ON THE PRODUCTS OF THE FISSION OF URANIUM AND THORIUM UNDER NEUTRON BOMBARDMENT

BY

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The first experiment of FERMI (1) on the transmutation of uranium and thorium by neutron bombardment already disclosed several interesting new features, which were investigated more closely in the following years especially by HAHN and MEITNER, and HAHN, MEITNER and STRASSMANN. In the case of uranium, ten different radioactive bodies were found to result from the transmutation (2). In the case of thorium, six transformation products with chemical properties similar to those of radium and actinium were found, apart from a radioactive thorium isotope which is probably converted into a protactinium isotope (3). It was found in both cases that the transformation products could not be arranged to form one single radioactive chain, and isomerism had apparently to be assumed in order to explain the large number of bodies.

At that time, all previous experience with nuclear transmutations suggested as possible starting processes only radiative capture of the neutron or capture with subsequent emission of an α-particle (or possibly of a proton). Neutron capture in uranium, followed by emission of a β-particle by the uranium isotope created, must lead to elements beyond uranium. It was therefore necessary to look for a chemical procedure which separates the elements beyond uranium from uranium itself and its lower neighbours. Precipitation with hydrogen sulfide in 2-normal hydrochloric acid was

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found suitable as the elements $Z = 85$ to 92 remain in the filtrate while the higher elements should be precipitated, because of their similarity to rhenium and the platinum elements, respectively. Seven different active bodies with the periods of 2.2 min., 16 min., 59 min., 2.5 h., 5.7 h., 66 h., and 66 days were found in the precipitate and were consequently assigned to transuranium elements. Three further periods (23 min., 40 sec., 10 sec.) were ascribed to uranium, although conclusive chemical identification was possible only in the case of the 23 min. period.

Following the remarkable observation by Curie and Savitch (4) of the occurrence of transmutation products of uranium chemically similar to rare earth elements, Hahn and Strassmann found, a few months ago, some further uranium products with unsuspected properties. These products were at first believed to be isotopes of radium and actinium (5); by careful chemical tests, however, it was found later that they were actually isotopes of barium and lanthanum (6). Thus it was necessary to assume the existence of an entirely new type of nuclear reaction, the splitting of the uranium nucleus into much smaller nuclei. Hahn and Strassmann found also that the activities produced in thorium, which had formerly been ascribed to isotopes of radium and actinium, were actually due to isotopes of barium and lanthanum.

As was pointed out by the present writers (7), this new type of nuclear reaction can be simply understood on the basis of the general ideas on the mechanism of nuclear transmutations developed in later years by Bohr (8). According to these ideas, a nucleus behaves in many respects similarly to a liquid drop, and the new type of transmutation, for which we proposed the name "fission",

may therefore be compared with the division of such a drop into two smaller droplets as a result of a deformation produced by an external disturbance. For highly charged nuclei, such a deformation will in fact require a relatively small energy, since the restoring influence of the surface tension arising from the short range nuclear forces will be largely compensated by the electrostatic repulsion of the separating fragments of the nucleus. In the course of the separation process, this repulsion will moreover give the fragments very large kinetic energies, of the order of 100 MeV, corresponding approximately to the large difference in the mass defects of the original nucleus and the final products. Just on account of this large energy release, direct experimental evidence of the fission processes was also soon obtained partly by the observation of the large ionisation produced by each of the high speed nuclear fragments (9), partly by the possibility of separating the new products from the bombarded uranium and thorium, afforded by the power of the high speed fragments to penetrate through thin foils of solid matter (10).

In our first note on this subject (7), we discussed the possibility that all the so-called "transuranium elements", precipitated by hydrogen sulfide, might originate in the fission of the uranium nucleus and consequently be isotopes of some light elements. Only the isotope of 23 min. period, which had been chemically identified with uranium, was assumed by us to result from radiative capture of the neutron. By our subsequent experiments, which have already been briefly reported and of which a detailed account is here given, this view has also been fully confirmed and in the meantime other investigators have independently arrived at the same conclusion (11).

Our experiments were based on the possibility of collecting the high speed fission fragments, which emerge from a considerable depth of the uranium layer bombarded, on a suitable receiving surface, just as nuclei of naturally active bodies projected by a-recoil can be collected as an active deposit. If then the "transuranium" activities are found among the collected nuclei, this is a proof that they originate by fission of the uranium nucleus and are, therefore, isotopic with lighter elements; the momentum imparted to a nucleus as a result of radiative neutron capture would in fact be very small and would enable only an exceedingly small fraction of these nuclei to escape from the uranium layer. Consequently the experiment consisted in collecting on a receiving surface the fission products of uranium under neutron irradiation and then applying the same chemical treatment to them which had previously been used for the separation of the "transuranium elements".

A "thick" layer (14 mg/cm²) of uranium hydroxide, 4×4 cm in area, was fixed on the underside of a glass plate. As a receiver, 1 mm below the uranium layer, we used the surface of about 10 cm³ water contained in a shallow trough made of paraffin wax. Both paraffin and water remain inactive under neutron bombardment, and no undesirable elements are present under the chemical operation. In each experiment a small fraction of the water was evaporated. As a test that the water had not been contaminated with uranium, it was always checked that the residue did not exhibit any persistent activity, corresponding to the natural radioactivity of uranium.

Neutrons were obtained on bombarding lithium with deuterons accelerated to about 800 KV. After a few preliminary experiments, several irradiations of one hour's dura-

tion were made. After each irradiation the water was sub-
jected to the treatment designed to separate the "trans-
uranium elements", with slight modifications in order to
be sure that the recently discovered activities (5, 6) would
stay in solution, in spite of the absence of uranium. The
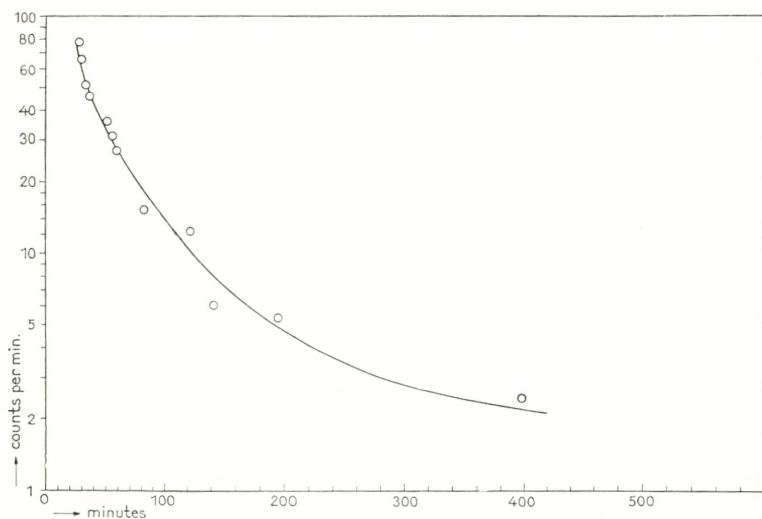procedure consisted in adding hydrochloric acid until the

Fig. 1.

solution was 2-normal, 150 mgm barium chloride, 10 mgm
lanthanum chloride, and 5—10 mgm platinum chloride in
order to get a precipitate of platinum sulfide acting as a
carrier for the elements under investigation; after treat-
ment with hydrogen sulfide, the precipitate was carefully
washed and dried. The activity of the filter was measured
with a GEIGER-MÜLLER counter with 0.1 mm aluminium
walls, as in the experiments of HAHN, MEITNER and STRASS-
MANN (2) and under the same geometrical conditions.

There was agreement between all the experiments and a
typical example of the decay of a precipitate is shown by the

circles in fig. 1. A check experiment, in which a uranium
sample after one hour's irradiation was subjected to the same
chemical treatment in bulk, is represented by the full line,
the intensity scale being suitably corrected. The circles are
seen to lie on the curve within the statistical errors, indicating
the presence, in the recoil sample, of the periods of 16 min.
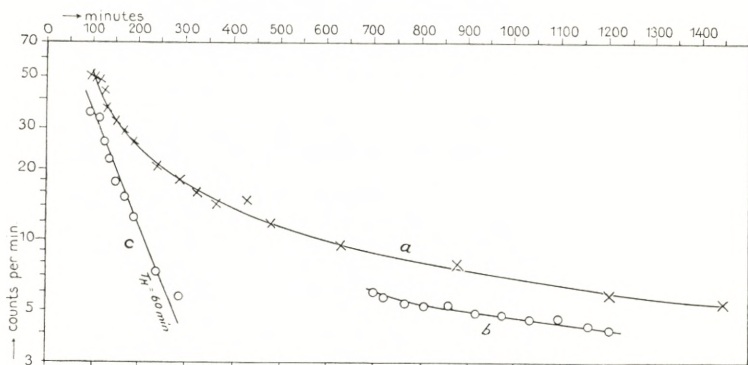and 59 min., and traces of the longer period substances.

Fig. 2.

In order to get better evidence for the long-lived bodies
we performed a longer irradiation, using three uranium
layers and three water receivers on top of one another.
More than three layers would not have increased the yield
much further, because of the increasing distance from
the target. The irradiation of $2^1/_2$ hours' total effective dur-
ation had to be interrupted twice on account of trouble
with the high-tension plant. One hour after the second
interruption it became clear that the difficulties could not be
removed within a short time and, consequently, we started
the chemical separation. These circumstances obviously
handicapped the shorter periods (15 min. and 59 min.)
while the relative intensity of the long period (5.7 hours and

66 hours) products was only little influenced by the inter-
ruptions. Fig. 2 shows the decay of the sulfide precipitate
(curve a) and, for comparison, an older curve by HAHN,
MEITNER and STRASSMANN, where counting was started
eleven hours after the end of a two hours' irradiation
(curve b); by that time only the two
long periods are left. It is seen that
the curves a and b are parallel, which
shows the presence of the bodies of
5.7 and 66 hours (and, of course,
the one of 2.5 hours which is formed
from the 66 hour body). If the activ-
ities of these long-lived bodies are
extrapolated back and deducted from
the measured activities at the begin-
ning of curve a, we obtain curve c,
which shows a decay of about 60 min.,
demonstrating the presence of the 59
min. body.

Several attempts were made to
detect the 2.2 min. period in the sulfide
precipitate. After an irradiation of 4
to 5 minutes, the chemical separation

Fig. 3.

was carried out as rapidly as possible. We found it possible
to start counting 7.5 min. after the end of the irradiation,
two minutes being lost with the transport of the water to the
chemical room and of the filter to the counter room. Fig. 3
shows curves from two such experiments. The first points
indicate a faster initial decay with a period of about two
minutes, but the evidence is not very clear cut. The rest of the
curve decays with a nearly unique period of about 17 min.,
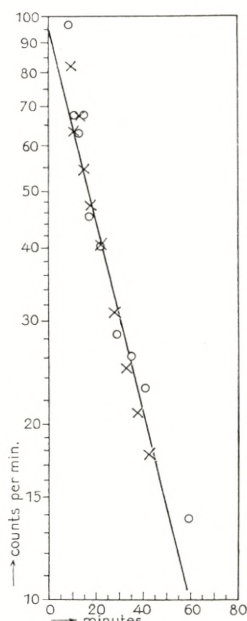demonstrating once more the presence of the 16 min. body.

In connection with experiments by Bjerge, Brostrøm and Koch (12), it has been shown by one of us (13) that the decay curve of a mixture of many components with different decay times, such as results from the fission of uranium or thorium, has very nearly a "standard shape" which can be calculated from purely statistical assumptions
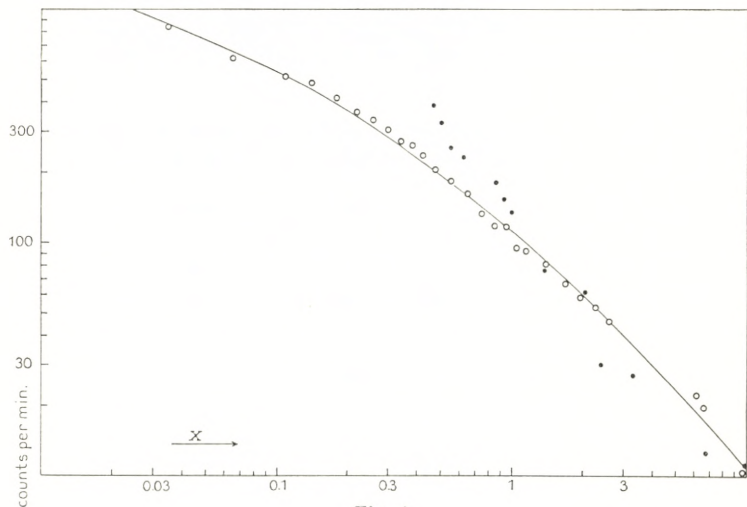

Fig. 4.

on the distribution of periods. For the intensity at a time t reckoned from the end of an irradiation of duration T the calculation gives the expression

$$J_T(t) = \text{const.} \left[x^{-\frac{1}{n}} - (1+x)^{-\frac{1}{n}} \right] \quad x = \frac{t}{T}$$

where n is the exponent in Sargent's rule, $\lambda = \text{const.} E^n$ connecting the decay constant λ and the upper limit E of the β-ray spectrum. In fig. 4 this expression is plotted (as full line) on a double-logarithmic scale. The circles represent the results of an experiment (12) in which a uranium

layer was irradiated for 100 minutes and the activity of
the recoil was measured without previous chemical treat-
ment. A very similar decay was found with a thorium
layer and it was pointed out that this similarity need not
imply any similarity in the distribution of individual
periods, but can be explained entirely on the basis of the
statistical argument. The dots, representing our own mea-
surements with H_2S-precipitates, do not, however, fit the
standard curve. Therefore the similarity of our curves
with the "transuranium curves" obtained by HAHN, MEIT-
NER and STRASSMANN cannot be explained on the basis of
the statistical argument but must be taken as evidence for
the identity of the corresponding products.

Similar experiments with thorium have been reported
by one of us (14). It was shown that the fission of thorium
gave rise to active elements chemically analogous to the
"transuranium elements", but that different periods occur.
Since these periods are not mentioned in the account of the
investigation of the uranium and thorium fission products,
which have been published in the meantime by BRETSCHER
and COOK (15), it may be worth while also to describe
these experiments somewhat more closely and to discuss
the decay curves in detail.

A "thick" layer of thorium oxide on a glass plate was
irradiated with neutrons $(Li + D)$ and the fission products
were collected on a water surface, the chemical treatment
of the water being the same as in the uranium experiment.
The thorium layer was covered with a celluloid foil of
about 0.3 mm air equivalent in order to avoid contamin-
ation of the water through α-recoil or traces of emanating
thoron. After an irradiation of about $2^1/_2$ hours, the decay
curves of fig. 5 were obtained. Curve a shows the decay
of the sulfide precipitate, and curve b shows the decay of

the carbonate precipitate obtained after subsequent neutral-
isation of the filtrate. The sulfide precipitate, after an initial
drop with a period of about 40 min., is seen to decay with
a period of 14.5 hours. For infinite irradiation, the longer
period would be about ten times stronger than the shorter
one. This might indicate that the 14.5 hour substance
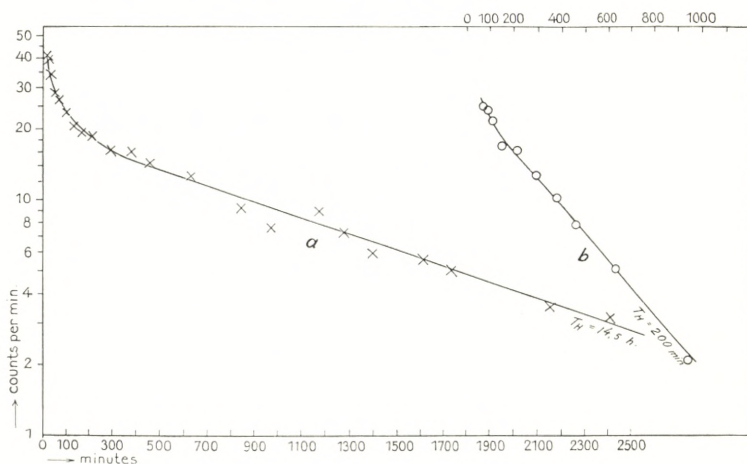produces a short-lived substance with correspondingly

Fig. 5.

penetrating β-rays; we have not, however, tested this hypo-
thesis. Curve b shows decay periods of about 20 min. (of
which only little was left when counting was started one
hour after the end of the irradiation), and 3.3 hours, in
good agreement with the periods found earlier by Meitner,
Hahn and Strassmann (3). After 21 hours no measurable
activity was left; this shows that no Th-B had got into the
water, since our chemical procedure would have collected
most of the Th-B with the carbonate precipitate.

The observation that, in spite of the general similarity
of the fission products of uranium and thorium, certain

periods are only found in uranium and certain other periods only in thorium, might be interpreted as an indication of a selectivity regarding the various modes of division of the nuclei undergoing fission. Such a selectivity would also seem to be disclosed in uranium by the distribution curve obtained by Booth, Dunning and Slack, for the kinetic energies of the fission fragments. This curve shows in fact two pronounced maxima corresponding to masses of the fragments just in the regions of atomic weights of about 100 and 136, where the most abundant uranium fission products revealed by chemical analysis are found. As well the striking similarity in the occurrence of the various fission products in uranium and thorium as the marked differences revealed in our present experiments might therefore be explained if the distribution curves of the fragments were in the two cases of similar type but showed minor deviations from each other. In particular, we might expect that, corresponding to the smaller atomic weight of thorium, the maxima would be somewhat displaced towards smaller atomic weights, giving rise to a greater abundance in thorium than in uranium of some of the fission products in the region immediately below the uranium maximum at highest atomic weight.

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