Artificial Radioactivity of Tantalum

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Radioactivity has been induced in tantalum by slow neutrons, fast neutrons, and fast deuterons. (1) Slow neutron capture leads to Ta^{182} with a lifetime of 97 days. (2) Fast neutron bombardment excites, in addition, an 8.2-hour period with the emission of electrons, K radiation of Ta, and γ-rays. The process responsible for these effects is probably the capture of one neutron with the ejection of two neutrons. The product nucleus, Ta^{180}, goes over to Hf^{180} largely by K electron capture; in this process either γ-rays are emitted or by their internal conversion extranuclear electrons ejected. (3) Deuteron bombardment of tantalum leads to a decay curve affected to such an extent by impurities that the tantalum activity can not be observed clearly.

PROBLEM

THE present paper deals with artificial radioactivity in tantalum bombarded with slow neutrons, fast neutrons, or fast deuterons. Tantalum has only one isotope, $_{73}Ta^{181}$, so that the attribution of radioactive nuclei artificially produced is comparatively simple.

Two periods produced in tantalum are known. A very weak activity with a long period of 200 ± 100 days, produced by slow neutrons from a radon—beryllium source, was observed by Fomin and Houtermans[1] and attributed to Ta^{182}. Heyn[2] reported another very weak activity produced by fast neutrons in Ta. Its half-life was determined by Pool, Cork, and Thornton[3] to 9.1 hours.

In the present investigation tantalum was bombarded with slow neutrons, fast neutrons from lithium, and 5.5 Mev deuterons in the cyclotron of the Radiation Laboratory of the University of California. Sheet tantalum furnished by the Fansteel Metallurgical Corporation was used. Its surface was carefully cleaned by scraping and washing in acids and distilled water. For experiments with deuteron bombardment surface impurities are most troublesome; therefore the tantalum samples were baked out at red heat in high vacuum in an induction oven. A thin platinum foil protected the tantalum in the chamber of the cyclotron from possible impurities knocked out by the deuteron beam from the metal walls. The platinum foil was baked in high vacuum in the same way as the tantalum. The decay curves of the radioactive products were measured with a Lauritsen electroscope.

(1) Slow neutrons

The long period of 200 days ±100 given by Fomin and Houtermans was confirmed. A more accurate value of the half-life is 97 ± 8 days. As there exists only one stable isotope, Ta^{181}, capture of slow neutrons must lead to Ta^{182}. The particles emitted are negative; this was determined by the Thibaud trochoid method. I am indebted to Mr. Ernest M. Lyman for the use of his apparatus. By the observed electron emission Ta^{182} goes over to the stable W^{182}. A very faint period of a few minutes probably belongs to an impurity.

(2) Fast neutrons

Fast neutrons were produced from lithium bombarded with deuterons of 5.5 Mev. The tantalum sample was protected from slow neutrons by a cadmium chamber filled with borax. The same long period as for slow neutrons resulted, due to neutron capture leading to Ta^{182}. In addition, an 8.2 ± 0.2-hour period was observed. (Fig. 1.) A very weak period of approximately 21 minutes may belong to an impurity and may be identical with the short period observed with tantalum bombarded by slow neutrons.

The 8.2-hour period is likewise excited if the lithium target is protected by a copper foil of 1/1000 inch which reduces the energy of the

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[1] V. Fomin and F. G. Houtermans, Physik. Zeits Sowjetunion **9**, 273 (1936).
[2] F. A. Heyn, Nature **139**, 842 (1937).
[3] M. L. Pool, J. M. Cork, and R. L. Thornton, Phys. Rev. **52**, 239 (1937); see Zeits. f. Physik **106**, 238 (1937).

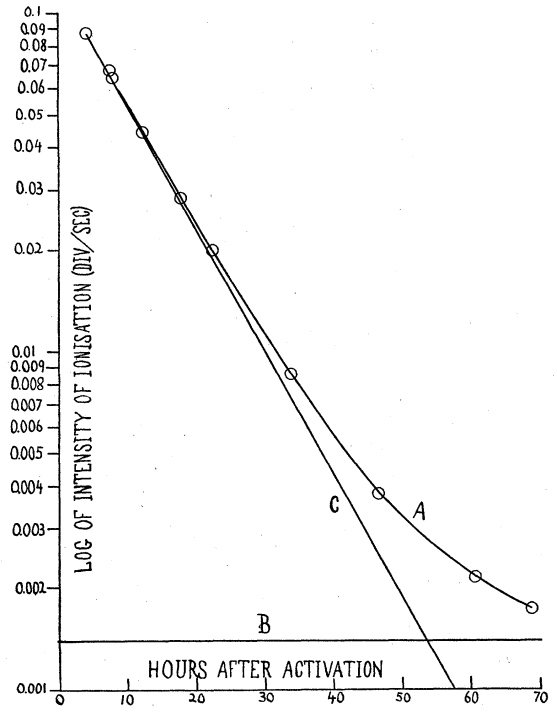

FIG. 1. Decay curves for tantalum bombarded by fast neutrons. Curve *A*, observed; curve *B*, 97-day period applied as correction; curve *C*, corrected curve, 8.2-hour period.

deuteron beam to a small fraction of its original value. This proves that neutrons as they emerge from Li without an appreciable contribution of kinetic energy from the beam are sufficient to excite the nuclear reaction.

The 8.2-hour β-particles have an absorption curve in aluminum with an end point at about 0.54 mm, corresponding to an upper limit of 0.48 Mev according to Feather's formula.

In order to determine at least a relative value for the yield of radioactive Ta excited by fast neutrons Ta samples were bombarded together with Cu samples arbitrarily chosen as a standard. Cu was bombarded for so short a time that the 10-min. period prevailed. By bombarding several sheets of Ta and Cu in touch with each other it was checked that no one sheet appreciably absorbed the neutrons and so reduced the effect on the following sheets. The saturation activity for Ta is 0.08 if the value for Cu is assumed to be one.[4] (This figure does not exactly express the probabilities of the effects of fast neutrons on Ta and Cu nuclei since it is not taken into account that in Cu electrons are able to emerge from greater depths than in Ta, both metals being uniformly affected through their whole bodies by neutrons.)

The 8.2-hour period is undoubtedly identical with the 9.1-hour period reported by Pool, Cork, and Thornton. A correction made for the superimposed long period of 97 days is responsible for the difference.

The 8.2-hour period must be attributed to tantalum itself and not to an impurity for the following reasons. First, its intensity is comparable with the intensity of the 97 day period induced in tantalum at the same bombardment. The ratio of the saturation activities for the short and the long period is approximately 1 : 3. Secondly, the possible impurities fail to show such a period. The Fansteel Metallurgical Corporation kindly informed us that the tantalum sample might contain 0.25 percent columbium and much smaller percentages of Fe, C, Li, Sn, Mn, and Mg. In a separate experiment, a piece of columbium was bombarded with fast neutrons. It failed to show the 8.2-hour period; in fact, columbium is almost completely inactive. None of the other impurities mentioned lead to an 8.2-hour period. It is to be concluded that this period belongs to tantalum itself.

The particles emitted by the 8.2-hour activity are negative; no positive particles could be observed.

The established processes caused by fast neutron bombardment are as follows (see Table I showing the stable isotopes).

(a) *Capture of the neutron.* This process is induced in tantalum by fast neutrons as well as by slow neutrons (Section 1). This is indicated by the 97-day period. Because the Li target gives off neutrons of a certain energy distribution the

TABLE I. *Stable isotopes.*

$_{71}$Cp	$_{72}$Hf	$_{73}$Ta	$_{74}$W
			183
	180	181	182
	179		
	178		
	177		
175	176		

[4] E. McMillan, M. Kamen, and S. Ruben, Phys. Rev. **52**, 375 (1937), footnote 3.

energy of the neutrons actually captured remains uncertain. It is only certain that thermal neutrons are excluded by borax and cadmium.

(b) *Capture of the neutron with ejection of a proton.* This would lead to Hf^{181}, an isotope known to have a lifetime of more than 30 days. Hence, this process is ruled out.

(c) *Capture of the neutron with emission of an α particle.* This process would lead to an isotope of cassiopeium, Cp^{178}, which would go to Hf^{178} with electron emission.

(d) *Capture of the neutron with emission of two neutrons.*[5] This process would lead to Ta^{180}.

Between the alternative Cp^{178} and Ta^{180} the decision was made on the basis of a chemical separation. I am much obliged to Dr. G. T. Seaborg for his detailed advice regarding the chemical method. Tantalum metal and a small amount of rare earth oxalate added to act as a carrier were treated with a mixture of hydrofluoric and nitric acids. This mixture was fumed with 9 normal perchloric acid. The T_2O_5 was filtered out. The rare earth remaining in the filtrate was precipitated by the addition of hydrofluoric acid and filtered out. The result was that the activity was concentrated in the tantalum, the rare earth scarcely showing an activity above background. It is concluded that the active isotope is Ta^{180} produced by the capture of the neutron with emission of two neutrons.

Ta^{180} is expected to go over to the neighboring Hf^{180} by either positron emission or capture of a K electron. This last process has been predicted on the basis of Fermi's theory of β disintegration. The first experimental evidence for this process was recently obtained by Alvarez[6] who observed that a certain radioactive vanadium isotope emits $K\alpha$ radiation as well as positrons. This indicates that this isotope may decrease its nuclear charge as well by K electron capture as positron emission. Theoretically it is known[7] that the ratio of these two alternative processes changes in favor of K electron capture for the heavier elements because of the stronger Coulomb force and the consequent closer approach of K electrons to the heavy nuclei. In agreement with this theoretical

[5] M. L. Pool, J. M. Cork, R. L. Thornton, Phys. Rev. **51**, 890 (1937).
[6] L. W. Alvarez, Phys. Rev. **52**, 134 (1937).
[7] C. Møller, Phys. Rev. **52**, 84 (1937).

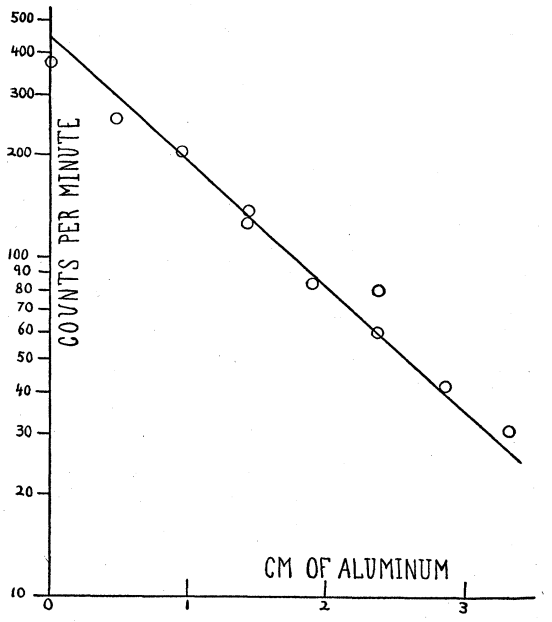

FIG. 2. Absorption curve for tantalum bombarded by fast neutrons. Circles indicate observations, solid line the absorption curve predicted for $K\alpha$ of hafnium.

argument, the survey reported by Pool, Cork, and Thornton[8] shows that positron emission is observed for many light nuclei, for a few nuclei of medium weight, but for no heavy nuclei. Therefore, it is not surprising that in Ta no positron emission is observed. In order to check the $K\alpha$ emission from the active tantalum the method of Alvarez was applied, that is, the absorption curve in aluminum was observed with a Geiger counter. The curve is given in Fig. 2. (Corrections are made for the background of the counter and, in addition, for a background of 40 counts per sec. due to a harder γ radiation.) The solid line represents the absorption curve as one might expect it from the $K\alpha$ line of hafnium, the next lighter element. The agreement seems sufficient to prove the emission of K radiation. It is inferred that K electron capture takes place. This result confirms the attribution of the activity to the Ta^{180} nucleus since K electron capture is not to be expected for the alternative Cp^{178} nucleus which presumably would be an electron emitter. The hard γ-rays are so weak that it is not possible to measure an absorption curve in order to determine their energy.

[8] M. L. Pool, J. M. Cork, and R. L. Thornton, Phys. Rev. **52**, 239 (1937).

The K radiation belongs to the electron emission of the 8.2-hour period, observed with the electroscope, and not to the 97-day period; it disappears completely after a few days. The 8.2-hour period is observed very sharply as a straight line on the semi-logarithmic diagram indicating the presence of only one period (Fig. 1). It follows that electrons and K radiation emerge from the same active nucleus. One may assume a branching disintegration leading to Hf^{180} by K electron capture or instead to W^{180} by electron emission.[9] But this interpretation would lead to the difficulty that W^{180} is not known as a stable isotope so that one would have to assume this isotope either as stable with a very small concentration or as unstable with a long life. (Since one might expect W^{180} to go over to Hf^{178} by α-emission, this emission was checked with ionization chamber and linear amplifier; no such α-emission could be observed. Thanks are due to Mr. Dean B. Cowie for his cooperation and the use of his apparatus.) More plausible than the idea of a branching reaction leading to an unknown W^{180} is the assumption that the electrons are emitted by a secondary process connected with K electron capture. Such a process would be the emission of a γ-ray which may be replaced by internal conversion, that is ejection of one of the extranuclear electrons (Auger process) as it is well known from the β-ray line spectrum. Such a γ-ray emission (or its alternative) is to be expected combined with K electron capture because, in general, the K electron capture must lead to an excited level of the new nucleus, the energy of which is available for radiation. (The conversion of the $K\alpha$ radiation into electron emission by an Auger process would be observed as emission of much softer β-rays, practically not to be separated from the β-rays with the harder limit just described.)

(3) Deuterons

The activity of tantalum, excited by 5.5 Mev deuterons was investigated with all possible accuracy and followed through several months. Mr. J. R. Richardson kindly took a series of cloud chamber photographs. Yet no definite conclusions can be drawn as the decay curve is too complicated. It seems to be a superposition of at least five or six periods. Most of them differ so widely that an analysis of the observed curve may be attempted. Almost all of them are sure to belong to impurities. This follows from a consideration of the established deuteron reactions. The stable isotopes are given in Table I. The reaction, Ta^{181} (d, n) W^{182}, would lead to the *stable* isotope W^{182}. Likewise, the reaction, Ta^{181} (d, α) Hf^{179}, would lead to a *stable* isotope, Hf^{179}. The only *radioactive* isotope to be expected[10] would be due to the Oppenheimer-Philipps reaction Ta^{181} (d, p) Ta^{182}. The resulting isotope Ta^{182} is known to have a lifetime of 97 days.

The overwhelming contribution of impurities observed after deuteron bombardment, in spite of the extreme care with which the sample was cleaned, is certainly due to the closer approach the deuteron has to light nuclei than to the heavy Ta nucleus.

After the gradual decay of some of the superimposed periods the 97-day period is actually apparent. The thick target yield is of the order of magnitude 10^9 deuterons per activated Ta atom provided that the activity observed for this period is not partly due to impurities with equal or longer periods. The masking is so strong that at present no more accurate figure for the Ta^{180} period can be obtained from deuteron bombardment, although with deuterons a much stronger activity can easily be induced than with slow neutrons. The superimposed periods can be attributed to definite impurities as far as it is possible to measure their lifetimes. Very intense is the 10-min. period of N^{13} due to the bombardment of carbon. Its intensity is not even materially reduced by the careful cleaning process described above. Hence the carbon impurity is probably contained in the body of the metal unless one assumes that the residual gas in the cyclotron deposits carbon from decomposed oil vapor on all surfaces. On the other hand, a period of 15 hours, emitting electrons, is materially reduced by the cleaning process; it probably belongs to Na. Several other periods are so much fainter that they cannot be measured with an accuracy sufficient for identification.

[9] See S. N. Van Voorhis, Phys. Rev. **50**, 895 (1936).

[10] In any such argument (Sections 2 and 3) the possibility of isomers is disregarded. So few isomers are known that this is plausible although by no means certain.

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Note added in proof: In the above argument for the disintegration of Ta^{180} no stable W^{180} is assumed to exist. Professor A. J. Dempster kindly informed me of his recent discovery of this isotope (Phys. Rev. **52**, 1074 (1937)). This removes the objection to the negative electron emission from Ta^{180}. This process must be considered as possible in addition to K electron capture, either due to a branching reaction or an isomer of Ta^{180}, both isomers disintegrating with periods of little difference.

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The Oppenheimer-Phillips Process

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The mechanism proposed by Oppenheimer and Phillips for the disintegration of nuclei by deuterons with proton emission (*d-p* reaction) is examined. A formula is derived which expresses the probability of this process in terms of the sticking probability of the neutron (§2) and the penetrability of the potential barrier. The importance of the finite (rather than zero) nuclear radius for the penetrability is pointed out and the penetrability is calculated for various values of the radius (§3). The energy distribution of the emitted protons is found to be given directly by the sticking probability of the neutron (§5). Therefore it may differ considerably from the distribution in "ordinary" nuclear reactions by containing relatively more high energy protons. A measurement of the energy distribution would allow direct conclusions about the width of low nuclear levels which is of importance for the theory of the α-decay and therefore of the nuclear radius (§5). The probability of the O-P reaction is compared with that of ordinary nuclear reactions. The O-P mechanism is found to prevail in the *d-p* reactions for nuclear charges of about 25 and higher; if the reaction leads to a nucleus which emits fast β-rays, the O-P mechanism will be valid at still lower charges. The relative probability of *d-p* as compared to *d-n* reactions is found to be (on the average) unity for very light nuclei, to decrease with increasing atomic number until the O-P process becomes prevalent, and to increase from there on. The excitation function of reactions with nuclei up to $Z\sim30$ is found to be an inadequate test for the O-P mechanism (§6). The question of secondary (cascade) disintegration following the *d-p* reaction is discussed and it is found that such disintegrations (e.g. *d-pn* or *d-p*α) should be rare with deuteron energies below the top of the potential barrier (§7).

§1. GENERAL

IN any ordinary nuclear reaction the first step is the entry of the incident particle as a whole into the initial nucleus. In the "compound nucleus" thus formed, the nuclear particles rearrange themselves until the compound nucleus breaks up into final nucleus and produced particle. However, according to Oppenheimer and Phillips,[1] this general scheme does not apply to reactions of the *d-p* type:[2] Here the incident deuteron does not enter the nucleus as a whole but splits up outside the nucleus into a proton which leaves as the "produced particle," and a neutron which is absorbed by the nucleus.

The Oppenheimer Phillips (O-P) process is, therefore, in principle *simpler* than the ordinary type of nuclear reactions. While the ordinary reactions are double processes, consisting of the formation and disintegration of the compound nucleus, the O-P process is a simple absorption, i.e. formation of the compound nucleus which is in this case identical with the final nucleus. The proton does not enter the reaction at all except by carrying away the surplus energy and momentum, this latter function being, of course, quite essential because otherwise a simple absorption process could never occur. Since the O-P process differs essentially from all other

[1] Oppenheimer and Phillips, Phys. Rev. **48**, 500 (1935).
[2] I.e., reactions produced by deuterons (*d*) with emission of a proton (*p*). For the nomenclature, see Livingston and Bethe, Rev. Mod. Phys. **9**, 245 (1937).