APPENDIX 8

Decay of Cerium-144

Research Report

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As part of an earlier program of investigation in this laboratory, studies were made of the gamma ray spectrum and the beta ray spectrum of cerium-144^(1,2). In the present work, some coincidence studies were made on one of the beta groups appearing in the cerium-144 decay and on the gamma rays appearing in the deexcitations from the energy levels of praseodymium-144.

Sources of cerium-144 were prepared from carrier free radioactive cerium-144 as supplied by the Oak Ridge National Laboratory. The sample material was more than two years old at the time of preparation of sources. No additional chemical purification was attempted. Sources for use in the beta crystal spectrometer were mounted on thin Formvar film on spectrometer ring mounts.

The gamma ray spectrum of cerium-144 in the energy range 20 kev to 180 kev is shown in Figure 1. This spectrum was determined using a 2" by 2" NaI(Tl) crystal. The pulse spectrum was analyzed by a Radiation Instrument Development Laboratory (RIDL) 200 channel analyzer. The spectrum gives clear evidence of gamma ray peaks at 34 ± 3 kev and 134 ± 2 kev. A rather broad peak at 80 kev is observed. An indication of a gamma ray group of energy near 100 kev is also shown. The resolution of the detecting assembly was 9.8 percent at 662 kev. The uncorrected relative intensities of the three groups of 34, 80 and 134 kev are 95, 35, and 100, respectively. These intensities are for the gamma radiation exclusive of internal conversion.

Gamma-gamma coincidence measurements were made using two of the 2" by 2" sodium iodide crystals placed at 90 degrees to one another. Gamma radiation of a particular energy was selected by means of a single channel analyzer and the 200 channel analyzer was used to analyze any coincident pulses from detector number two. Insertion of a 1.6 μ sec. delay in channel one gave direct observation of random coincidences under identical experimental conditions.

Coincidence spectra were taken setting the single channel analyzer for 34 ± 2 , 80 ± 2 , 100 ± 2 kev gamma radiation. Five hour true and random counts were taken at each setting. Figure 2 shows the spectrum of gamma radiation of cerium-144-

Figure 1 Cerium 144. Single Channel Spectrum of Gamma Rays in the Ce^{144} - Pr^{144} Decay.

Figure 2 Cerium 144. Spectrum of Gamma Rays in the Ce144-Pr144 Decay in Coincidence with the 34 keV Gamma Ray.

praseodymium-144 in coincidence with the 34 kev gamma radiation. There is evidence for coincident radiation of 34, 55, 82 and 101 kev energy with only weak coincidences at 135 kev energy. Figure 3 shows the gamma ray spectrum of cerium-144-praseodymium-144 in coincidence with the 80 kev gamma radiation. Here the expected coincidences with radiation of energy about 35 kev is observed as well as coincidences with gamma radiation of about 54 kev energy. There are no indications of coincidences with gamma radiation of energy greater than 54 kev and less than 180 kev (channels 100 to 199 are not shown). Figure 4 shows the gamma ray spectrum of cerium-144-praseodymium-144 in coincidence with the 100 kev gamma radiation. Here coincidences are observed with radiation of about 34 kev energy (channels 100 to 199 are not shown). No coincidences were observed between gamma rays of 34 kev and gamma rays of energy less than 180 kev (channel 100 to 199 are not shown) except for relatively weak coincidences with 34 kev.

Additional knowledge of the gamma ray decay of the excited states of praseodymium-144 was obtained by means of a new coincidence technique. With this technique, as in the case of the gamma-gamma coincidences reported above, the chance counting rates in each channel are determined under conditions exactly duplicating those used for getting the coincidence data. However, for random events channel one is delayed 1.6 μ sec. as before. The technique for measurement of coincidence gamma rays in cascade has been outlined by Hoogenboom⁽³⁾ and was adapted for use with the 200 channel analyzer here. A simple summation circuit was constructed. This circuit employs a cathode follower and balance control to sum the inputs from two scintillation detector phototubes. Isolation of the separate input signals from one another is maintained.

As the summed output of the separate input signals it is possible to make three selections: 1. signal A only, 2. signal B only, 3. signal A plus B, each depending on the output pulse amplitude selected. For random events in A and B

Figure 3 Cerium 144. Spectrum of Gamma Rays in the Ce 144-Pr 144 Decay in Coincidence with the 80 keV Gamma Ray.

Figure 4 Cerium 144. Spectrum of Gamma Rays in the Ce144—Pr144 Decay in Coincidence with the 100 kev Gamma Ray.

corresponding to scintillations from gamma rays of energy E_A in A and E_B in B, the output of this circuit will consist of pulses corresponding to E_A , E_B and E_A plus E_B . If the pulse amplitude selector of channel one is set to observe only pulses of energy E_A plus E_B and this output is used to trigger the 200 channel analyzer only when a pulse of this energy appears (the input of the analyzer may be taken as the signal from 'A'), then sharp peaks will appear in the spectrum observed by the multichannel analyzer corresponding only to signals of energy E_A and E_B .

The use of the Hoogenboom method with the equipment in service here can be illustrated with cobalt-60. Here gamma rays of energy 1.17 and 1.33 Mev are in coincidence in all decays. Detectors A and B observe gamma rays of 1.17 and 1.33 Mev plus Compton and background radiation in both cases. The output of the summing circuit will then consist of pulses from the 1.17 and 1.33 Mev gamma rays plus summed pulses related to 2.50 Mev energy. When a coincidence occurs and the two detectors simultaneously feed pulses into the summation network a 2.50 Mev pulse does appear. The signals are balanced and the single channel analyzer is set to trigger the 200 channel analyzer only when a sum pulse appears. The analyzer then will distinguish the pulse in channel 'A' which caused the coincidence. For cobalt-60 this pulse may be either 1.17 Mev or 1.33 Mev. All Compton and scattered radiation will be excluded. Figure 5 shows the cobalt-60 spectrum obtained by this technique. The very good resolution here is obvious from the peak to valley ratio of more than eight to one.

In application of the above method to more complicated systems, for example, cerium-144-praseodymium-144, the data becomes only slightly more complex. When a crossover gamma ray is present a peak will always occur corresponding to this gamma ray energy because a signal from the crossover transition will give the proper pulse height to trigger the analyzer and the instrument will observe the crossover gamma ray as a single peak in the coincidence spectrum.

Figure 5 Cobalt 60. Gamma Rays in Coincidence Cascade from the -2.50 Mev Energy Level. In Coincidence: 1.17 Mev and 1.33 Mev.

In the cerium-144-praseodymium-144 decay the 134 keV level deexcites by emission of 134 keV gamma rays or by emission of two or more coincident gamma rays whose energy sum is 134 keV. Figure 6 is the spectrum of coincident gamma rays whose sum is 134 keV. The two detectors were placed at 90 degrees for this determination. Cascades of 101 ± 4 keV plus 33 ± 4 keV and 91 ± 4 keV plus 43 ± 5 keV are observed. Note that the 33 and 91 keV gamma rays do not give a 134 keV sum nor do the 43 and 101 keV gamma rays therefore, the latter pairs are not in coincidence. Figure 7 is the spectrum of coincident gamma rays the sum of which is 134 keV as observed in cerium-144-praseodymium-144 when the two detectors were placed at 180 degrees to each other. Cascades of 43 ± 3 keV plus 91 ± 2 keV and 33 ± 3 keV plus 101 ± 2 keV are observed. The peaks are more clearly defined in this case.

The beta-gamma coincidence spectrum of cerium-144 was measured using an anthracene crystal coupled to a multiplier phototube to detect the beta radiation. The gamma rays were detected by means of a sodium iodide crystal. The total and random coincidence rates were both directly determined as in the case for gamma-gamma coincidences by delaying the gamma channel for the randoms determination.

The beta spectrum in coincidence with 134 keV gamma radiation yields the coincidence Kurie plot shown in Figure 8. This plot shows a linear region from 80 keV to 150 keV and extrapolates to give an upper energy limit for the coincident beta group of 190 ± 5 keV. It was hoped that an inner beta group of endpoint energy less than 190 keV might be observed by this method assuming that the excited state to which the decay occurs deexcites to the 134 keV level since no crossover gamma radiation of energy over 134 keV was observed. The plot does not indicate that an inner beta group of endpoint energy less than 190 keV is present in the decay of cerium-144.

Recent publications have reported on the decay of cerium-144 to praseodymium-144. Kickok, McKinley and Fultz⁽⁴⁾ measured the beta spectrum and conversion electron spectrum and conclude that energy levels of 134 and 80 keV must exist in praseodymium-144

Figure 6 Cerium 144. Gamma Rays in the Decay of Ce 144—Pr 144 in Coincidence Cascade from the 134 keV Energy Level. In Coincidence: 91 and 43 keV, 101 and 33 keV.

Figure 7 Cerium 144. Gamma Rays in the Decay of Ce144-Pr144 in Coincidence Cascade from the 134 kev Energy Level. In Coincidence: 91 and 43 kev, 101 and 33 kev.

Figure 8 Cerium 144. Kurie plot of the Beta Spectrum in Coincidence with the 134 keV Gamma Ray.

Freeman⁽⁵⁾ has reported energy levels of 58, 80, 99, 134 and 166 kev. Sengupta, Bhattacharyya, Lahiri and Mukherjee⁽⁶⁾ reported energy levels of 80, 99 and 134 kev. Geiger, Graham and Ewan⁽⁷⁾ conclude from internal conversion measurements that energy levels of 133.4, 99.9, 80.0 and 59.0 kev exist in praseodymium-144. The latter researchers also report gamma rays as follows: 33.5(M1), 40.88(M1), 53.36(M1), 58.96(M3), 80.03(M1), 99.85(E2) and 133.42(M1) kev.

An attempt to fit the gamma energy measurements and gamma-gamma coincidence observations into a general decay scheme was made. The observations are consistent with energy levels at 58, 80, 100 and 134 kev as have been reported by previous investigators⁽³⁻⁶⁾.

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