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EXTENSION OF ALPHA- AND BETA-DECAY SYSTEMATICS OF PROTACTINIUM ISOTOPES

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EXTENSION OF ALPHA- AND BETA-DECAY SYSTEMATICS OF PROTACTINIUM ISOTOPES

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January 30, 1950

As part of our program for obtaining data to extend the scope of the systematics of alpha-radioactivity[1] and to obtain more data pertaining to the energy surface in the heavy region of elements, we have made some pertinent new measurements on protactinium isotopes.

The heaviest isotope of protactinium hitherto reported is that of mass 234 and hence it would be of interest to know the beta-emission properties of heavier isotopes in order to tie them in with the known radioactive data in this general region. Since low-energy deuteron bombardment of U^{238} might be expected to lead to Pa^{235} and Pa^{236} by (d,αn) and (d,α) reactions and proton bombardment to Pa^{235} by the (p,α) reaction, these irradiations were made in the 60-inch cyclotron at energies of 19 and 9.5 Mev, respectively.

The protactinium was chemically separated following the bombardment of natural uranium by a procedure which involved a number of manganese dioxide cycles coupled with extractions of protactinium from aqueous into organic solvents. The manganese dioxide cycles consisted of precipitating this compound from the solution of uranium in nitric acid, followed by centrifugation, dissolution of the solid with hydroxylamine solution, dilution, and reprecipitation. The dissolved precipitate from the last cycle was acidified, salted with ammonium nitrate, and the protactinium extracted with diisopropyl ketone, several washings with salted solutions being made to insure good separation from fission products. The protactinium was then washed back into a low acidity aqueous solution and after

acidification was extracted into a benzene solution of thenoyltrifluoroacetone which forms a complex ion with the protactinium. This solution was then evaporated to dryness on a platinum counting plate leaving a weightless deposit of protactinium.

The protactinium from the 19-Mev deuteron bombardment contained 23.7 ± 0.5 minute and 27-day beta-particle emitters. The cross section for the formation of the former is about 2×10^{-27} cm^2 on the assumption that it is formed from U^{238} while that for the latter is about 4×10^{-27} cm^2 on the assumption that it is due to Pa^{233} produced in the reaction $U^{235}(d,\alpha)Pa^{233}$. A small amount of two intermediate periods were present but these most probably can be attributed to a small amount of zirconium and niobium fission product contaminants. The 23.7-minute activity was also found in the protactinium fraction from the bombardment of U^{238} ($U^{238}/U^{235} = 2300$) with 9.5-Mev protons in a yield corresponding to a cross section of about 3×10^{-29} cm^2. These observations are consistent with the assignment of the 23.7-minute activity to Pa^{235}, produced in the reactions $U^{238}(d,\alpha n)Pa^{235}$ and $U^{238}(p,\alpha)Pa^{235}$.

An aluminum absorption curve taken on this 23.7-min. activity shows that the beta-particles have a range of about 600 mg corresponding to an energy of about 1.4 Mev according to the Feather relationship.[2] Gamma-rays are either absent or present in amounts too small to detect by absorption experiments.

If one closes a decay cycle involving Pu^{239}, Np^{239}, Pa^{235}, and U^{235} using the values 0.7 Mev for the beta-disintegration energy[3] of Np^{239}, 5.24 Mev for the alpha-disintegration energy[3] of Pu^{239} and 4.6 Mev for the estimated alpha-disintegration energy[1] of Np^{239}, a beta-disintegration energy of 1.34 Mev is obtained for Pa^{235}. This measurement,

therefore, gives added weight to the somewhat uncertain measured beta- and estimated alpha-disintegration energies of Np^{239}, and hence adds to the reliability of data needed for the construction of the energy surface in this region.

We also were interested in determining the partial half-life of Pa^{230} for alpha-disintegration. A half-life of some hundreds to thousands of years would be predicted from the alpha-systematics by obtaining the alpha-disintegration energy from mass number versus energy plots (5.5 Mev) and taking into account the prohibition introduced by the presence of two odd nucleons in determining its alpha-disintegration rate.[1] In this experiment the alpha-particles of the Pa^{230} were not measured directly but its half-life for alpha-disintegration was inferred by measuring the equilibrium amount of its alpha-disintegration product, the beta-particle emitting 29-hour Ac^{226}, on the assumption that the predicted negligible branching decay of Ac^{226} by electron capture is correct. (The half-life of Ac^{226}, originally reported as 22 hours,[3] has been more recently determined to be 29 hours.[4])

The actinium was chemically separated from a sample containing a large amount of Pa^{230} and the Ac^{226} was identified and the amount measured through an alpha pulse analysis in which the alpha-particles of its daughters, 30-minute Th^{226} and daughters, were measured. In a bombardment of a thick thorium target with 60-Mev deuterons for 10 microampere-hours in the 184-inch cyclotron a sample of Pa^{230} corresponding to 1.3×10^6 negative beta disintegrations/minute (as determined by alpha pulse analysis of the daughter U^{230} decay chain) was chemically isolated as described above. The actinium was chemically separated from this protactinium fraction by carrying it on lanthanum and cerium fluorides which were

redissolved and reprecipitated several times and further purified after dissolution by removing thorium through repeated zirconium phosphate precipitations. At the end of the procedure, the cerium(III) was oxidized to cerium(IV) with sodium bismuthate, the cerium(IV) removed on a zirconium phosphate precipitate, and the actinium activity precipitated with a small amount of lanthanum fluoride. After dissolution in hydrochloric acid, the soluble chloride of the carrier when evaporated on platinum gave a thin plate suitable for alpha pulse analysis.

The actinium fraction from the protactinium containing 1.3×10^6 beta-disintegrations/minute of Pa^{230} contained 300 alpha-disintegrations/minute of Th^{226} which decayed with the 29-hour half-life of its beta-emitting Ac^{226} parent. Correcting for chemical yield (as determined by 10.0-day Ac^{225} tracer) and taking into account the fact that Pa^{230} has a half-life of 17 days[5] and also decays by electron capture in a ratio of 10 as compared to its decay by negative beta-particle emission,[6] these data lead to a partial half-life for alpha-emission of about 1400 years $\pm$ 20%, which corresponds to a prohibition by a factor of about 100 in the alpha decay, presumably due to the presence in Pa^{230} of two odd nucleons.[1]

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