-Krönig type because of the uncertainty in the magnitude of these effects. These L x-ray intensities lead to a ratio of curium to plutonium L conversion of 3.6 assuming only K and L_1 electron capture.

There is considerable question as to the origin of the americium L x-rays. It was observed that these lines decayed to some extent and then remained constant with time. The americium L x-rays which did not decay with a characteristic 16-hour half-life are thought to be due to self-excitation of the bulk Am^{241} in the sample, and a correction for this effect has been applied to the intensities. In addition, the presence of x-rays with the Am^{242m} decay capable of exciting the americium L_2 and L_3 shells indicates the observed relative abundances are merely upper limits. From the intensity of the americium L x-rays and on the basis of L conversion alone, an upper limit of 6 percent may be set for a predominantly L-converted isomeric transition. Although no evidence has been found for the presence of radiations or conversion electrons of an isomeric transition of energy greater than about 30 kev, there is a possibility that the isomeric transition is either not energetic enough to convert in the L shell of americium or produces extremely low-energy L conversion electrons. In such a case the upper limit of 6 percent for the isomeric transition does not hold.

The conversion electron and beta spectra of these isomers were studied using a double-focusing $\pi\sqrt{2}$ magnetic beta-ray spectrometer. The conversion electron spectrum of Am^{242m} is shown in Fig. 2. A spectrum containing essentially the same electron lines but which exhibited somewhat poorer resolution was observed by O'Kelley, et al.[2] The energies and relative intensities of the electron lines of Am^{242m} are listed in Table II. The absolute uncertainty on the electron energies is ± 1.5 kev. The present interpretation of these conversion electron lines indicates they arise from the L shell conversion of two gamma rays.

Table I. X-rays following the decay of Am^{242m}

Line	Transition	Observed energy (kev)	Siegbahn's extrapolated energy (kev)	Corrected relative intensity
Cm $L\alpha_2$	$L_3 - M_4$	14.75 ± 0.03	14.74	4
Cm $L\alpha_1$	$L_3 - M_5$	14.97 ± 0.03	14.96	27
Cm $L\beta_2$	$L_3 - N_5$	18.09 ± 0.02	18.10	16
Cm $L\beta_1$	$L_2 - M_4$	19.47 ± 0.02	19.38	100
Cm $L\gamma_1$	$L_2 - N_4$	22.79 ± 0.04	22.63	40
Cm $L\gamma_3$	$L_1 - N_3$	23.30 ± 0.06	23.25	6
Cm $L\gamma_6$	$L_2 - O_4$	23.62 ± 0.12	23.46	12
Am $L\alpha_2$	$L_3 - M_4$	14.44 ± 0.06	14.41	1
Am $L\alpha_1$	$L_3 - M_5$	14.61 ± 0.03	14.61	4
Am $L\beta_1$	$L_2 - M_4$	18.89 ± 0.02	18.80	6
Pu $L\alpha_2$	$L_3 - M_4$	14.08 ± 0.03	14.08	2
Pu $L\alpha_1$	$L_3 - M_5$	14.28 ± 0.03	14.28	10
Pu $L\beta_2$	$L_3 - N_5$	17.29 ± 0.03	17.25	9
Pu $L\beta_1$	$L_2 - M_4$	18.33 ± 0.03	18.27	27
Pu $L\beta_3$	$L_1 - M_3$	18.62 ± 0.04	18.52	7
Pu $L\gamma_1$	$L_2 - N_4$	21.46 ± 0.04	21.38	16
Pu $L\gamma_3$	$L_1 - N_3$	22.06 ± 0.10	21.97	7
Pu $L\gamma_6$	$L_2 - O_4$	22.24 ± 0.10	22.13	7
Pu K x-rays		102		37

Table II. Electron energies and intensities for Am^{242m}

Gamma Energy (kev)	Electron Energy (kev)	Conversion shell	Intensities (arbitrary units)
41.0	17.3	Cm L_{II}	520
	22.0	Cm L_{III}	380
	35.5	Cm M_{II}	380
	36.5	Cm M_{III} or M_{IV}	
	39.9	Cm N (Pu M?)	100
43.3	20.9	Pu L_{II}	210
	25.5	Pu L_{III}	150
Cm L x-rays	~16.5	Cm M and N	500

The two most abundant lines were assigned to the L conversion of a 41 kev gamma ray in Cm^{242}. The 43 kev gamma ray is thought to be present in the electron capture branch of the decay to Pu^{242} giving rise in part to the observed L x-rays of plutonium. The conversion electron intensities yield a ratio of curium to plutonium L shell vacancies of 2.5. It should be noted that these intensities are subject to uncertainties in resolution, scattered electron background, and an unknown contribution from L Auger electrons. The L_2/L_3 conversion ratios are 1.4 ± 0.4 for each gamma ray. Since El and most magnetic radiations are expected to exhibit marked L_1 conversion, the radiation is most likely electric quadrupole or higher multipole electric radiation. The assignment of E2 character to both transitions is consistent with the theoretical calculations of Gellman et al.[10] and the regularities noted among first excited states of even-even nuclei.[11]

Two gamma rays of 42.3 and 44.8 kev energy were barely discernible using the bent crystal spectrometer. Their energy difference compares favorably with a 2.3 kev difference from conversion electron data. Experiments performed by Church give energies of 42.2 ± 0.3 and 44.6 ± 0.3 kev, respectively, for the gamma rays following Am^{242m} beta and electron capture decay.[3] These energies agree well with those from bent crystal spectrometer measurements and are considered to be the best values. The absence of conversion electron lines of any other gamma ray is consistent with the low intensity of the isomeric transition but does not preclude an isomeric transition of extremely low energy.

The beta spectrum of each isomer was determined with separate samples. Results agreed with previous determinations,[2] the beta end point of Am^{242m} occurring at 620 ± 10 kev while that of Am^{242} was at 585 ± 10 kev. Both spectra produced straight-line Fermi-Kurie plots within experimental error. No conversion electron lines were observed in the beta spectrum of the long-lived Am^{242} sample which could not reasonable be ascribed to the 60 and 43 kev transitions in the decay of Am^{241}. Beta-L x-ray

coincidence measurements were performed on both isomers using an anthracene crystal for detecting beta particles and a thallium-activated sodium iodide crystal with a beryllium window for observing L x-rays. The L x-rays due to the alpha decay of Cm^{242} which has a branching ratio of 26.3 percent to the first excited level of Pu^{238} were used as a standard in determining the branching ratios of the Am^{242} isomer beta decay. A comparison of an Am^{242m} sample with the Cm^{242} standard gave a branching ratio of 51 ± 5 percent to the first excited state of Cm^{242} in the beta decay of Am^{242m}, after suitable corrections were made for differences in fluorescence yield and absorption of L x-rays. From a similar measurement on Am^{242}, a branching ratio of 45 ± 10 percent was calculated. The remainder of the beta transitions, in each case, proceed to the ground state of Cm^{242}. Thus the beta spectrum of each isomer should consist of two components differing by approximately 43 kev in energy. These components were not resolved experimentally. If it is assumed that the measured beta particle energies are the respective ground state transitions, the separation between the two Am^{242} isomers is approximately 35 kev. However, in view of the uncertainties in the beta spectrum endpoints and the inability to resolve each spectrum into its components, the energy spacing between the Am^{242} isomers is rather uncertain.

The branching ratio $\beta^-/EC = 4.2$ for Am^{242m} has been reported from an analysis of products from a long neutron irradiation of Am^{241}.[12] The ratio of curium to plutonium L shell vacancies has been determined by two experimental methods, L x-ray (3.6) and conversion electron (2.5) intensities. These data may be used in conjunction with the experimentally known ratio of beta decay of Am^{242m} to the ground and first excited states of Cm^{242} and the experimental β^- to electron capture branching ratio to calculate an electron capture branching ratio to the ground and first excited states of Pu^{242}. Using L x-ray intensity data, the electron capture branching of Am^{242m} to the first excited state of Pu^{242} is 60 percent, while if the conversion electron intensity data are used, the branching decay to the first

excited level of Pu^{242} is 87 percent. The lower value, 60 percent, is thought to be more accurate in that the electron capture branch of the Am^{242m} decay is expected to be very similar to the β^- branch since the energy levels of the daughter nuclei, Pu^{242} and Cm^{242}, are similar and the energies for beta decay and electron capture (closed decay cycles[13]) are nearly equal.

The K electron capture branching of Am^{242} was detected with the scintillation spectrometer by observing plutonium K x-ray radiations in a sample of americium 241, 242, and 243 from which the Np^{239} had been removed chemically. (The product of Am^{243} alpha decay, Np^{239}, has prominent plutonium K x-rays associated with it.) It was also necessary to employ a silver absorber to decrease the intensity of the 60 kev gamma ray of Am^{241}. Making suitable correction for absorption, escape peak, and the presence of a 75 kev gamma ray of Am^{243}, the ratio of K x-rays to 60 kev gamma was calculated. Combining this information with the known mass composition of the sample, a ratio of β^- to K electron capture of approximately 9 was calculated for Am^{242}.

III. DECAY SCHEME

The decay scheme of the Am^{242} isomers is shown in Fig. 3. The energies of the first excited levels of Pu^{242} and Cm^{242} compare favorably with those of other even-even nuclei of plutonium and curium.[14] In addition, the complex alpha structure of Cf^{246} shows an energy spacing of 42.7 kev for the first excited level of Cm^{242} which is in excellent agreement with the value determined here.[15] Using the measured beta energy and branching ratio, the log ft for both beta transitions of Am^{242m} is 6.9. Similarly, the log ft for electron capture to both levels of Pu^{242} is 7.3 as calculated from equations for allowed f[16] using energies derived from closed decay cycle calculations. These log ft values indicate first forbidden transitions, $\Delta I = 0, 1$ yes, for both beta and electron capture decay. The long-lived Am^{242} has a log ft of 11.6 for beta decay to both levels of Cm^{242}. Although the percentage of

electron capture decay to the ground and first excited state of Pu^{242} is unknown for this isomer, a log ft of 12.3 for K electron capture may be calculated if one assumes approximately equal population of these two levels. These rather large log ft values for Am^{242} may indicate second forbidden transitions.

The spins of the two Am^{242} isomers are still rather uncertain. Since Am^{242m} decays to both the ground and first excited states of its daughters with approximately equal probability as evidenced by the ft values, a spin of unity with odd parity would be consistent with the first forbidden beta and electron capture decay. There are less data available on the decay of the long-lived Am^{242}. Since the beta decay of this isomer also apparently populates the ground and first excited states of Cm^{242} with equal probability, a relatively low spin is indicated. Inconsistent with this postulate of low spin are spin changes of at least two units with no parity change required by the large ft values for the beta branching of this isomer. The absence of an appreciable isomeric transition may be explained either by a very long halflife due to a large spin difference between isomers or by a very small energy difference between isomers which would make the isomeric transition difficult to detect. At present the problem of spin and parity assignments must be left unresolved. Perhaps the most useful information for solving this problem would be a further study of the decay characteristics of the long-lived Am^{242}.

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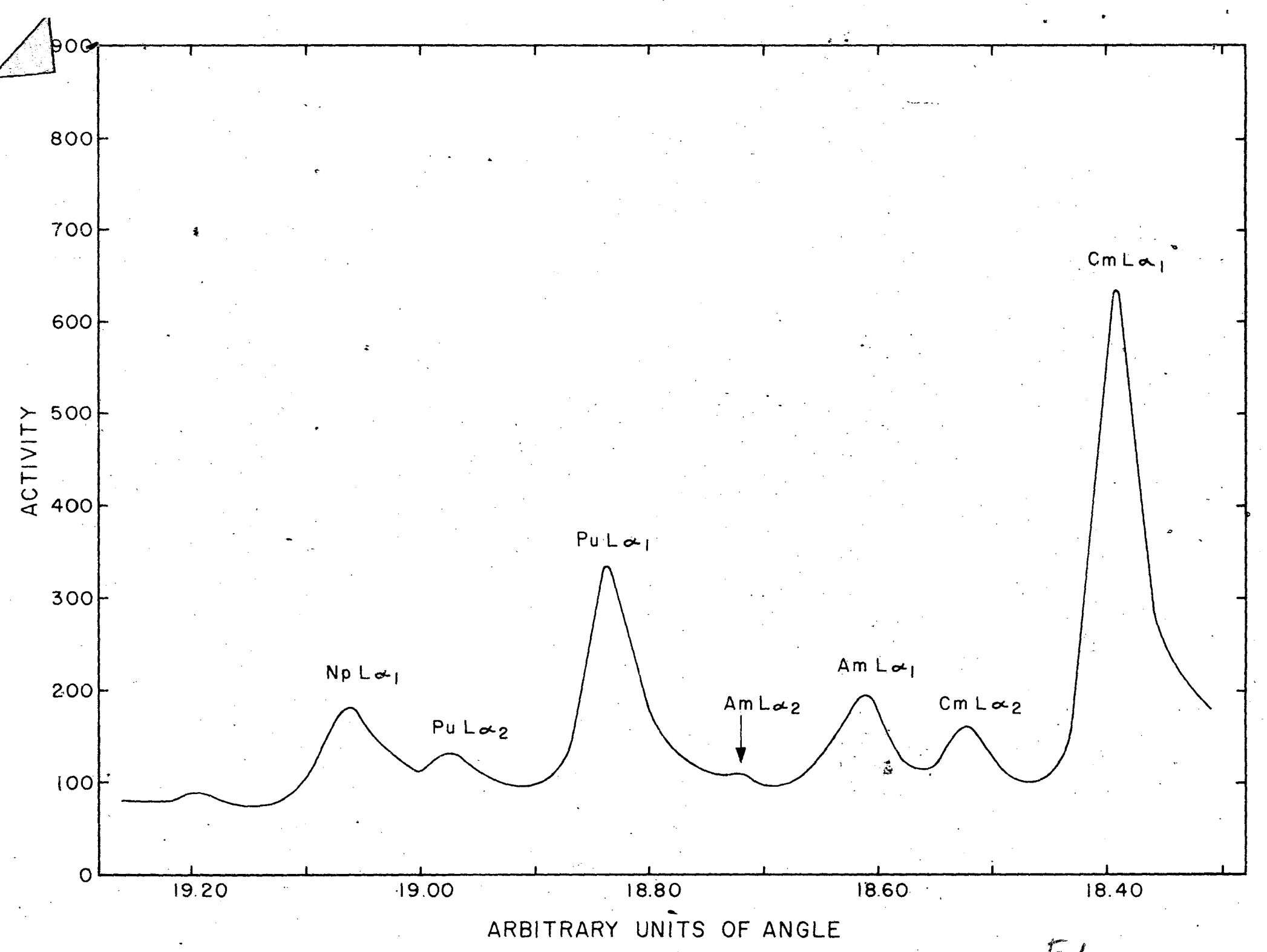
Figure Captions

Fig. 1. L α x-ray spectrum of a sample containing Am^{241} and Am^{242m}.

Fig. 2. Conversion electron spectrum of Am^{242m}.

Fig. 3. Decay scheme for the isomers of Am^{242}.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Cm Lα1
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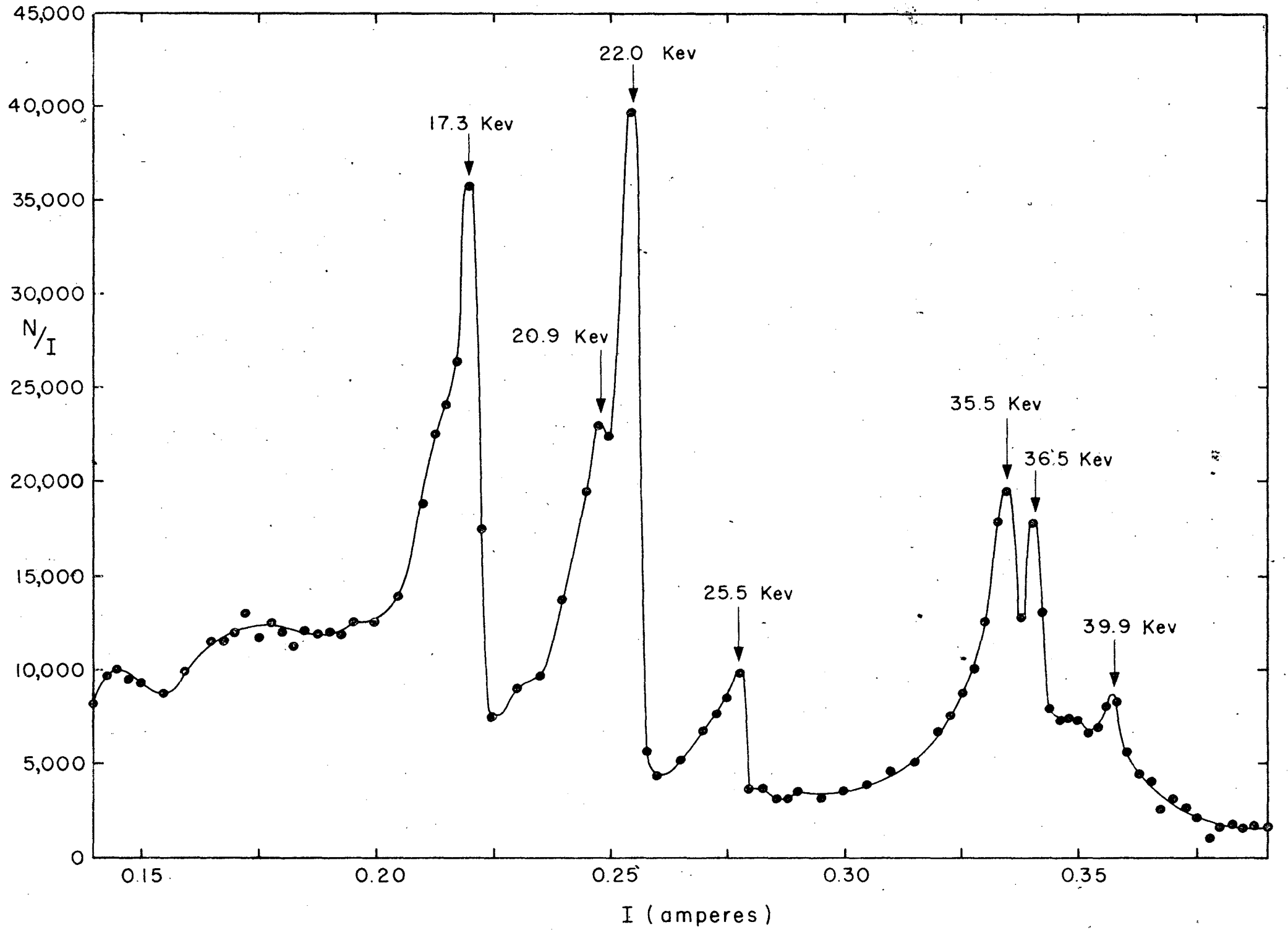
18.40
ARBITRARY UNITS OF ANGLE

F-1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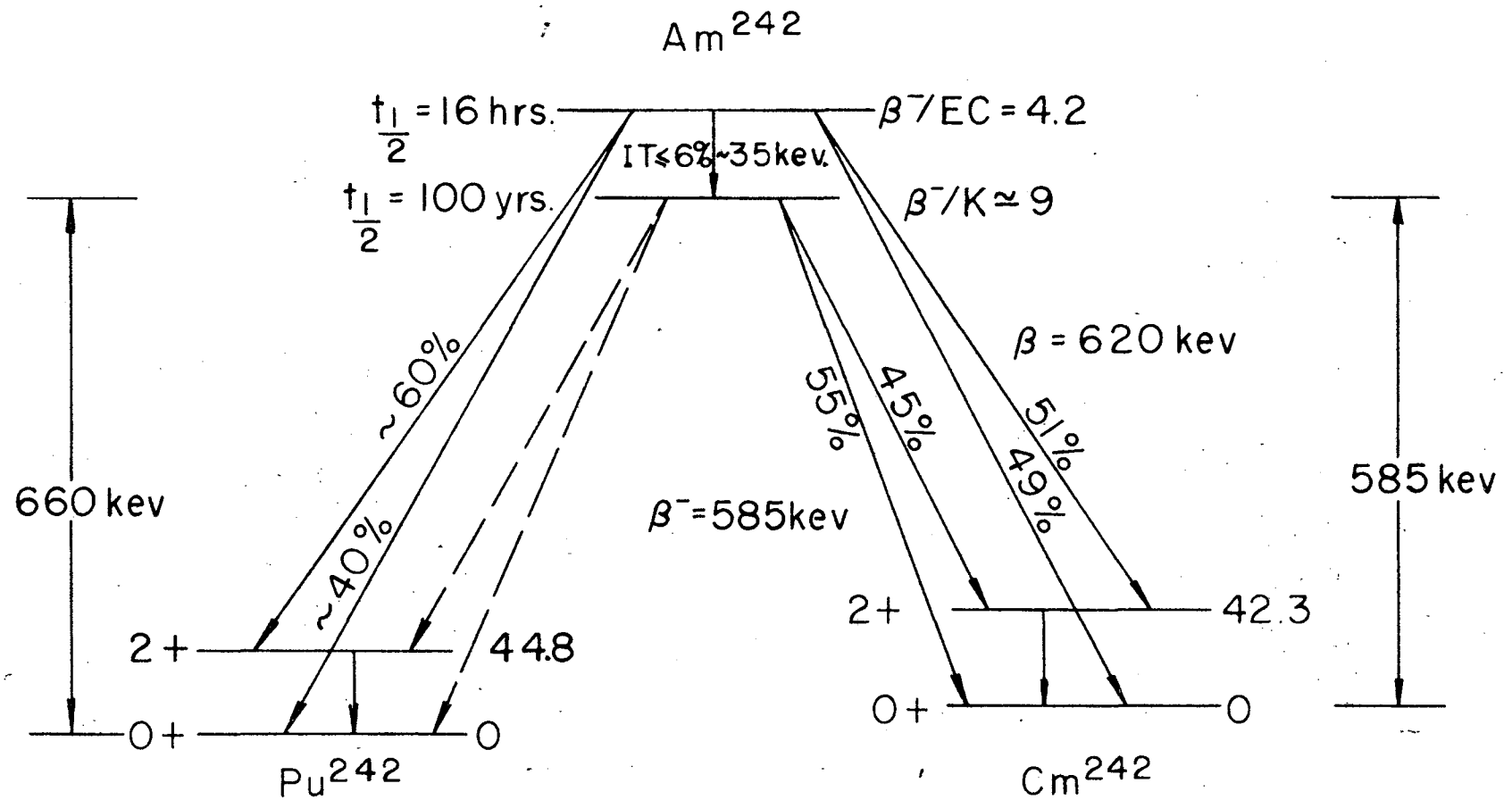
Am242
t1/2 = 16 hrs.
β⁻/EC = 4.2
IT≤6%~35kev.
t1/2 = 100 yrs.
β⁻/K ≃ 9
~60%
~40%
β = 620 kev
55%
45%
51%
49%
660 kev
β⁻=585kev
585 kev
2+
44.8
0+
0
Pu242
2+
42.3
0+
0
Cm242

F-3

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