Neutron-Induced Radioactivity of the Noble Metals

EDWIN MCMILLAN, MARTIN KAMEN
Radiation Laboratory, Department of Physics, University of California, Berkeley, California

AND

SAMUEL RUBEN*
Department of Chemistry, University of California, Berkeley, California

(Received July 19, 1937)

WE have started a study of the effects produced by neutrons on the elements in the neighborhood of platinum, and some of the results, although admittedly incomplete, seem sufficiently striking to merit publication at this time. Strong activities are induced in Ir, Pt and Aμ by slow neutrons, and in these metals and Hg by fast ($Li+H^2$) neutrons. Os, Tl, Pb and Bi give only comparatively weak activities with either type of neutrons. Our results so far on the strong activities, in order of increasing complexity, are given below.

MERCURY

This element has been reported to give a weak activity of 40 hr. half-life with slow neutrons,[1] and a strong activity of 43-min. half-life with fast neutrons.[2] We find the fast neutron 43-min. period to be indeed very strong, corresponding to an intensity with 10 μA of 5.5 Mev deuterons striking the Li target, after exposure to saturation, of 18 divisions per second on the electroscope.[3] Also a new period of 25 hrs., with a saturation intensity of 2 div./sec., was found. This was only weakly activated by the exposures used, and its properties have not been studied. The particles emitted by the 43-min. activity are negative (determined by a Thibaud trochoid apparatus, for the use of which we are indebted to Mr. Ernest Lyman). A chemical separation showed that this (as well as the 25-hr. period) is a mercury isotope. From this information we can place the 43-min. period in the system of isotopes, as being most probably Hg^{203}.

The 43-min. beta-particles are very soft; the absorption curve in aluminum shows a well-defined end point at 0.130 ± 0.005 g/cm², giving an upper limit of 0.42 Mev by Feather's formula. This low energy for such a short life does not fit at all on the usual Sargent curves. There is also a strong gamma-ray, with an absorption in Pb indicating a possible complex structure with energies in the range 70–250 Kev.

GOLD

With slow neutrons, the well-known 2.7-day period[4] is produced with a saturation intensity of 860 div./sec. The absorption curve of the beta-rays has an end point at 0.31 ± 0.01 g/cm² of Al, corresponding to an upper limit of 0.78 Mev. The strong gamma-ray has an absorption in Pb indicating an energy of about 460 Kev. The beta-particles are all negative.

Fast neutrons on Au produce a much more complex activity. There is a 13-hr. period of saturation intensity 1 div./sec., and what appears at first to be a 3.3-day period of saturation intensity 6 div./sec. On following the decay of this, however, the logarithmic plot deviates from a straight line after about 10 days, and seems to be going into a slope of 4–5 days. This shows that the apparent 3.3-day period is a mixture, presumably of the 2.7-day period and a longer one in about equal amounts. We have shown that neither resonance nor thermal neutrons are responsible for producing the 2.7-day period here, since a sample surrounded by a thick sheet of

* Abraham Rosenberg Fellow.

[1] Anderson, Nature **137**, 457 (1936).

[2] Heyn, Nature **139**, 842 (1937).

[3] One division per second corresponds to about 10^4 beta-particles per second emitted from the target. All intensities given are calculated for an exposure to saturation with a 10 μA deuteron beam, although of course different intensities and times were actually used. For the Li neutron exposures, the samples were about 2 cm from the target, surrounded by a layer of B_2O_3 containing about 1/2 g/cm² of B. For the slow neutron exposures, they were about 5 cm from a Be target, enclosed in a large block of paraffin. The samples were all of about the same size (~0.1 g) but in some cases the beta-particles are so soft that even the thin sheets used do not allow them all to escape. No attempt has been made to correct for this.

[4] Fermi, Amaldi, D'Agostino, Rasetti, and Segrè, Proc. Roy. Soc. **146A**, 483 (1934).

gold is activated as strongly as one without this, and the boron layer is sufficient to stop all thermal neutrons.

Chemical separation shows that all these periods belong to gold isotopes, and they are also found to emit only negative electrons. This leads to some difficulty in placing the 13-hr. and 4–5-day periods in the system of isotopes, since only one space (Au^{196}) satisfies the necessary conditions for both of them. Au^{195} would be expected to emit positive electrons, unless a new stable Hg isotope (Hg^{195}) should exist. It seems possible that we have here another case of nuclear isomerism.[5]

Iridium

With slow neutrons, we find the known 19-hr.[4] and 2-month[6] periods, with saturation intensities 120 and 280 div./sec., respectively. We find also a 1.5-min. period with intensity 28 div./sec. This is easily missed if the longer periods are activated strongly, and the observations delayed until several minutes after activation, but by using short exposures it is brought out prominently. It was observed in three different iridium samples, one of which is known to be of very high purity, and we feel that it cannot be due to a contamination. We have done no chemistry on iridium, but by analogy with other slow neutron processes all three of these periods should be attributed to the two isotopes Ir^{192} and Ir^{194}, so that we may have another case of isomerism here.

The 19-hr. beta-rays have an end point in Al of 0.99 ± 0.02 g/cm^2, corresponding to an energy of 2.1 Mev. The 2-month beta-particles are much softer.

With fast neutrons, a very long period (presumably 2 months) is produced with saturation intensity 10 div./sec., as well as a complex array of shorter periods that we have not succeeded in analyzing, using the present data. The 1.5-min. period is present with a saturation intensity of 0.2 div./sec., and the 19-hr. period is buried in the midst of a continuously curving logarithmic plot, so that we cannot be sure of its presence. It is certainly less intense relative to the 2-month period than with slow neutron activation, just as is the 1.5-min. period, so that we can provisionally assign the 2-month period to Ir^{192} and the other two to Ir^{194}. The other unresolved periods again offer difficulties, since all the emitted electrons are negative. No chemical separation has been made.

Platinum

Platinum with slow neutrons gives three periods, 31 min., 18 hrs., and 3.3 days, the saturation intensities being, respectively, 1.2, 0.8, and

[5] Snell, Phys. Rev. **51**, 1011 (1937), and Bothe and Gentner, Naturwiss. **25**, 284 (1937), find such a case in bromine.

[6] Fomin and Houtermans, Physik. Zeits. Sowjetunion **9**, 273 (1936).

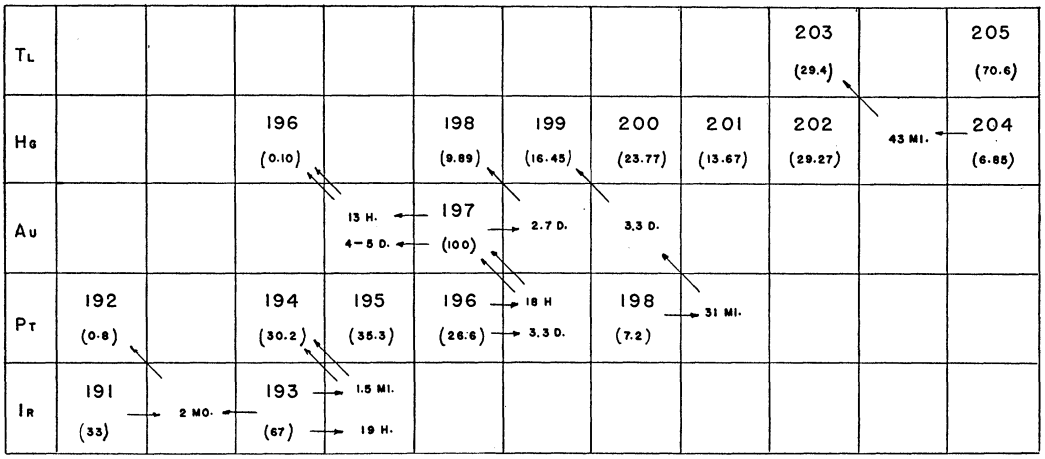

FIG. 1. Isotope chart, showing provisional assignment of identified activities. Figures in parentheses are percentage abundances of stable isotopes. Horizontal arrows indicate neutron capture or ejection, and diagonal arrows indicate β-particle emission.

1.2 div./sec. These might be assigned to the isotopes Pt^{193}, Pt^{197}, and Pt^{199}, but the observation that they all give negative beta-particles makes this unlikely, since Pt^{193} cannot form a known stable isotope by emitting a negative electron. A chemical separation helped to clear up this situation—the gold precipitate showed a 3.3-day activity. Reference to the isotope chart shows that one would expect Pt^{199} to form unstable Au^{199}. We made successive separations of gold from activated platinum to find which platinum period is its parent, and found that it does not come from the 18-hr. period, but most probably does come from the 31-min. period. When two separations $1\frac{1}{4}$ hr. apart were made, the second gold fraction showed about the right ratio of activity to the first to agree with this interpretation. The platinum fraction carries the 18-hr. activity (this incidentally shows that it is not due to Ir contamination), and also, even after repeated separations of gold, still has a period of about 3.3 days. Thus there seem to be two active bodies with the same period (within our present accuracy), one of which fits into the scheme very nicely, while the other (the Pt isotope) again causes trouble. The best that we can think of doing is to make the 18-hr. and 3.3-day platinum periods another pair of isomers. Au^{199} accounts for about 1/3 of the total 3.3-day activity; the agreement between the cross sections for the 31-min. and 3.3-day periods is spurious, since the latter has an extremely soft beta-radiation which can get out of only a fraction of the thickness of the sample.

Pt with fast neutrons gives, like Ir, what appears to be a very complex set of periods, ranging from about 30 min. to more than 5 days. One chemical separation made two days after activation showed that all the long period activity is isotopic with Pt, except for a trace of Au activity, which was much weaker relative to Pt than after slow neutron activation. (We should mention that any active Os would be lost in this and the other separations during the solution in aqua regia, so that we cannot exclude the possibility of its presence. With this exception, all the separations were made to distinguish an element from its two lower neighbors, and in some cases its upper neighbor.) The beta-particles from Pt activated with fast neutrons were found to be nearly all negative, but there was some evidence of a weak long period positron activity.

The results of this work so far do not seem to be capable of any simple explanation without the introduction of a fantastic number of isomeric nuclei, but other explanations may be suggested. Some of the arguments based on the nonexistence of certain stable nuclei may be wrong, or one might assume that some of the active bodies found are excited states of stable nuclei. A search was made using a linear amplifier for α-particles from Au, Ir, and Pt activated with both slow and fast neutrons, and none were found, so no explanation involving α-particle emission can be valid. It also seems strange that so little evidence for the formation of positron emitters is found, although there would appear to be ample chance for their production. We are continuing the work, and expect to publish a fuller account later.

In conclusion, we wish to express our gratitude to Professor Ernest O. Lawrence for his particular interest in this investigation. The financial support of the Research Corporation, the Chemical Foundation, and the Josiah Macy, Jr. Foundation is greatly appreciated.