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LIBERATION OF NEUTRONS IN THE NUCLEAR EXPLOSION OF URANIUM IRRADIATED BY THERMAL NEUTRONS

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The discovery was first announced by Hahn and Strassmann[1], that uranium under neutron irradiation is split by absorbing the neutrons into two lighter elements of roughly equal weight and charge, being accompanied by a very large amount of energy release. This leads to the consideration that these fission fragments would contain *considerable excess* of neutrons as compared with the corresponding heaviest stable isotopes with the same nuclear charges, assuming a division into two parts only. A part of these excess neutrons was found, in fact, to be disposed of by the subsequent β-ray transformations of the fission products,[2] but another possibility of reducing the neutron excess seems to be a *direct emission* of the neutrons, which would either be emitted as a part of explosion products almost instantaneously at the moment of the nuclear splitting or escape from highly excited nuclei of the residual fragments. It may, therefore, be expected that the explosion process would produce even larger number of secondary neutrons than one per nuclear fission.

Soon after the discovery of this new type of reaction, it has been attempted by several workers[3–8] to observe this pronounced emission of neutrons from uranium irradiated by neutrons. It is of interest to find more closely whether and to what extent the emission of neutrons takes place in uranium by absorbing *thermal* neutrons, and also to estimate the number of the neutrons released per fission in connection with the possibility of a cumulative process of exo-energetic transmutation chains. In some of the previous experiments[3,4,5,6] a comparison

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1) O. Hahn and F. Strassmann, *Naturwiss.*, **27**, 11 (1939).

2) Numerous *active* products, for example, with atomic numbers lying between 35 (*Br*) and 57 (*La*) were revealed by chemical work.

I. Curie and P. Savitch, *Compt. rend.*, **208**, 343 (1939); O. Hahn and F. Strassmann, *Naturwiss.*, **27**, 89 *and* 163 (1939); P. A. Heyn, A. H. W. Aten jun. and C. J. Bakker, *Nature* **143**, 516 (1939); P. Abelson, *Phys. Rev.*, **56**, 1 (1939) and *numerous subsequent reports.*

3) H. von Halban, F. Joliot and L. Kowarski, *Nature*, **143**, 470 *and* 680 (1939).

4) H. L. Anderson, E. Fermi and H. B. Hanstein, *Phys. Rev.*, **55**, 797 (1939).

5) H. L. Anderson, E. Fermi and L. Szilard, *Phys. Rev.*, **56**, 284 (1939).

6) L. Szilard and W. H. Zinn, *Phys. Rev.*, **55**, 799 (1939).

7) G. von Droste and Reddemann, *Naturwiss.*, **27**, 371 (1939).

8) J. L. Michiels, G. Parry and G. P. Thomson, *Nature*, **143** 760 (1939).

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was drawn between the numbers of thermal neutrons present in a large water tank with and without uranium in the water under the irradiation of photo-neutrons from a Ra γ-Be source at the centre of the tank. Since in those experiments (made in such a system as composed of a mixture of uranium and water), however, parts of the neutrons are considered to be absorbed into uranium and to induce the fission before reaching down to thermal energies, it seems desirable to make measurements on the *separated* effects due to thermal neutrons alone. The present experiment is, therefore, intended to find some information regarding a net increase in the number of presumably fast neutrons released from uranium by absorbing the primary thermal neutrons alone. In the following, a short report of the experiments is given, the detailed account of which is to appear soon in the *Memoirs of the College of Science, (A), Kyoto Imperial University.*

In order to ascertain a net gain in the number of secondary neutrons from uranium irradiated by thermal neutrons, a series of experiments were carried out.

To obtain thermal neutrons, a cylindrical paraffin block, 20 cm in diameter and 25 cm in height, was made, in the central cavity of which was placed the usual neutron source of 50 mg Ra α-Be. The container of uranium oxide powder was a thin concentric double walled, cylindrical copper vessel, 25 cm in the outer diameter, 21 cm in the inner diameter and 25 cm in height, and the cylindrically zonal space between the double walls was able to contain the powder of U_3O_8. The paraffin cylindrical block (with neutron source) was placed inside the inner wall of the vessel.

The outer surface of the vessel was completely covered on all sides by the cadmium sheet shields 1 mm thick, which prevented thermal neutrons from escaping to affect the detector. An *inner* cadmium sheet cylinder (about 20.5 cm in diameter) was also placed, if desired, so as to cover the whole cylindrical surface of the paraffin block on its outside, and it served as a shield to prevent the slowed down source neutrons from interacting on uranium. Accordingly, the uranium oxide could be either screened from the thermal neutrons or exposed to them by simply setting this inner shield in or out of the place.

As a detector for slow neutrons, a *boron* cylindrical ionization chamber, 2.2 cm in diameter and 20 cm in length, lined with a thin layer of boron, was used. The ionization pulses from the chamber were fed to a grid of a linear amplifier of Wynn-Williams-type,[9] through which the frequency of these pulses was counted from the records of kicks on the moving sensitized photographic papers by use of

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a usual oscillograph. In order to reduce the background due to γ-rays from the source, a thick lead block was interposed between the uranium container and the top of the horizontally placed ionization chamber. The whole apparatus was adequately enclosed in a thick walled (about 15 cm) paraffin chamber. (the inner content: $30 \times 70 \times 26$ cm)

The present method of neutron detection is eventually based on measuring the number of the neutrons in question as that of only the neutrons which have been slowed down on the whole inner walls of the enclosed paraffin chamber after passing through the outer cadmium shields with relatively high velocities. So a simple comparison of the counts of neutrons per definite time interval measured with and without the inner cadmium shield, the outer cadmium shield being in place, would allow us to detect and measure the net increase in the number of relatively fast neutrons inside the outer cadmium cylinder.

A number of sets of such *alternating* measurements (in the presence and absence of the inner cadmium shield) were made per 10 minutes interval on the oscillograph records, when one of the semi-circular zonal parts between the concentric double walled cylinders of the vessel was filled with 2.5 kg of U_3O_8. The semi-circular uranium part was placed in one case (i) closely to the ionization chamber, and in another case (ii) on the side opposite to the ionization chamber with respect to the neutron source. In the former case, the front of the outer convex semi-circular wall of the uranium part was kept at a distance of 64 cm from the top of the cylindrical ionization chamber along its axial direction, and in the latter case, the uranium part was, at its remotest part, farther away by 25 cm (the outer diameter of the uranium vessel) from the ionization chamber along the axial line.

In this way, we found i) 1.8 percent increase of secondary neutrons upon the primary in the first case, and ii) 3.8 percent increase in the second case. These increases must be taken to be caused only by the secondary emission of *nonthermal* neutrons due to the absorption of *thermal* neutrons alone. It seems also reasonable to consider that a remarkable difference of these increases in the number of counts observed between the first and the second cases may be associated with probably *high* energies of the secondary neutrons. The secondary neutrons emitted in the process concerned are considered in the second case to be more strongly slowed down by passing through the medium of the paraffin

9) B. Arakatsu, K. Kimura and Y. Uemura, *Memoirs of the Faculty of Science and Agriculture, Taihoku Imperial University*, 18, 83 (1936).

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cylindrical block 20 cm in diameter.

We also looked for *delayed* fast neutrons[10] in the *present arrangement*, but observed no indication which might be due to the delayed emission after the quick removal of the source from the paraffin block.

In order to estimate the energies of the secondary fast neutrons, a set of paraffin sheets (37 mm thick each) was interposed between uranium oxide and the ionization chamber, and the alternating change of the counts—with and without the inner cadmium shield—was observed for every different thickness of the increased paraffin layers.

From the result of the above experiments made in the same geometrical arrangement as in the case (i), it is shown that the number of secondary neutrons first increases with the increased paraffin sheets and then after passing through a maximum it decreases. The result obtained in the same geometry as in the case (ii) shows that the number of secondary neutrons is reduced to about one half through a paraffin sheet (37 mm thick), while the removal of the paraffin sheet exerts no appreciable influence on the predominant increase of the original count. In view of the results concerned we have some reason to conclude that the secondary fast neutrons emitted from uranium, when it is exposed to thermal neutrons, possess a kinetic energy of *about 10 Mev.* in order of magnitude.

By measuring the total absorption of thermal neutrons by uranium oxide, it was found that 7 percent of the primary thermal neutrons emitted by the source is absorbed through uranium oxide, and its total absorption cross section due to fission and other possible processes in the present experimental arrangement was calculated to be 9.6×10^{-24} cm^2 by use of the relation of the exponential absorption. This result means that the number of the secondary fast neutrons produced does not exceed the total number of thermal neutrons absorbed within uranium oxide by all possible processes, and that a nuclear chain reaction is not maintained in the ordinary uranium oxide exposed to thermal neutrons alone.

In order to estimate the *average number* ν of the secondary fast neutrons liberated per fission of uranium for thermal neutron absorbed, let σ_f be the cross section of thermal neutrons for this fission process alone, n the number of the nuclei per unit volume and r the path length of neutrons travelling through

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10) R. B. Robert, R. C. Meyer and P. Wang, *Phys. Rev.*, 55, 510 (1939); R. B. Robert, L. R. Hafstad, R. C. Meyer and P. Wang, *Phys. Rev.*, 55, 664 (1939); E. T. Booth, J. R. Dunning and F. G. Slack, *Phys. Rev.*, 55, 876 (1939).

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uranium oxide. It follows, then, that $\nu\sigma_f nr$ should correspond to the ratio of the number, C, of the observed *net gain* of secondary neutrons from uranium oxide to that, A, of thermal neutrons emitted by the source, namely to $\left(\frac{k'}{k}\right)\cdot\frac{C}{A}$, where k and k' are the average efficiency factors of the detector for slow neutrons and for fast neutrons resulting from fission respectively. Thus we have

$$\nu=\left(\frac{k'}{k}\right)\frac{1}{\sigma_f nr}\cdot\frac{C}{A}.$$

It is here assumed that the contributions of all fast and slow neutrons of different energies to the detecting efficiency of the neutron detector in the paraffin chamber are of the same order of magnitude. Since it is considered to be possible in the geometry of the case (ii) to obtain the similar condition to that as required above, provided that it would still give some lower value of ν, $\frac{k'}{k}$ is assumed to be equal to unity in good approximation. In calculating ν, we use the value of σ_f ($=2\times10^{-24}$ cm^2) for thermal neutrons, measured by the direct observation of Anderson, Booth, Dunning, Fermi, Glasoe and Slack.[11] Putting the numerical values in the above relation (and also by taking into account the effect of the reduction in the number of neutrons due to the absorption within uranium oxide), we find

$$\nu=2.6.$$

Thus, by the present investigation it was made possible to decide conclusively the remarkable *net increase* of secondary fast neutrons caused only by the thermal neutrons absorbed, and also to estimate the *mean number of* ν as deduced above. From the considerations of the order of the amount of energy involved in the fission neutrons, it may be more probable that the neutron release takes place preferably at the moment following within a *very* short interval from the main division of the compound nucleus into the heavy fragments of highly exited states.

It is reasonably seen that remarkable fractions of the neutron excess and of the fission energy are both certainly disposed of by the direct emission of neutrons, and that further fractions are also carried off by the subsequent β-ray emission of the explosion products.

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11) H. L. Anderson, E.T. Booth, J. R. Dunning, E. Fermi, G. N. Glasoe and F. G. Slack, *Phys. Rev.*, **55**, 511 (1939).

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