Radiations from Indium (114) and Barium (140)*

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Indium (114) emits soft conversion electrons (range ~20 mg/cm²) and hard nuclear beta-rays of energy 1.89 Mev. Lead absorption gives quantum energies of 0.15 Mev and 0.70 Mev whereas coincidence absorption yields a maximum quantum energy of 0.90 Mev. The harder quanta are associated with an inner beta-ray group constituting less than 1.5 percent of the total beta-radiation, if with any group at all. A gamma-gamma-coincidence rate of $(0.25\pm0.02)\times10^{-3}$ coincidence per gamma-ray was noted.

Barium (140) emits nuclear beta-rays having a maximum energy of 0.91 Mev and two gamma-rays having quantum energies of 0.14 Mev and 0.6 Mev as indicated by absorption in lead. Beta-gamma-coincidence data, obtained from Ba^{140} in equilibrium with La^{140} and from Ba^{140} freshly separated from its daughter element, show that the beta-ray spectrum is complex and that about twenty percent of the disintegrations proceed by way of the inner beta-ray group which is coincident in time with gamma-radiation.

See note added in proof.

INTRODUCTION

THE radiations of the isomers of In^{114} have been studied intensively by many investigators. Lawson and Cork[1,2] report a maximum beta-ray energy of 1.98 Mev (cloud chamber and magnetic field) for the 72-second In^{114}. The value for the converted gamma-ray of $*In^{114}$ was reported as 0.19 Mev[2]. A more recent classified report,[3] gives spectrometric values of 1.98 Mev and 0.186 Mev for these radiation energies. A recent report by Cork *et al.*[4] gives 190.9 kev as the energy of the highly converted gamma-ray of $*In^{114}$ and 0.81 Mev as the energy of a relatively hard gamma-ray emitted by the two isomers in equilibrium.

No comment is made in their paper regarding the presence of the harder gamma-ray. Its energy value, obtained by lead absorption, is simply quoted in their Table I. The presence of the hard quantum stimulated some interest in the matter on the part of the writers so that some radioactive indium was obtained from the Oak Ridge pile for investigation. Chemical separations were carried out for the removal of Ag, Cu, Fe, Pb, Ca, and Ni as contaminants indicated by spectrographic analysis.

The historic barium (140), parent element of the 40-hour lanthanum (140), is among the products of uranium fission.[5] Among the few papers in the open literature dealing with this activity are an absorption value of 1.2 Mev and a spectrometric measurement giving 1.05 Mev[6] as the maximum energy of the nuclear beta-rays. The gamma-radiation is reported to have an energy of 0.54 Mev. Classified measurements include beta-ray end points at 0.4 Mev and 1.0 Mev and lead absorption measurements, giving a quantum energy of 0.5 Mev.[7] Still other classified measurements give an absorption limit of 1.1 Mev for the beta-rays[8] and a spectrometric value of 0.529 Mev for the hard gamma-ray.[9] Additional classified data[10] give good agreement with the values already quoted.

For this investigation, Ba^{140} was produced in the fission process at the Oak Ridge pile. Measurements of radiation energies and coincidence rates of Ba^{140} were carried out when it was in equilibrium with La^{140} as well as when freshly separated from it.

A complete description of the general methods and techniques used in obtaining the data to follow has been previously given.[11]

In^{114}

A thin source of the chemically purified $*In^{114}-In^{114}$ was placed before a single G-M counter, and the absorption curve of Fig. 1 was obtained. The end point, occurring at 0.87 g/cm² in aluminum, corresponds to a maximum energy of 1.89 kev as calculated by Feather's equation.[12] In the region of small absorber thickness, (< 20 mg/cm²), the conversion electrons of the 48-day metastable state of $*In^{114}$ are clearly evident. The curve closely resembles one previously published by Lawson and Cork.[2] If the portion of the curve relating to the 72-second period is extrapolated to zero absorber thickness, it can be shown that the number of nuclear beta-rays present is about equal to the number of conversion electrons, indicating a conversion coefficient of the order of one hundred percent.

The gamma-radiation of the purified indium source was absorbed in lead as plotted in Fig. 2. It is immedi-

* Assisted by the joint program of ONR and the AEC.

[1] J. L. Lawson and J. M. Cork, Phys. Rev. **56**, 291 (1939).

[2] J. L. Lawson and J. M. Cork, Phys. Rev. **57**, 982 (1940). Other references to measurements carried out prior to 1940 may be found in this paper.

[3] P. W. Levy, Plutonium Proj. Rept. Mon P-250 (February, 1947), p. 26, quoted by Seaborg and Perlman in their recent isotopic table.

[4] Cork, Shreffler, and Fowler, Phys. Rev. **74**, 1657 (1948).

[5] O. Hahn and F. Strassman, Naturwiss. **27**, 11 (1939).

[6] W. Rall and R. G. Wilkinson, Phys. Rev. **71**, 321 (1947).

[7] D. W. Engelkemeier, Plutonium Proj. Rept. CC-1959 (August, 1944), quoted by Seaborg and Perlman.

[8] H. A. Levy and L. G. Stang, Plutonium Proj. Rept. CC-1204, (January, 1944), p. 9, quoted by Seaborg and Perlman.

[9] V. A. Nedzel and M. B. Sampson, Plutonium Proj. Rept. CC-2283 (October, 1944), quoted by Seaborg and Perlman.

[10] Plutonium Proj. Rept., "Nuciei formed in fission," Rev. Mod. Phys. **18**, 513 (1946).

[11] C. E. Mandeville and M. V. Scherb, Nucleonics **3**, 2 (1948).

[12] N. Feather, Proc. Camb. Phil. Soc. **34**, 599 (1938).

ately clear that two gamma-rays are present. Resolution of the curve into its two components gives linear absorption coefficients corresponding to quantum energies of 0.15 Mev and 0.70 Mev. Taking into account the variation with energy of the quantum efficiency of the counter, it is estimated that the soft radiation is about six times as intense as the hard gamma-rays. The harder gamma-ray is undoubtedly that previously noted by Cork *et al.*[4] It cannot be said whether the softer quanta are emitted with the disintegration of the 72-second period or whether they are unconverted gamma-rays emitted at the 48-day level. Because of the inaccuracies of absorption methods, the latter possibility cannot be dismissed.

A source of $*In^{114}-In^{114}$ was placed behind a thick aluminum block so that recoil electrons ejected from the block by the gamma-rays would traverse two G-M counters in coincidence. Aluminum absorbers were placed between the two counters to obtain the coincidence absorption curve of Fig. 3. From a previously published calibration curve,[13] an energy of 0.90 Mev is deduced. The disagreement between the two values of the energy of the harder gamma-ray may be simply a result of the inaccuracies of absorption methods. When a source of radioactive indium was placed between two gamma-ray counters in coincidence, a gamma-gamma-coincidence rate of $(0.25\pm0.02)\times10^{-3}$ coincidence per gamma-ray was measured. Several different geometries were employed, but the gamma-gamma-coincidence rate persisted, attesting the validity of the effect. A search was made for beta-gamma-coincidences, but

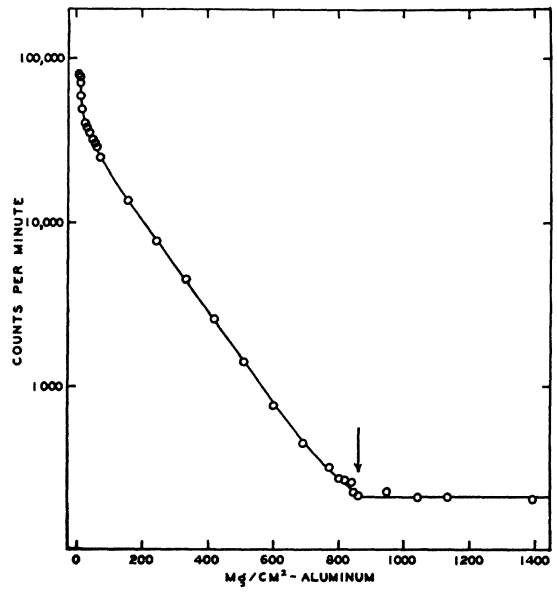

FIG. 1. Absorption in aluminum of the charged particles of $*In^{114}-In^{114}$. Soft conversion electrons and hard nuclear beta-rays are present. The end point corresponds to a maximum beta-ray energy of 1.89 Mev as calculated by Feather's equation.

[13] C. E. Mandeville and M. V. Scherb, Phys. Rev. **73**, 1434 (1948).

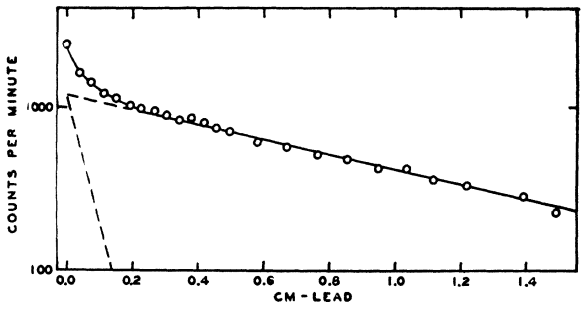

FIG. 2. Absorption in lead of the gamma-rays of indium (114). The linear absorption coefficients taken from the curve correspond to quantum energies of 0.15 Mev and 0.70 Mev.

none were detected. Assuming that a genuine coincidence rate as large as the accidental rate could be detected, knowing the calibration of the gamma-ray counter of the beta-gamma-coincidence counting arrangement, and assuming that the beta-rays of a low intensity inner beta-ray group are followed by about 0.8 Mev of gamma-ray energy, it was concluded that the softer beta-rays have an intensity less than 1.5 percent of the principal group at 1.89 Mev. This conclusion rests upon the assumption that the hard gamma-rays are indeed coupled with a nuclear beta-ray spectrum. Naturally, it cannot be said whether the supposed beta-ray spectrum originates at the 72-second level or at the 48-day level.

Ba^{140}

The aluminum absorption curve of the beta-rays of Ba^{140}, separated from La^{140}, is shown in Fig. 4. The measurements were completed within 40 minutes after the chemical separation. The end point corresponds to a maximum beta-ray energy of 0.91 Mev.

The gamma-rays of Ba^{140} were absorbed in lead as shown in Fig. 5. The starting time of each curve after the initial separation of barium from lanthanum is indicated at the right of the drawing. Each curve was completed in less than twenty minutes after the starting time; that is, the first curve was commenced five minutes after chemical separation and completed within twenty-five minutes after separation. It is evident that the first curve, the five minute curve, is readily analyzed into two components. The linear absorption coefficients taken from the curve correspond to quantum energies of 140 kev and 0.6 Mev, respectively. These values are only approximate since they are subject to the inaccuracies of the absorption method. It appears, however, that the softer gamma-ray has not been previously reported. The curves of Fig. 5 show how the gamma-count increases with time and with the growth of La^{140}, a hard gamma-ray emitter. The curvature in the region of low energy becomes less and less apparent with time and as the activity of the daughter element increases. This shows that the soft radiation, so clearly evident on the curve taken five minutes after

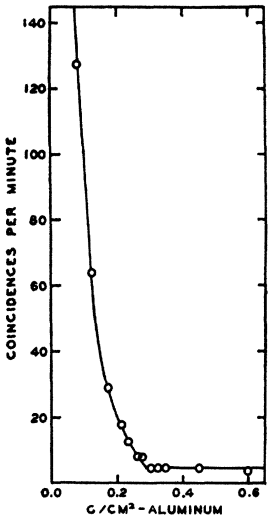

FIG. 3. Coincidence absorption of the recoil electrons of the gamma-rays of In^{114}. The quantum energy calculated from the end point is 0.90 Mev.

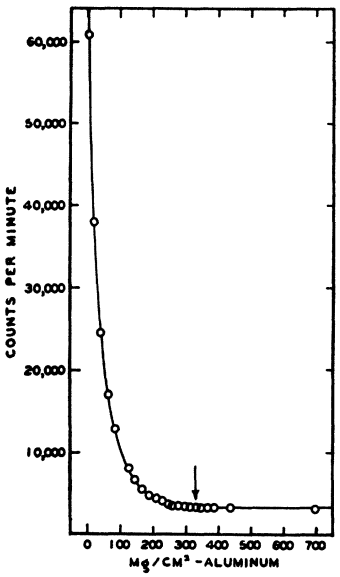

FIG. 4. Absorption in aluminum of the beta-rays of Ba^{140}. The end point corresponds to an energy of 0.91 Mev as calculated by Feather's equation.

chemical separation, is definitely emitted in the disintegration of Ba^{140}.

The beta-gamma-coincidence rate of Ba^{140}, in equilibrium with La^{140}, is shown in curve *A* of Fig. 6. The curve is seen to rise from an extrapolated value of 1.0×10^{-3} coincidence per beta-ray at zero absorber thickness to 1.8×10^{-3} coincidence per beta-ray in the vicinity of 0.34 g/cm². The coincidence rate beyond this absorber thickness is characteristic of La^{140} alone. Below 0.34 g/cm², the coincidence rate of La^{140} is reduced by the presence of the beta-rays of Ba^{140} which are obviously accompanied by much less gamma-ray energy than are those of La^{140}. Were the beta-rays of Ba^{140} accompanied by no gamma-rays whatever, the beta-gamma-coincidence rate at zero absorber thickness would be exactly half as great as it is beyond 0.34 g/cm². From the curve, it is seen that this is not quite the case. This fact alone shows that a small number of beta-gamma-coincidences are present in the disintegration of Ba^{140}.

Curve *B* is a plot of the genuine beta-gamma-coincidence rate of Ba^{140} alone. Barium was chemically separated from lanthanum at each point of the curve. Approximately fifteen minutes were required to prepare and mount the barium source after each separation. Coincidences were observed for about eight minutes at each point so that each measurement was completed in less than twenty-five minutes after the chemical separation. Only brief observations could be made at each point of curve *B* because of the rapid growth of La^{140}. Even very small traces of La^{140} are sufficient to invalidate any Ba^{140} measurements. This results from the fact that La^{140} is characterized by radiation energies and coincidence rates which are very high as compared with those of pure Ba^{140}. The aforementioned brevity of the coincidence measurement at each point of curve *B* made statistical accuracy difficult, and measurements were almost impossible to carry out at appreciable absorber thicknesses. Although the coincidence rate appears to be decreasing with increasing absorber thickness, the statistical accuracy of the points at larger absorber thickness makes it impossible to say that the coincidence rate eventually decreases to zero to indicate a complex spectrum. However, this conclusion can be reached by another rather simple argument.

It has been previously shown[13] in this laboratory that although the beta-ray spectrum of La^{140} is complex, the beta-gamma coincidence rate appears to be constant, independent of the beta-ray energy. It was also concluded that on the average, each beta-ray of La^{140} is followed by 2.3 Mev of gamma-ray energy.* Extra-

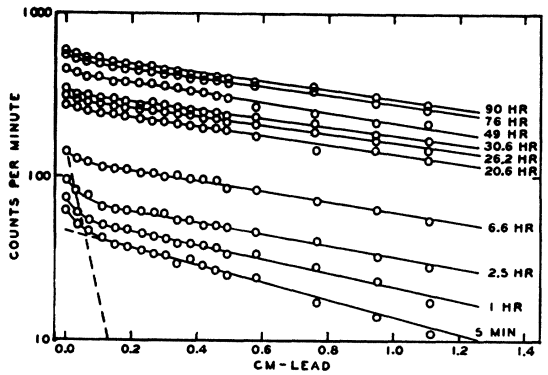

FIG. 5. Absorption in lead of the quantum radiations of $Ba^{140}-La^{140}$. The time after the initial chemical separation at which each curve was commenced is given at the right of the figure. A time of less than twenty minutes was required to complete each curve. The curve begun five minutes after the separation is analyzed into two components having energies of 140 kev and 0.6 Mev.

* In connection with the gamma-radiation of La^{140}, it should be mentioned that the energy of the hardest gamma-ray was erroneously reported as 2.16 Mev (see reference 13). This estimate was obtained from a preliminary calibration curve for the coincidence counting set which was based upon an unduly low value for the maximum energy of the gamma-rays from Ga^{72}. Taking the energy of the hardest gamma-ray of Ga^{72} to be 2.51 Mev, the maximum energy of the gamma-rays of La^{140} becomes 2.38 Mev.

polation of curve B to zero absorber thickness gives a beta-gamma-coincidence rate of 0.11×10^{-3} coincidence per beta-ray. Since curves A and B were taken with a fixed geometry common to both curves, $1.8/0.11 = 2.3/0.14$.** In other words, at zero absorber thickness, the beta-rays of Ba^{140} are followed on the average by 0.14 Mev of gamma-ray energy. Therefore, the 0.54 Mev gamma-ray cannot be associated with each beta-ray of Ba^{140}. The obvious explanation is that the beta-ray spectrum of Ba^{140} is complex.*** From the coincidence data given herein, it can be concluded that the inner beta-ray group constitutes about twenty percent of the totality of the beta-rays emitted by Ba^{140}. This calculation is based upon the assumption that the harder beta-ray group is associated with a ground state transition in Ba^{140} and hence non-coincident with any gamma-rays. This assumption may be only an approximate truth, because the position of the 140 kev gamma-rays in the disintegration scheme of Ba^{140} is not known. This estimate of relative intensity of the beta-ray spectra is in good agreement with one previously obtained from aluminum absorption and Feather analysis.[7] Returning to Fig. 5 it can be seen that the five minute curve gives 160:480 as the ratio of counts arising from the 0.14 and 0.6 Mev quanta. Assuming that the G-M counter is four times as efficient for the hard radiation as for the soft quanta, the ratio becomes 640:480. Thus, the two quanta are of roughly equal intensity and may be in cascade. Difficulties relating to even minute quantities of La^{140} made it impossible to make reliable gamma-gamma-coincidence measurements in Ba^{140}.

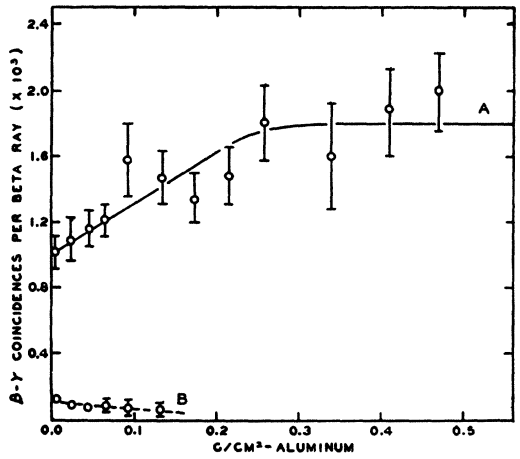

FIG. 6. Curve A is a plot of the beta-gamma-coincidence rate of Ba^{140} in equilibrium with La^{140} as a function of the surface density of aluminum placed before the beta-ray counter. Curve B is the coincidence rate of Ba^{140} alone. A chemical separation of Ba^{140} from La^{140} was made just prior to taking each point of curve B.

APPENDIX I. CHEMICAL PROCEDURE FOR THE PURIFICATION OF INDIUM

Neutron irradiated indium foil plus additional carrier elements silver, copper, iron, lead, calcium, and nickel, known from a spectrographic analysis to be present in the indium as impurities, were dissolved in 6N nitric acid. Potassium chloride was added to the solution, precipitating all of the silver and part of the lead. These chlorides were removed by filtration, and the filtrate was made slightly ammoniacal, precipitating indium, iron, residual lead, copper, and nickel as hydroxides. The filtrate, deep blue in color from the ammonia complexes of copper and nickel, contained no appreciable activity and was therefore discarded. The hydroxides were dissolved in 6N sulfuric acid, and the acid solution was reduced with 30-mesh zinc pellets, precipitating indium, copper, lead, and nickel as metals. Repeated reductions with zinc were necessary to reduce all of the indium. The metals were dissolved in 6N nitric acid and converted to sulfates by adding 1 cc of 18N sulfuric acid and evaporating to SO_3 fumes. The solution was diluted to 10 percent sulfuric acid and filtered to remove the insoluble lead sulfate. The sulfate solution of indium and traces of copper and nickel was made slightly ammoniacal and filtered. The precipitate was dissolved in two cc of 3N hydrochloric acid, and the acid solution was nearly neutralized with ammonia. Five grams of sodium bisulfite was added, and the solution was boiled for 15 minutes, then filtered. The precipitate of basic indium sulfite was washed with cold water, dried at 100°C and used in the experiments.

APPENDIX II. SEPARATION OF BARIUM FROM LANTHANUM

Barium chloride was precipitated from the barium-lanthanum solution with 12N hydrochloric acid. The precipitate was separated from the supernatant liquid by centrifugation, washed with 6N hydrochloric acid, dissolved in hot water, and reprecipitated as barium chloride with 12N hydrochloric acid, centrifuged and counted.

Note added in proof: Since this paper was submitted for publication, the measurements of Boehm and Preiswerk (Helv. Phys. Acta **22**, 331 (1949)), have appeared. They report gamma-gamma-coincidences but no beta-gamma-coincidences in In^{114}, in agreement with the findings of the writers. By critical absorption, they have shown that the hard gamma-rays of In^{114} are related to the K-capture process.

** It is here supposed that the energy-quantum efficiency curve of the gamma-ray counter is linear with energy so that the beta-gamma coincidence rate is also proportional to the average amount of gamma-ray energy associated with the beta-rays. This is a good assumption, because the gamma-ray counter is constructed of materials made of the light elements.

*** In making beta-gamma-coincidence measurements on both In^{114} and Ba^{140}, the coincidence resolving time was varied from 0.10 microsecond to 1.0 microsecond. No change in the genuine beta-gamma-coincidence rate of either element was noted. The small beta-gamma-coincidence rate of Ba^{140} and the lack of coincidences in In^{114} therefore cannot be explained by a time delay in this time interval.