Neutron-Deficient Activities of Praseodymium

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Neutron-deficient radionuclides of praseodymium were produced by proton bombardment of enriched isotopes of cerium and neodymium. Two unreported activities were found, 22-min Pr^{135} and 70-min Pr^{136}; the gamma and beta spectra of each were determined. Further study of 2.0-hr Pr^{138} and 4.5-hr Pr^{139} was also made, and gamma spectra of each are shown. The half-life of Pr^{137} is shown to be <5 min or >1 year. Confirmation of assignment of 3.4-min Pr^{140} is made.

PRASEODYMIUM-135

AN enriched stable isotope of Ce^{136} was bombarded with 22.4-Mev protons in the Oak Ridge National Laboratory 86-inch cyclotron. Ion exchange methods were used to separate the products.[1] Following separation, the praseodymium fraction was found to contain a 22-min activity, a 70-min activity, 4.5-hr Pr^{139}, and 19.3-hr Pr^{142}. A second bombardment was made with the energy of the incident proton beam adjusted to 9.5 Mev by use of aluminum absorbers. Following separation the praseodymium fraction was found to contain a 70-min activity and 19.3-hr Pr^{142}; thus the 22-min activity was produced by either a $(p,2n)$ or $(p,3n)$ reaction. The 22-min activity was not observed as a result of bombarding enriched stable Ce^{138} or natural cerium with either 22.4 or 9.5-Mev protons, thus eliminating its assignment to a mass higher than 136. In another bombardment of enriched Ce^{136} with 22.4-Mev protons the praseodymium fraction from an initial separation was permitted to decay; then, after a second separation, 22-hr Ce^{135} was found to be present in the cerium fraction. Thus, the 22-min activity is the parent of Ce^{135} and is assigned to Pr^{135}. A gamma-ray spectrum as measured with a NaI(Tl) scintillation spectrometer is shown in Fig. 1. The spectrum shows the presence of positron emission with gammas of 0.080, 0.22, and 0.30-Mev energy. Because of problems of counting efficiency and internal conversion it was not practical to distinguish between the x-rays due to electron capture and those due to internal conversion. However, it is reasonably certain that the activity decays by both electron capture and positron emission. The 0.033-Mev peak is the x-ray and the 0.51-Mev the annihilation peak. More energetic radiation was not observed. The gamma-ray spectrum was determined once every 20 minutes for ten half-lives and the contribution of the longer lived components of the praseodymium fraction subtracted in order to obtain a spectrum for the 22-min activity.

The maximum energy of the positron emitted was determined by use of a scintillation spectrometer consisting of a cylindrical anthracene crystal $1\frac{1}{2}$ in. in diameter and 1 in. in thickness mounted on top of a DuMont type 6292 photomultiplier. Pulses from the photomultiplier tube were fed into a preamplifier. These were further amplified by a linear amplifier and fed into a single-channel pulse height analyzer and a scaler. The beta spectrum was determined several times over a period of ten half-lives. Decay of each experimental point was plotted and the decay curve analyzed for the 22-min component. The gamma background was determined in a similar manner, by using an appropriate absorber, and subtracted. The results were plotted with counting rate *versus* pulse height as coordinates. A Kurie plot indicated a value of 2.5 ± 0.1 Mev. Comparison with a phosphorus-32 curve obtained under identical conditions gave agreement with this value. An aluminum absorption curve also agreed within these limits.

PRASEODYMIUM-136

As stated in the previous section, bombardment of enriched stable isotope Ce^{136} with 9.5-Mev protons followed by ion exchange separation gave praseodymium activities with half-lives of 70 min and 19.3 hr. The 3.4-min Pr^{140} had decayed by the end of the chemical separation and there was not enough Ce^{138} present to produce a detectable amount of 2.0-hr Pr^{138}. The

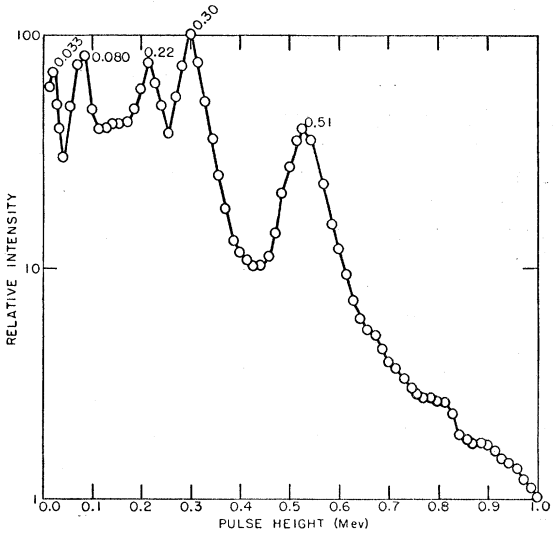

FIG. 1. Gamma-ray spectrum of 22-min Pr^{135}.

[1] B. H. Ketelle and G. E. Boyd, J. Am. Chem. Soc. **69**, 2800 (1947).

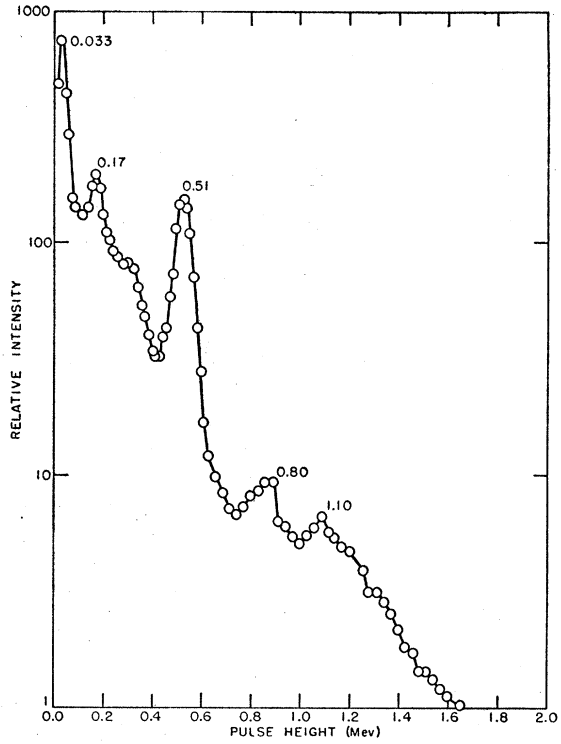

FIG. 2. Gamma-ray spectrum of 70-min Pr^{136}.

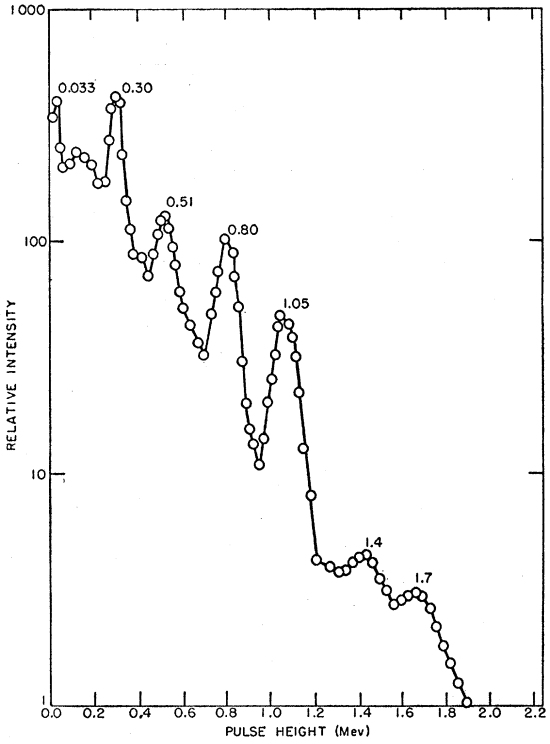

FIG. 3. Gamma-ray spectrum of 2.0-hr Pr^{138}.

19.3-hr Pr^{142} was produced by a (p,n) reaction on Ce^{142}. The 70-min activity was not observed as a result of bombarding enriched Ce^{138} or natural Ce with 9.5-Mev protons. This assigns the 70-min activity to Pr^{136}. A gamma-ray spectrum, Fig. 2, shows it to decay by positron emission with a gamma of 0.17-Mev energy, and with possibly more energetic gammas of low intensity with energies of 0.80 and 1.1 Mev. The energy of the positron was determined in the same manner as described for Pr^{135} in the previous section. It was found to be 2.0±0.1 Mev.

PRASEODYMIUM-137

A 1.4-hr activity has been reported by others[2] to be assigned to Pr^{137}. However, we were unable to confirm this assignment. Enriched stable isotope Ce^{138} was bombarded with 22.4-Mev protons and separation was performed by means of ion exchange, as before. This separation was completed within 2.5 hours after completion of bombardment. The praseodymium fraction was collected and cerium was added as carrier before the material was precipitated and recovered. After a delay of 7 hours a separation was again performed on the praseodymium fraction and the cerium fraction was recovered from the second separation. This fraction was examined by means of a gamma-ray scintillation spectrometer and the absence of the 0.26-Mev gamma ray associated with 36-hr Ce^{137} was confirmed. The x-ray peak and a 0.17-Mev peak were the only radiations observed, both of which are associated with 140-day Ce^{139}. The Ce^{139} is present because of the decay of 4.5-hr Pr^{139}. Decay of the cerium fraction has also been followed for 90 days and at this time shows a slope corresponding to a half-life of 140 days, thus proving that the 1.4-hr activity previously reported is not the parent of Ce^{137}. Since the unknown Pr^{137} and 4.5-hr Pr^{139} activities would be produced with approximately the same cross section, it is possible to calculate, with corrections for time of bombardment, dilution factors, chemical yields, and isotopic abundance of the Ce^{138} and Ce^{140} in the enriched sample of Ce^{138}, the maximum half-life of activities longer than 4.5 hours. From these results it can be said that the half-life of Pr^{137} is >1 year. In other short bombardments an upper limit of <5 min can be placed on the half-life of Pr^{137}. In summation, limits of >1 year or <5 min may be placed on the half-life of Pr^{137}.

PRASEODYMIUM-138

Bombardment of enriched stable isotope Ce^{138} with 9.5-Mev protons followed by ion exchange separations yielded two activities in the praseodymium fraction, 2.0-hr Pr^{138} and 19.3-hr Pr^{142}. Previous reports[2,3] on the half-life of Pr^{138} were confirmed with a value of 2.0±0.1 hr. The 2.0-hr activity was not observed by

[2] Dahlstrom, Foster, and Thompson [private communication to J. M. Hollander (October 1952)]; Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953).

[3] B. J. Stoner, Phys. Rev. **81**, 8 (1951).

bombarding natural cerium with 9.5-Mev or 22.4-Mev protons. This would assign to it a mass lower than 139 and, since it is produced by 9.5-Mev protons on enriched Ce^{138}, confirmation of assignment to Pr^{138} is also made. A gamma-ray spectrum, Fig. 3, shows the presence of positron emission. Gamma rays of 0.30-, 0.80-, and 1.05-Mev energy are in disagreement with values obtained by others.[3] The 0.033-Mev peak is the x-ray and the 0.51-Mev is the annihilation peak. Here again the fraction of x-rays associated with electron capture and internal conversion was not determined. There is also the possibility of more energetic gamma rays of 1.4- and 1.7-Mev energy and of a very low intensity relative to the others. Energy of the positron was measured with a scintillation spectrometer as previously described and by aluminum absorption curves. A value of 1.4±0.1 Mev was found, which is in agreement with that previously reported.

PRASEODYMIUM-139

Bombardment of natural cerium with 22.4-Mev protons followed by chemical separation yielded two praseodymium activities, 4.5-hr Pr^{139} and 19.3-hr Pr^{142}; the 3.4-min Pr^{140} decayed before completion of the separation. The 4.5-hr activity was not observed as a result of 9.5-Mev proton bombardment of natural cerium. It was also shown, in the section of this paper on Pr^{137}, to be the parent of Ce^{139}. Thus, its half-life and assignment are confirmed.[2] To obtain a sample containing less 19.3-hr Pr^{142}, neodymium enriched in mass 142 was bombarded with 17.5-Mev protons; the 4.5-hr Pr^{139} was produced by the (p,α) reaction on Nd^{142}. At this energy, the $(p,\alpha n)$ reaction is eliminated. The neodymium contained 93.0 percent Nd^{142} and 0.4 percent Nd^{145}. The 19.3-hr Pr^{142} would be produced from Nd^{145} by the reaction $Nd^{145}(p,\alpha)Pr^{142}$. However, calculations show it to be much less from this bombardment than from the (p,n) reaction on natural cerium. A gamma-ray spectrum, Fig. 4, shows the presence of positron emission with gamma rays of 0.17-, 1.3-, and 1.6-Mev energy. The 0.17-Mev gamma ray is the same as that associated with Ce^{139}. The fraction of x-rays due to electron capture was not determined. A gamma-ray spectrum of Pr^{139} produced by the reaction $Ce^{140}(p,2n)Pr^{139}$ agreed exactly with that shown in Fig. 4, which was produced by $Nd^{142}(p,\alpha)Pr^{139}$. The

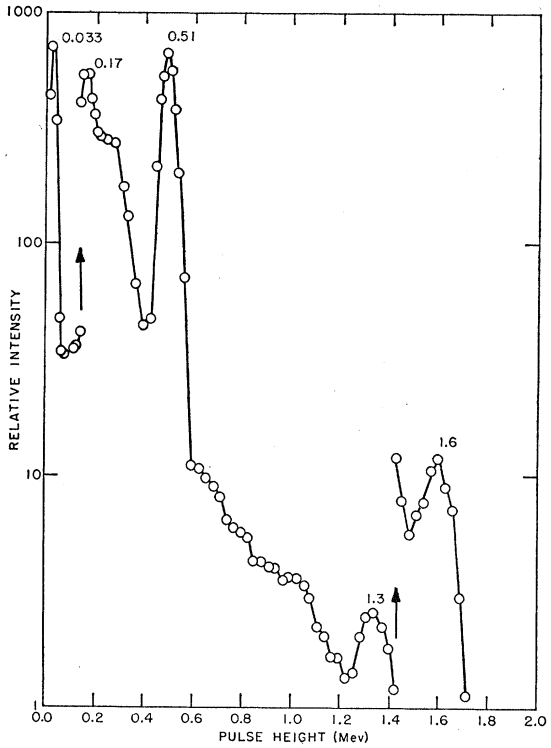

FIG. 4. Gamma-ray spectrum of 4.5-hr Pr^{139}.

values of the gamma-ray energies do not agree with those reported by others.[3] However, since this work shows agreement with two different methods of production, the values are assumed to be correct. The energy of the positron was determined by techniques already described and the 1.0- ±0.1-Mev value obtained is in agreement with that previously reported.

PRASEODYMIUM-140

Confirmation of assignment of a 3.4-min activity to Pr^{140} was made by bombarding natural cerium with 9.5-Mev protons; also by bombardment of praseodymium with 22.5-Mev protons, which resulted in the $Pr^{141}(p,pn)Pr^{140}$ reaction. No further attempt was made to study its decay characteristics because of experimental difficulties encountered due to the short half-life involved.