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ON THE SPONTANEOUS EMISSION OF NEUTRONS FROM THE URANIUM NUCLEUS

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ABSTRACT. Using a boron-lined ionisation chamber filled with BF_3, the spontaneous emission of neutrons from the uranium nucleus has been detected. It is suggested that they are produced as a result of the spontaneous fission of uranium and that 3 neutrons are emitted per each such process.

INTRODUCTORY

Soon after the discovery of the induced fission of the uranium nucleus due to neutron bombardment, it was realised that in whatsoever way the ${}_{92}U^{239}$ nucleus divided itself, there must be an excess of neutrons over that contained in normal nuclei of the same atomic number. There seemed to be two possibilities of getting rid of this neutron excess :

(1) by the emission of a β-ray, a neutron may be transformed into a proton, thus reducing the neutron excess by two units.

(2) by the direct liberation of the neutrons taking place either as a part of the fission itself, or as an "evaporation" process from the resulting nuclei which would be formed in an excited state.

The evidence for the first process was obtained from the successive β-transformation of many of the fission products. The direct emission of neutrons during the second process, was sought for and obtained by several experimenters, employing various methods.

Von Halban, Joliot and Kowarski (1939), measured the distribution of neutrons surrounding a neutron-source placed at the centre of a large container filled with a solution of uranyl nitrate. Another set of measurements was made in which a solution of ammonium nitrate of the same strength was used instead of uranyl nitrate. They found a 5% higher intensity in the former case and associated it with the production of secondary neutrons due to the induced fission process in uranium. The average number of neutrons produced per fission was calculated to be 3.5 ± 0.7. Dodé et al (1939) also demonstrated that the neutrons produced during induced fission were fast neutrons.

Further experimental evidence of neutron emission during induced fission was obtained by Anderson, Fermi and Hanstein (1939), and Haenny and Rosenberg (1939).

The experimental investigations on the spontaneous fission of the uranium nucleus, carried out by the author, (Chatterjee, 1944) raised the interesting question whether the process of spontaneous fission of U is likewise accompanied

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by the emission of neutrons. The following experiment was therefore undertaken to verify the correctness of the above hypothesis and also to determine the half-life period of the corresponding process. A preliminary report of our findings was published in a note in Science and Culture (1944).

PRINCIPLE OF THE METHOD OF MEASUREMENT

A proportional counter, lined with a thin layer of boron powder and filled with BF_3 gas is used. Owing to the comparatively high gas-pressure (~ 1 atm), the counter is used as an ionisation chamber, in conjunction with a linear amplifier and a thyratron-operated mechanical recorder. The rate of counting of such an arrangement may be represented by

$$A + B + C,$$

where A represents the number of slow neutron-induced boron disintegrations, the resulting α-particles being counted.

B is the number of recoil nuclei produced by fast neutrons traversing the counter.

C is the number of other particles of high specific ionization passing through the counter.

It is evident that

$$A = VN_B \rho \sigma_B v_B + (N_A \rho / \mu) R_B \sigma_B .$$

$$B = VN \int_{v_2}^{v_1} i(v) \sigma(v) \, dv .$$

$$C = S \int_{v_4}^{v_3} I(v) \, dv$$

where V = volume of the counter.

N = the total number of nuclei per c.c. in the counter.

N_A = Avogrado's number.

N_B = the number of B^{10} nuclei per c.c.

ρ = the density of neutrons per c.c. with energies in the $1/v$ region.

σ_B = the capture cross-section for B^{10} for neutrons of velocity v_B.

R_B = the range of α-particles in boron.

μ = atomic weight of boron (B^{10}).

$i(v)dv$ = the current of fast neutrons of velocity between v and $v + dv$.

$\sigma(v)$ = the recoil cross-section of the nuclei of the gas for neutrons of velocity v.

S = the cross-sectional area of the counter.

$I(v)dv$ = the current per cm^2, of particles of high specific ionization, of velocity between v and $v + dv$.

The limits of integration, v_1 is the highest and v_2 the lowest neutron velocity producing measurable recoils, v_3 the the highest and v_4 the lowest velocity of a particle traversing the counter and producing enough ions to be recorded as a count.

It is evident that the counter is essentially a slow neutron detector, because $\sigma_B >> \sigma(v)$. It can also be seen that the factor C is due practically entirely to the radio-active contamination of the inner surface of the counter and that of the boron-layer; since the number of penetrating heavily ionizing particles in the cosmic rays at the sea-level is extremely low. Likewise, the contribution of factor B (due to cosmic ray fast neutrons at sea-level) is negligible. In fact, the back-ground of such a counter is practically solely determined by

(*i*) the radio-active contamination of the internal surface layer of the counter and (*ii*) the presence of slow neutrons in the atmosphere.

Owing to the high-value of the half-life period for the spontaneous fission of the uranium nucleus (as obtained in a separate experiment and described in details in the *Trans. Bose Res. Inst.*, (1945) the expected number of neutrons emitted by one gram of uranium per minute is extremely small in comparison with the back-ground count. The sensitivity of our arrangement has, therefore, been sought to be enhanced by

(1) employing a large quantity of U_3O_8 (corresponding to 1.2 kg. of metallic uranium),

(2) surrounding the U_3O_8-container with a thick paraffin cylinder in order to slow down the neutrons, and

(3) increasing the efficiency of the slow-neutron counter by using an optimum thickness of the boron layer on the inner surface and filling it with BF_3-gas at a pressure of 50 cms. of Hg.

EXPERIMENTAL ARRANGEMENT

The experimental arrangement is shown diagrammatically in Fig. 1. The neutron counter consisted of a brass-tube 5 cms. in diameter and 26 cms. in length. The two open ends were closed with ebonite discs fitted with amber plugs and earth guard rings. In order to prevent the occurrence of any paralysis of the subthreshold counter (Montgomery and Montgomery, 1940), due to an electron back-ground of high intensity, the field-strength in the neighbourhood of the central wire was reduced by replacing it with a thick brass-rod of about

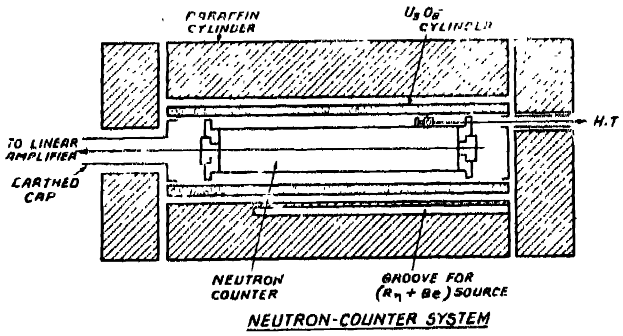

FIG. 1

2 mms. diameter. The inner surface of the outer brass-tube was lined with a thin layer of amorphous boron powder ($\sim$0.1 mm. thick) so that the range of the α-particles produced by the $_5B^{10}$ (n, α) $_3Li^7$ reaction was somewhat greater than the stopping-power of the layer. The counter tube was filled with BF_3 gas at a pressure of 50 cms. of Hg. The surface of the ebonite end-pieces within the counter was protected from the action of BF_3 gas by a thin layer of paraffin, while only piecein was used for making the counter gas-tight. A potential of +1200 volts was given to the body of the counter, while the inner electrode was connected directly to the grid of the first tube of the proportional amplifier. The counter was operated in the ionisation chamber region. The pulses were recorded by a thyratron operated mechanical counter.

The U_3O_8-container consisted of a hollow double-cylinder made of galvanized iron sheet, whose inner diameter was just large enough to accommodate the neutron-counter tube within it. Thus we had a layer of U_3O_8 of about 1 cm. thick surrounding the counter tube. The metal container was earthed. The U_3O_8 was prepared by igniting uranium-nitrate crystals.

The uranium container was further surrounded by a bigger double-cylinder of galvanised iron sheet containing paraffin of about 6 cms. thickness. The two open ends of the paraffin cover were also closed with paraffin bricks of suitable size.

The necessary measurement comprises of the following steps:

(1) Determination of the back-ground of the counter, when it is surrounded by the paraffin cover.

(2) Determination of the effect of slow neutrons by interposing the uranium container between the neutron-counter and the paraffin cover.

(3) Determination of the effect of fast neutrons, any other penetrating ionising particle or γ-rays emitted by U by keeping the uranium container in situ and removing the paraffin cover.

(4) calibration of the over-all efficiency of the neutron counter under working conditions by means of a weak standard (Rn + Be) source.

The calibration of the detector was performed by a method similar to that adopted by Booth, Dunning and Slack (1939) in their investigation on the delayed neutron emission from Uranium after the removal of the source of irradiation. For this purpose, the U_3O_8 container was replaced by a weak (0.11 mc.) Rn + Be source. In order to maintain the symmetry of the arrangements, as far as practicable, the (Rn + Be) source was contained in a narrow glass tube of about the same length as that of the uranium cylinder and inserted into a narrow groove in the inner surface of the paraffin cover as shown in Fig. 1. Care was taken to start the standardisation measurements about 4 hours after admitting radon gas into the glass tube containing Be, when the calculated rise of RaC from the pure emanation attained the maximum values. The effective number of neutrons per sec. per mc. from the (Rn + Be) source was taken to be 15,000 as recommended by Dunning *et al.*

RESULTS

The experimental results are presented in the following table:—

TABLE I

Expt.	Disposition of the experimental arrangement	Counts per minute	Remarks
A	Counter+paraffin cover	0.83 ± 0.02	Back-ground effect
B	Counter+U_3O_8 cylinder + paraffin cover	2.97 ± 0.04	Counts of slow neutrons per minute = B−A = 2.14
C	Counter+U_3O_8 cylinder	0.85 ± 0.04	Effect of neutrons emitted directly by uranium (without being slowed down by paraffin) C−A = 0.02
D	Counter+(Rn+Be) source (0.11 mc.)	206	* Efficiency of the counter = 1/480

* Determined from the ratio of the actual number of counts recorded by the neutron counter to the number of neutrons emitted by the Rn+Be source when placed in position indicated in Fig. 1.

It will be recognised from a comparative study of (C-A) with (B-A) that uranium emits essentially fast neutrons. When these are slowed down by scattering in the paraffin cover, they produce an effect which is approximately three times the back-ground effect. Incidentally, it is also clear that the γ-ray emitted by U has practically no influence on the number of counts per minute. That the observed increase in the number of counts is really due to the detection of slow neutrons was further verified by covering the counter with a cadmium foil of 0.5 m.m. thickness, when the number of counts per min. practically dropped down to the back-ground level.

Now, since the efficiency of our detector is only 1 in 480, the actual number of neutrons emitted by 1.2 Kg of uranium is 1027 per minute. Assuming, as before, that U^{238}, the most abundant isotope (99.2%) of uranium, undergoes spontaneous fission, and that a single neutron is emitted per fission process, the corresponding half-life period comes out to be 3.8×10^{15} years.

DISCUSSION OF THE RESULT

We have used two different methods to measure the half-life period of the products of the spontaneous fission of the uranium nucleus. The first one described in the Transactions of the Bose Research Institute (1945), is based upon the actual counting of the frequency of fission by measuring the large impulses produced by them in an ionisation chamber or a proportional counter. The second method described in the present paper counts the number of neutrons emitted per given mass of uranium nucleus. Now, in order to calculate the mean half-life of the spontaneously disintegrating nucleus, we have to make some assumption as to which of the three isotopes of U, is undergoing fission.

For the sake of definiteness, we assume that the most abundant of the isotope, *viz.*, U^{238} is undergoing fission. Based on this assumption, it is found that

$$T_{f_1} = 1.3 \times 10^{16} \text{ years by method (1)}$$

and

$$T_{f_2} = 3.8 \times 10^{15} \text{ years by method (2).}$$

We find that there is a discrepancy of the order of 3.4 in the ratio of the values of the mean life of spontaneous fission of U nucleus given by the methods (1) and (2). The second value is based on the assumption that associated with each spontaneous fission only one neutron is emitted. This discrepancy is removed if the assumption is made that on an average about 3.4 neutrons are emitted instead of one. This value is of the same order as that found by Dode' (*loc. cit.*) for the emission of fast neutrons associated with the induced fission of U nucleus. The value found will be independent of any particular assumption as to which particular isotope undergoes fission. It is obvious that the absolute value of T_f will change if it is found that some other isotope of uranium is responsible for the spontaneous fission ; but the ratio of the two T_f, which gives the number of neutrons emitted per nuclear fission is independent of any such particular assumption. More precise experiment is in progress.

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