Letters to the Editor

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Atomic Energy from U^{238}

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May 29, 1940*

THE recent experiments of Nier, Booth, Dunning, and Grosse[1] and of Kingdon, Pollock, Booth, and Dunning[2] have confirmed Bohr's prediction[3] that the nuclear fission produced in U by thermal neutrons is to be attributed practically entirely to the rarer isotope of mass 235. This is present in an amount only 1/139 as great as that of the 238 isotope, so it is natural to conclude that only 1/140 of any quantity of U can be considered as a possible source of atomic energy if slow neutrons are to be used. It seems likely, however, that this is wrong, that the energy of a large part of the U^{238} nuclei can also possibly be made to undergo fission by slow neutrons in an indirect way. This is of obvious importance for practical utilization of the atomic energy of U.

The indirect use of U^{238} can come about through the fission either of the U^{239} nuclei formed by the capture of neutrons, or of the nuclei descending from U^{239}. In a forthcoming paper[4] it is shown that $_{93}$EkaRe^{239} and $_{94}$EkaOs^{239}, the probable long-lived descendants from U^{239}, would be expected to undergo fission upon capture of thermal neutrons. A similar argument leads to the conclusion that U^{239} itself would probably also give fission with thermal neutrons. For such fission to be of consequence it is necessary that nearly one U^{239} atom be produced for every U^{235} atom lost. That this condition can be satisfied may be seen as follows. Consider N secondary neutrons. A number Np will suffer resonance capture by U^{238} to give U^{239}, p being the probability for such capture. The remaining $N(1-p)$ neutrons will become thermal. Of these, $N(1-p)w$ will produce fission of U^{235} nuclei and $N(1-p)(1-w)$ will be captured by U^{238} nuclei, w being the probability that a thermal neutron will produce fission. It is assumed that no other capture processes are important for the thermal neutrons, that a non-capturing method of slowing the neutrons needed for developing the divergent chain reaction has been found. The total number of U^{239} nuclei formed is thus $Np+N(1-p)(1-w)$ and the number of U^{235} nuclei lost is $N(1-p)w$. The condition that every U^{235} nucleus be replaced by a U^{239} or descendant nucleus becomes $Np+N(1-p)(1-w)>N(1-p)w$ which reduces to $1/(1-p)>2w$. Since $w<1$ this condition can be met if $p\sim0.5$ as it was in the experiments of Halban, Joliot, Kowarski, and Perrin.[5] Presumably it can be made so with other methods of slowing down the neutrons. Some concentration of the U^{235} isotope may be necessary if p is to be ~0.5. Since the number of neutrons evolved per fission[5] is ~3 the divergent chain can develop if more than one on the average gives fission and the present condition be satisfied if more than one gives U^{239}.

It seems likely, however, that of the three nuclei $_{92}U^{239}$, $_{93}$EkaRe^{239}, and $_{94}$EkaOs^{239} only the last might be expected to have a large cross section for fission. The excess energy in the $_{92}U^{240}$ and $_{93}$EkaRe^{240} nuclei after capture of neutrons by $_{92}U^{239}$ and $_{93}$EkaRe^{239}, respectively, would be small compared to what it is in the $_{92}U^{236}$ nuclei coming from $_{92}U^{235}$. The cross sections for fission would thus be correspondingly small. In $_{94}$EkaOs^{240}, on the other hand, the excess energy would be even larger than in $_{92}U^{236}$ and a large cross section for fission would be expected. It might thus be necessary to allow U from which the $_{92}U^{235}$ had been used up to lie idle for several times the as yet unknown half-life of $_{93}$EkaRe^{239} until a sufficient quantity of $_{94}$EkaOs^{239} had developed in it, after which it could again be used. This could be repeated until all of the $_{92}U^{238}$ had been converted.

* This letter was received for publication on the date indicated but was voluntarily withheld from publication until the end of the war.
[1] Nier, Booth, Dunning, and Grosse, Phys. Rev. 57, 748 (1940).
[2] Kingdon, Pollock, Booth, and Dunning, Phys. Rev. 57, 749 (1940).
[3] N. Bohr and J. A. Wheeler, Phys. Rev. 56, 426 (1940).
[4] L. A. Turner, Phys. Rev. 57, 951 (1940).
[5] Halban, Joliot, Kowarski, and Perrin, J. de phys. et rad. 10, 428 (1939). See also L. A. Turner, Phys. Rev. 57, 334 (1940).

Radioactive Element 94 from Deuterons on Uranium

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January 28, 1941*

WE are writing to report some results obtained in the bombardment of uranium with deuterons in the 60-inch cyclotron.

The uranium was bombarded in the form of U_3O_8 and the deuterons had to pass through a 2-mil thickness of aluminum foil before hitting the uranium target. The carefully purified element 93 fraction contained a beta-activity whose aluminum absorption curve (taken on an ionization chamber connected to an FP-54 tube and also on a Lauritsen electroscope) was distinctly different from the absorption curve of a sample of the 2.3-day 93^{239} (formed from uranium plus neutrons) taken under identical conditions. The upper energy limit of the beta-particles from this new 93 activity is about 1 Mev, compared with about 0.5 Mev for 93^{239}. The ratio of gamma-ray to beta-particle ionization is about five times larger than for 93^{239} The initial part of the absorption curve of this 93 from uranium plus deuterons is very similar to the initial part of the absorption curve of 93^{239}. Of course the production of 93^{239} is expected in the deuteron bombardment of ura-

nium, from the reaction U^{238} $(d, n)93^{239}$. It is impossible to deduce from the absorption curve the relative intensities of the new 93 and of 93^{239}, since the initial parts of the individual absorption curves of these two activities might well be nearly identical. The rate of decay of the high energy beta-particles (0.5–1 Mev) and gamma-rays from the 93 of uranium plus deuterons was determined. This gave a half-life of about 2 days for the new 93. This activity is probably to be assigned to 93^{238}, 93^{236}, or 93^{235} formed in the reaction $U^{238}(d,2n)93^{238}$, $U^{235}(d,n)93^{236}$, or $U^{235}(d,2n)93^{235}$, respectively.

The growth of alpha-particles, which might be due to the element 94 daughter of the 2-day 93, was then looked for. We did observe the growth of alpha-particles in the very carefully purified, as well as in the semi-purified 93 fractions, and the growth curves indicate a half-life of roughly 2 days for the parent of the alpha-emitter. The final alpha-particle count amounts to several hundred counts per minute for a bombardment of 200 microampere-hours. This work was done with a proportional type counter. We plan to re-determine the alpha-particle growth curve more accurately using an ionization chamber and linear amplifier with the help of a magnetic field to bend out the very strong beta-particle background. The alpha-particles have a range of approximately 3.9 cm in air.

This alpha-activity is chemically separable from uranium and 93. The chemical experiments so far indicate a similarity to thorium and the activity has not yet been separated from thorium. More chemical experiments definitely must be performed before it can be regarded as proved that the alpha-particles are due to an isotope of element 94.

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Radioactive Element 94 from Deuterons on Uranium

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WE should like to report a few more results which we have found regarding the element 94 alpha-radioactivity formed in the 16-Mev deuteron bombardment of uranium. We sent a first report[1] of this work in a Letter to the Editor of January 28, 1941. We have in the meantime performed more experiments in order to study the chemical behavior of this alpha-radioactive isotope. The radioactivity can be precipitated, in what is probably the +4 valence state, as a fluoride or iodate by using a rare earth or thorium as carrier material and as a peroxyhydrate by using thorium as carrier material. However, in the presence of the extremely strong oxidizing agent persulfate ion ($S_2O_8^{--}$), plus Ag as a catalyst, this radioactive isotope is oxidized to a higher valence state which does not precipitate as a fluoride. The oxidizing agent bromate ion (BrO_3^-) is not sufficiently powerful to oxidize it to this higher valence state and hence the radioactivity comes down as a fluoride even in the presence of bromate ion. With the help of persulfate ion it has been possible to separate quantitatively this radioactivity from thorium, by using the beta-active UX_1 as an indicator for thorium. These experiments make it extremely probable that this alpha-radioactivity is due to an isotope of element 94. The experiments are being continued.

* This letter was received for publication on the date indicated but was voluntarily withheld from publication until the end of the war.
[1] G. T. Seaborg, E. M. McMillan, J. W. Kennedy and A. C. Wahl, Phys. Rev. 69, 366 (1946).

Search for Spontaneous Fission in 94^{239}

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December 4, 1941*

THIS report describes the experimental procedure and gives the results obtained in our search for spontaneous fission in 94^{239}. The observations were made on a 3.5-microgram sample of 94^{239}, the preparation of which was described in a report of July 24, 1941, from this laboratory. This sample, when placed on one electrode of an ionization chamber connected to a linear amplifier adjusted to record counts due to fissions, gave zero counts in 139 hours. In another, independent experiment, done in a similar manner with entirely different apparatus, this sample gave zero counts in 209 hours. These experiments set a lower limit of the order of 10^{14} years for the "half-life" of 94^{239} with respect to spontaneous fission. These results show that it is probable that the half-life for spontaneous fission of 94^{239} is as long or longer than that of U^{235}, assuming the British results (40 spontaneous fissions per min. per g of pure U^{235}).

The details follow: The 3.5-microgram sample of 94^{239} was mounted on a platinum-coated copper disk, along with its cerium fluoride carrier in which the thickness amounted to $\sim$0.3 mg per cm^2. All the measurements described herein were made with shallow ionization chambers so constructed that this disk could be inserted to serve as one of the electrodes. Consideration of this geometry indicates that ionizing particles emitted from a sample on the disk could be counted with an efficiency of about 45 percent; tests of the alpha-particle counting rate with a thin, weighed uranium sample on a similar disk confirmed this estimate when the gain of the linear amplifier used was set sufficiently high. Since in each fission process two ionizing particles are emitted in opposite directions, an upper limit of about 90 percent efficiency for fission counting is set by the geometrical arrangement. Actually, because of the fluctuating background ionization due to alpha-particles from the sample, it was expedient to adjust the amplifier gain in such a way that a slightly lower counting efficiency was obtained. With the use of a neutron source and a thin uranium sample mounted like the 94^{239} sample, two independent calibrations of this efficiency were made, as follows: (1) oscillographic observation of pulses showed the fraction of fission pulses too small to actuate the thryatron recording circuit; (2) the recorded fission rate was compared with the expected rate calculated from the weight of the thin uranium sample, the slow neutron fission cross