Fission and chain decay of uranium*

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1. BASIC PHENOMENA

A thorough radiochemical analysis of radioactive elements formed as a result of irradiation of uranium and thorium by neutrons led, as is well known, Hahn and Strassmann[1] on making more precise measurements of a number of interesting features discovered by them and by Curie and Savitch[2] to a remarkable result. They succeeded in proving with certainty that one of the products obtained on irradiating uranium by neutrons is barium, while according to the scheme generally accepted at that time that had been proposed by Fermi *et al.*[3] transuranic elements should be obtained under those conditions.

On the basis of the facts described in the above articles Meitner and Frisch[4] suggested that as a result of the capture of a neutron by a uranium or a thorium nucleus what happens is not emission of a β-particle, as was supposed by Fermi[3] but fission (or the disintegration) of the compound nucleus into two approximately equal parts. On the basis of the Bohr theory of heavy nuclei[5] they visualized the fission mechanism in the following manner. The surface energy[6] of heavy nuclei due to their large charge will, as a rough approximate calculation indicates, be not very great. Therefore it is possible that a uranium nucleus is not very stable with respect to changes of shape, and as a result of the initiation of oscillations of the nucleus excited upon neutron capture it can fall apart into two nuclei of approximately the same size. The electrostatic repulsion of these two nuclei will communicate to them a kinetic energy of the order of 200 MeV. A quantity of the same order of magnitude can be obtained if one utilizes for the calculation the values of the mass defect of uranium and of elements from the middle part of the periodic system.

Experimental confirmation of this assumption was published by Frisch[7] in the English journal "Nature" of 18th February 1939 (the note—a letter to the editor—is dated 16 January). In an ionization chamber covered inside by a layer of uranium and irradiated by neutrons from a radium-beryllium source of 300 mCi strong ionization pulses appeared. A linear amplifier connected to the chamber in its turn was connected to a thyratron so adjusted that it responded only to pulses corresponding to not fewer than $5 \cdot 10^5$ ion pairs. When the source was placed at a distance of 1 cm from the chamber, the number of pulses amounted to approximately 15 in the course of 1 minute.

Surrounding the neutron source placed at a distance of 4 cm from the chamber by paraffin doubled the effect.

Changing the thyratron adjustment it was possible to establish that the largest pulses corresponded approximately to two million ion pairs with the path length of the ionizing particle being equivalent to not more than 0.8 cm of air. Taking a reasonable ratio between the atomic weight and the effective charge the above numbers yielded for the atomic weight of the fragment a value of not less than 70.

Similar results were obtained with thorium. However, in this case surrounding the source by paraffin did not increase the effect, but on the contrary slightly diminished it.

Frisch notes another experiment proposed by Meitner which was expected to be performed in the near future: on a metallic plate placed in front of the uranium layer bombarded by neutrons an active deposit should be produced of light atoms forming in the course of fission of uranium.

Practically simultaneously with the work of Meitner and Frisch an article by Joliot[8] appeared similar in its content. This work was reported at a meeting of the French Academy of Sciences on 30 January 1939. The reasoning is based on the same experimental facts. Joliot produced the following possible scheme for fission:

$$U_{92}^{238}+n_0^1=Rb_{37}^{98}+Cs_{55}^{141}.$$

From fragments that have a considerable excess of neutrons compared with the stable nuclei of the same atomic number as a result of a series of β-emissions one can expect the appearance of Pr_{69}^{141} and Mo_{42}^{98} as final products.

Joliot's experiment was carried out in the following manner. The neutron source (700 mCi of radon and beryillium) was placed inside a brass cylinder of 20 mm diameter and 5 cm in height; a layer of uranium oxide was deposited on the external surface of the cylinder. All this was placed in a bakelite cylinder with an internal diameter of 26 mm. In separate placement within the bakelite cylinder of a neutron source or a cylinder covered with uranium oxide on the surface (the internal one of course) of the bakelite no activity was observed. But on their being placed there together resulted in the appearance of activity on the bakelite. Placing between the surfaces of brass and bakelite layers of mica with different stopping power it was possible to verify that the path in air of particles ejected from uranium was appoximately 3 cm.

Knowing the amount of uranium oxide deposited on the brass cylinder the intensity of the neutron source and the value of the activity appearing on the bakelite it was possible to estimate the effective cross section of the uranium nucleus for neutrons initiating the fission process. It turned out to be of the order of 10^{-25} cm^2.

Joliot observed similar effects also in the case of thorium. Fission of heavy nuclei irradiated with neutrons was also confirmed by Jentschke and Prankl[9], Droste[10] and Thibaud and Moussa.[11]

It should be noted that the possibility of fission of heavy nuclei was pointed out on the basis of general considerations of stability of different nuclei by Ida Noddack[12] already in 1934. But her remark remained unnoticed, none of the experimenters tried, and if anybody did try, then

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apparently unsuccessfully, to observe the phenomenon predicted by her.

Bohr who at the beginning of 1939 was in America was informed by telephone concerning the interpretation proposed by Meitner and Frisch of the data of the Hahn-Strassmann experiments and acquainted American physicists with this idea. In the course of several days in a number of American laboratories that had powerful neutron sources experiments were carried out which also supported the Meitner and Frisch hypothesis and brought out certain essential details of the phenomenon.

Roberts, Meyer and Hafstad[13] working with neutrons from a lithium target bombarded by deuterium ions of 1 MeV energy and using an ionization chamber for detecting the decay products obtained a positive result for uranium and thorium; for bismuth and lead, thallium, mercury, gold, tin, and silver the effect was by a factor of at least a thousand weaker than for uranium. On acting by γ-rays (3 μA of protons of 1 MeV energy bombarding a target containing lithium or fluorine) no fission was observed.

Surrounding the uranium or thorium by paraffin in order to slow down the bombarding neutrons and using a cadmium screen (which, as is well known, absorbs slow neutrons) in order to exclude slow neutrons, Roberts, Meyer and Hafstad have shown that in the case of uranium fission can occur either under the action of fast neutrons (with a limiting energy of not lower than 0.5 MeV, but lower than 2.5 MeV), or under the action of slow neutrons. In the case of thorium only fast neutrons can give rise to fission.

Similar results were obtained by Fowler and Dodson[14] and also by Green and Alvarez.[15] The last two also showed that between the instant of a neutron colliding with uranium and the instant when fission occurs not more than $3 \cdot 10^{-3}$ s elapse.

Abelson[16] showed using chemical methods and analysis of the emitted x rays that among the products of irradiation of uranium by neutrons there is present iodine,—this also is a confirmation of the correctness of the hypothesis of fission. Feather[17] having discovered an asymmetry in the distribution of fragments showed that between the instant of a neutron colliding with a uranium nucleus and the instant of fission a time elapses shorter than is required for slowing down the nucleus which has acquired a certain velocity as a result of capture of the fast neutron that gave rise to fission, i.e., not more than 10^{-12} s.

D. Corson and R. Thornton[18] observed fission of uranium in a Wilson chamber containing a mixture of air and vapors of alcohol and water with a total pressure of 15 cm of Hg. Uranium in the form of UO_3 was placed in the chamber on thin collodion films. On a stereoscopic photograph given in the paper by Corson and Thornton (cf., Fig. 1) it can be seen that as a result of fission two strongly ionizing particles emerged in opposite directions. The photograph is very interesting because of the presence of a fork (near the lower end of the track) which makes it possible to estimate the mass of the fragment obtained as a result of fission. The density of the side branch of the fork is so great that it cannot be produced by a proton and consequently is

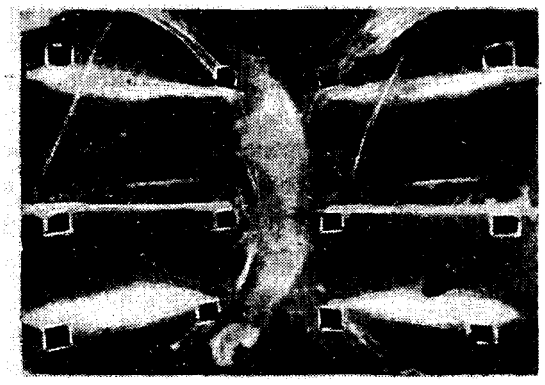

FIG. 1. Photograph from paper[18] by D. Corson and R. Thornton.

the track of an ion of carbon, nitrogen or oxygen. The fact that the main branch of the fork practically is not deflected from the initial direction leads to a value for the mass of the fragment not smaller than 75.

On some photographs it appeared as if there were more than two particles, but the authors do not regard this material as sufficiently reliable.

An investigation of the fission process in a Wilson chamber was also carried out by Perfilov.[19] On one of his photographs he observed a fork which cannot be ascribed to an elastic relation of two particles since the branch of the fork makes an angle greater than 90° with the main track. The author points out that perhaps we are here dealing with subsequent fission of the fragment itself.

Zhdanov, Mysovskiĭ and Mysovskaya[20] observed fission of uranium with the aid of tracks produced by fragments in a special photographic emulsion.

One can assume that fission events can occur not in a unique manner, i.e., that the masses and charges of the fragments obtained from uranium can vary within certain limits. E. McMillan[21] made an attempt to distinguish different possible types of fragments by using the possible difference in their path lengths. In front of the uranium irradiated by neutrons several layers of thin cigarette paper were placed (the air equivalent of a layer is approximately 1 cm). After exposure the activity was found on the first three layers of paper. The curves of the variation of activity with time for the second and the third layer turn out to be the same within the limits of accuracy of the experiment. The activity of the irradiated uranium itself was quite different; it exhibited a strong activity with a half period of 25 min that corresponded, as McMillan supposed, to a uranium isotope formed by resonance capture of a neutron by uranium.[22] Also an activity was observed with a period of two days.

Thus in the course of the first months of 1939 in a number of laboratories undoubted proof was obtained of the correctness of the hypothesis of the fission of uranium and thorium nuclei under the action of neutrons and the basic characteristics of this process were clarified.

Somewhat later (July 1939) Grosse, Booth and Dunning[23] investigated the behavior of protoactinium under neutron bombardment and just as in the case of

thorium fission was observed only under the action of fast neutrons.

We present here numerical values characterizing the physical phenomena associated with the process of uranium fission (the methods of determining a number of these quantities will be presented below).

The (total) energy of the fragments turned out to be approximately 150–200 MeV in agreement with theoretical concepts.

The fission cross section for radon-beryillium neutrons is $\sim 0.1 \cdot 10^{-24}$ cm^2 (Refs. 8, 24). For monochromatic neutrons with an energy of 2.4 MeV the fission cross section of uranium amounts to $0.5 \cdot 10^{-24}$ cm^2 (for thorium—$0.1 \cdot 10^{-24}$ cm^2).[25]

The fission cross section of uranium for thermal neutrons is $2 \cdot 10^{-24}$ cm^2.[24] The cross section for the radiative capture of uranium for thermal neutrons (with the formation of U^{239} and the subsequent β-emission with the formation of ecarhenium) is $(1.3 \pm 0.45) 10^{-24}$ cm^2 (Ref. 26) or $1.2 \cdot 10^{-24}$ cm^2 (Ref. 65) (the error is not stated).

The energy of the neutrons (with respect to the fragment of the nucleus from which the neutron emerged) ejected in the course of fission of a uranium nucleus under the action of slow neutrons is equal to ~ 2 MeV.[61] Since the neutron can emerge at any angle with respect to the direction of motion of the fragment of the nucleus the neutron energy can lie within the limits from 1 to 3 MeV.[61]

The number of neutrons ejected in the act of fission occuring as the result of capture of a slow neutron is characterized by the following numbers: 3.5 ± 0.7 (Ref. 62), 2.3 (Ref. 61), 2.4 (Ref. 64).

Both the energy and also the number of neutrons ejected in the act of fission brought about by a fast neutron have not been determined with any degree of accuracy, although fairly detailed investigations were carried out in this direction.[27]

2. THE MECHANISM OF FISSION

The general theory of phenomena occurring in the nuclei as a result of their interaction with neutrons was given by Bohr[28] and by Bohr and Wheeler.[29] The theory specifically of fission was also independently worked out by Frenkel' [30] and by Flügge and Droste.[31] In this section we shall restrict ourselves to the presentation of the qualitative side of the problem.

Bohr regarded the process of nuclear fission similarly to the previously known nuclear reactions as occurring in two stages. At first a compound nucleus is formed from uranium and a neutron in which the energy associated with the capture of the neutron is in a form reminiscent of the thermal motion of a liquid or a solid. The second stage is either the radiation of this energy or its transfer into such a form which can lead to the disintegration of the compound nucleus. In the case of the ordinary reactions represented by the emission of a proton, a neutron or an α-particle we are dealing with a concentration of a significant portion of the excitation energy on one of the particles situated near the surface, and this is analogous to the evaporation of a molecule from a liquid drop. In the case of the phenomenon of fission a significant part of the energy must be transferred into a special kind of motion of the nucleus as a whole that leads to such a deformation of the surface of the nucleus that can lead to the rupture of the nucleus "similarly to the formation of two drops of a liquid from a single drop" (quotation from Ref. 28). Bohr shows that in case of a sufficiently high nuclear charge the probability of fission is indeed of the same order of magnitude as the probability of radiation (radiative capture of the neutron) or the probability of the inverse evaporation of the neutron (equivalent to inelastic scattering of the neutron).

Bohr assumes that the whole complicated set of events in the phenomenon observed in bombarding uranium (or thorium) by neutrons reduces to two main processes: radiative capture of a neutron, leading to the formation of a β-radioactive isotope of uranium and to fission which can occur in different ways, i.e., with the formation of fragments having different masses and charges.

Meitner, Hahn and Strassmann[22] showed that in the case of uranium and thorium the capture of a neutron with the formation of a radioactive isotope is of a resonance nature. Uranium in this respect has been investigated more thoroughly: for it the resonance energy of neutrons is equal to approximately 25 eV. Near the resonance the effective cross section of uranium for neutron capture is equal to approximately 10^{-21} cm^2; this number enables one to ascribe the radiative capture of a neutron only to the main uranium isotope (of atomic weight 238), since the light isotope whose abundance is 0.007 in order to provide an effective cross section of 10^{-21} cm^2 would have to have a cross section exceeding the upper limit admissible by theory. However since the increased probability of capture of neutrons near 25 eV does not lead to an increase in the number of fissions one can assert that for the excited nucleus of U_{92}^{239} produced in the process the probability of radiation is much higher than the probability of fission. The unexcited nucleus of U_{92}^{239} which results after radiation is unstable only with respect to β-decay (as a result of which one obtains the only "surviving" transuranic element Eca Re_{93}^{239}.

In the capture of fast neutrons both in the case of uranium and thorium the probability of fission, which increases (in accordance with Bohr's concepts) with increasing excitation energy more rapidly than the probability of radiation, attains quite large values. The cross section for fast neutrons according to the theory must be of the order of nuclear dimensions, which is what in fact is observed.

The question remains concerning the nature of fission under the action of slow neutrons. Bohr assumes that this process is due to the capture of slow neutrons by the uranium isotope of mass 235. The fact that the decay products under the action of slow neutrons are the same as under the action of fast neutrons is explained by the possibility of an entire spectrum of fragments both from U_{92}^{239} and from U_{92}^{235}. Since the neutron binding energy in the nucleus with an even atomic number is considerably greater in the case of an even atomic weight than in the case of an odd atomic weight the excitation energy of the compound nucleus U_{92}^{236}

is considerably greater than in the case of U_{92}^{239}. Therefore even slow neutrons can guarantee a greater fission probability (compared to the probability of radiation). As a result of the great fission probability the levels will be broadened, whose density will be very great due to the high excitation energy. Even a continuous spectrum can occur. As a result the cross section for the capture of slow neutrons (leading to fission) will be inversely proportional to the velocity, and this is observed experimentally.[24] Accordingly at "medium" neutron energies (from 1 to 10^6 eV) fission will not be observed, since for the isotope of mass 235 their velocity will be too great, while for the isotope of mass 238 it will be too small.

3. FISSION PRODUCTS OF URANIUM AND THORIUM

As a result of the work of Hahn and Strassmann[1] and the later discovery of uranium fission[4,7,8] the schemes of articifially radioactive families—the transuranic elements arising as a result of bombarding uranium with neutrons[22] that had been proposed by the same Hahn and Strassmann together with Meitner were completely swept away. It became obvious that all the accummulated material had to be revised in the light of the new fact—the fission of heavy nuclei, and subsequent investigations were directed towards finding elements from the middle part of the periodic system of elements.

The fact that substances which were thought to be "transuranic" in fact are representative of the middle of the periodic system was very graphically proven by Meitner and Frisch.[32] They showed that a radioelement separated from the irradiated uranium together with platinum coincides with respect to the decay half-life with the radioelement separated by them together with platinum from water above which at a distance of 1 mm an irradiated layer of uranium was placed. Consequently, the "transuranic elements" are obtained in the course of fission of a uranium nucleus since by itself the capture of a neutron with the emission of a light particle could not have communicated to the nucleus an energy sufficient to kick it out from the irradiated layer through air and into water.

Khlopin, Passvik-Khlopina and Volkov[33] found some difference in the nature of the decay of fragments collected on a glass surface (after separating them out with platinum) and a similar deposit obtained directly from irradiated uranium. In the former case after 30–40 hours a complete decay of activity is observed, while in the latter case after the expiration of this period of time an activity is observed with a greater decay period of approximately 70 hours.

A number of essential data on the nature of fission products was obtained by Abelson.[34] Along with chemical separation Abelson for the identification of the nature of the carriers of activity used the possibility of observing x rays emitted by them. Prior to the discovery of uranium fission Abelson assumed that the radiation observed by him was L-rays of "transuranium" with a decay period of 72 hours. Some discrepencies in the values of the absorption coefficient of this radiation were attributed to unsatisfactory geometrical conditions of the measurement.

Having reexamined and improved the method in connection with the discovery of uranium fission Abelson at first quite definitely found that the radiation observed by him corresponds to the K-group of iodine. A further analysis established the presence of a number of radioactive isotopes of antimony, tellurium and iodine.

Let us examine the path used to establish the genetic relationships in one of the "families" obtained by Abelson. After irradition an active deposit was separated from uranium with tellurium. After a week (as a result of which all the tellurium atoms with short periods have practically completely decayed) extraction of tellurium from the deposit was carried out. The activity of this tellurium grew in the course of approximately 10 hours after separation, which indicates the formation of a daughter substance with a period of approximately 2.5 hours which is iodine according to all its chemical properties and its x rays. An exact measurement of the decay period of this iodine obtained from a long period tellurium gave a value of 2.4 hours. For tellurium itself the decay half-life of 77 hours was obtained which corresponds to the period of 66 hours described by Meitner, Hahn and Strassmann.[2] The difference is apparently associated with the presence of an active tellurium with a half-period of 30 hours which appreciably distorts the decay curve in the course of the first two to three days. According to Meitner, Hahn and Strassmann[2] the substance with the decay half life of 66 (77) hours is obtained from a substance with a half-period of 59 minutes. Abelson having conducted a number of separations of tellurium from the irradiated uranium at 10-minute intervals (100 mg of tellurium were introduced into solution for each separation) and measuring the activity of the precipitates obtained found a very strong decrease in the activity of the subsequent sediments compared with the preceding ones by approximately a factor of 3.5. From this it follows that the 77-hour tellurium is obtained from approximately a 5-minute antimony (or from a 5-minute tin and antimony with a still shorter period). Since among the products obtained from uranium there is a 5-minute antimony then it probably is the predecessor of the 77-hour tellurium.

Abelson assumes that the 2.4 hour iodine transforms into one of the stable isotopes of xenon of atomic weight 132, 134, or 136.

In the case of this family of antimony-tellurium-iodine-xenon examined by us there is no possibility of obtaining an exact identification, i.e., to determine the atomic weights as well as the atomic numbers. But in some other cases (70-minute and 10-hour tellurium and the 8-day iodine) one succeeds in determining also the atomic weight, and specifically in those cases when there are available data concerning artificially radioactive isotopes of the corresponding elements obtained by irradiation with slow neutrons.

We present a compilation of data obtained by Abelson concerning the radioactive isotopes of antimony, tellurium and iodine found among the products of the fission of uranium.

Antimony	Tellurium	Iodine	Atomic Weight
80 hours	10 hours		127
4.2 hours	70 min		129
	30 min (isomer)	8 days	131
	30 hours (isomer)	8 days	131
5 min	77 hours	2.4 hours	132, 134 or 136
10 min	43 min	54 min	
10 min	60 min	22 hours	

A similar method was used by Feather and Bretcher[35] to discover iodine among the fission products.

Khlopin, Passvik-Khlopina and Volkov[36] obtained for the half-period of iodine the values of 3.7 and 28 hours. The authors assume that the difference of their numbers from the numbers obtained by other authors is associated with the fact that they (Khlopin *et al.*) used more accurate methods of separation of haloids.

For tellurium Khlopin, Passvik-Khlopina and Volkov[37] obtained a half-period of 56 hours (Abelson—77 hours).

Hahn and Strassmann further showed[38] that the 66 hour substance in fact consists of two substances, specifically that in addition to tellurium there is also a 66-hour molybdenum. This molybdenum apparently is identical with the molybdenum obtained by Seaborg and Segre[39] by irradiating molybdenum with slow neutrons.

Hahn and Strassmann[40] found among the fission products isotopes of strontium and also of the inert gases. In particular the presence of xenon is indicated by the fact that active isotopes of cesium were found by blowing air through a cooled vessel containing a solution of a uranium salt and being irradiated.

Heyn, Aten and Bakker[41] found by a similar method an isotope of rubidium with a half-period of 16 ± 2 min which forces one to assume among the fission products of uranium the presence of krypton. Krypton with a half-period of 3 hours which transforms into an 18-minute isotope of rubidium also found by Langsdorf[42] among the fission products of thorium. Since the 18-minute half-period appears in the case of neutron bombardment of rubidium[43] and since krypton with an atomic weight 86 is stable, the 18-minute half-period should apparently be ascribed to the isotope of rubidium with atomic weight 88, and not 86. The 3-hour krypton was also observed among the fission products by Hahn and Strassmann.[44] Heyn, Aten and Bakker[41] found that xenon has a half-period of approximately 1 min and found that the 12-minute barium is obtained only from the primary fragments but not from the gas. This indicates that the 12-minute barium is either a primary fission product, or is formed from a very rapidly decaying xenon, or is formed from cesium obtained directly in the course of fission and not from xenon.

On the basis of the above facts Hahn and Strassmann[45] consider that one of the possible schemes of fission is the following:

$$U_{92}+n_0=Kr_{36}^{88}+Ba_{56},$$

$$Kr_{36}^{88}\xrightarrow[3\text{ hours}]{\beta}Rb_{37}^{38}\xrightarrow[17\text{ min}]{\beta}Sr_{38}^{88},$$

$$Ba_{56}\xrightarrow[14\text{ min}]{\beta}La_{57}\xrightarrow[-2.5\text{ hours}]{\beta}Ce_{58}.$$

We also present without going into a detailed discussion of the sources the most probable schemes of families occurring among the fission products of uranium (according to Hahn and Strassmann[45]):

$$Xe_{54}^{139}\xrightarrow[?]{\beta}Cs_{55}^{139}\xrightarrow[6\text{ min}]{\beta}Ba_{56}^{139}\xrightarrow[86\text{ min}]{\beta}La_{57}^{139},$$

$$Xe_{54}\xrightarrow[\sim15\text{ min}]{\beta}Cs_{55}\xrightarrow[33\text{ min}]{\beta}Ba_{56}\xrightarrow[300\text{ hours}]{\beta}$$
$$\times\xrightarrow[300\text{ hours}]{\beta}La_{57}\xrightarrow[\sim36\text{ hours}]{\beta}Ce_{58},$$

$$Sr_{38}\xrightarrow[7\text{ min}]{\beta}Y_{39},$$

$$Kr_{36}\xrightarrow[\text{very short}]{\beta}Rb_{37}\xrightarrow[80\text{ sec}]{\beta}Sr_{38}\xrightarrow[6\text{ hour}]{\beta}Y_{39}\xrightarrow[3.5\text{ hours}]{\beta}Zr?,$$

$$Kr_{36}^{88}\xrightarrow[3\text{ hours}]{\beta}Rb_{37}^{88}\xrightarrow[17\text{ min}]{\beta}Sr_{38}^{88}.$$

Thibaud and Moussa,[46] Dodson and Fowler,[47] Bretcher and Cook,[48] Hahn and Strassmann,[44] and also Khlopin, Passvik-Khlopina and Volkov[36] found among the uranium fission products bromine which indicates the possibility of an even more asymmetric fission of the uranium nucleus than those already discussed. Hahn and Strassmann[44] consider the half-periods of the two bromine isotopes discovered by them to be equal to 35 and 230 min.

Among the fission products of thorium a number of products has been found which are observed in the case of uranium: Ba—300 hours[49] and 86 min.,[50] Mo and Te 66 hours,[49] I—2.4 hours,[49] Rb—18 min,[50] Cs—10 and 33 min.[50]

Nishina, Yasaki, Esoe, Kimura and Ikawa[51] in investigating the fission products of thorium and uranium found activity of sediments obtained with a large number of metals; however their publication is only of a preliminary nature.

Meitner[52] in analyzing the fission products of thorium obtained results which appear to indicate the presence of some products which have not been observed in the case of uranium, specifically, in obtaining an active deposit with the aid of H_2S, half-periods of 40 min and 14 hours were obtained which have not been observed in the case of uranium.

We also note that according to Joliot[53] in the case of uranium irradiation with slow neutrons results in the appearance of long-period activities more often than irradiation with fast neutrons. On the other hand Bjerge, Bros-

trom and Koch[54] obtained completely identical curves of the decrease of activity in the case of irradiation of uranium by fast or by slow neutrons.

4. EMISSION OF NEUTRONS AND γ RAYS IN FISSION

Instantaneous and delayed emission. The strong excitation both of the compound nucleus (associated with the capture of a neutron) and of fragments obtained in the process of fission gives every reason to assume that the fission process is accompanied by emission of neutrons. Neutrons can be "evaporated" from the compound nucleus or from the fragments, can be "splashed out" in the course of the act of fission similarly to the formation of small droplets in the interval between the main drops into which a jet of liquid is broken up;[29] finally, neutron radioactivity of fragments can occur—emission of neutrons after a relatively long time after the act of fission (associated with the excitation of the nucleus as a result of β-decay).

Experimental study of the time of emission of neutrons in fission indeed showed[55–57] that a practically instantaneous emission of almost the entire number of emitted neutrons occurs along with which, however, there is also observed a certain number of delayed neutrons.

A direct measurement of the upper limit on the time of delay of the process of fission was carried out as we have already indicated by Green and Alvarez[15] who obtained for the upper limit of the delay of the process of fission $3 \cdot 10^{-3}$ s. However, there is no justification for rejecting the possibility of the emission of the main number of neutrons not simultaneously with the act of fission but as a result of disintegration of the fragments. Basing themselves on this consideration Gibbs and Thomson[57] investigated experimentally whether neutron emission might take place after a certain interval of time after fission. Using an interrupted source of neutrons[58] with a period of 0.005 s they showed that the principal bulk of neutrons is emitted not later than 0.001 s after the impact of the neutron which gives rise to fission.

Delayed neutrons. The emission of neutrons by uranium with a delay was observed by Roberts, Meyer and Wang.[55] An ionization chamber lined with boron was placed at a distance of several centimetres from a lithium target bombarded with deuterons. Both the target and the counter were surrounded by paraffin. Under such conditions the pulses in the ionization chamber ceased as soon as the deuteron bombardment was stopped. If, however, between the chamber and the target a vessel was placed containing 100 μg of uranium nitrate then one could observe ionization pulses in the course of approximately 1.5 min from the instant of cessation of bombardment. Initially approximately one neutron per second was oberved in the chamber. This neutron activity fell off with a period of 12.5 ± 3 s. Also gamma activity with approximately the same period was observed. Therefore it was necessary to establish whether the observed delayed neutrons are photoneutrons, or are they directly emitted by fission products (primary or secondary ones). Roberts, Hafstad, Meyer and Wang[56] having somewhat improved the technique of their experiment demonstrated with total certainty that direct emission of neutrons took place. They also showed that along with the aforementioned approximately 12-second γ radiation there are also present at least three other γ radiations with greater periods. Both the delayed neutrons and also all the observed γ rays were observed (similarly with the fission process itself) under the action either of fast or of thermal neutrons and were not observed under the action of neutrons of intermediate energies (from carbon). In the case of action of fast neutrons (lithium-deuterium) the effective cross section of uranium for obtaining delayed neutrons amounted to $4 \cdot 10^{-26}$ cm^2.

The energy of the delayed neutrons determined by the tracks of atoms in a Wilson chamber amounted to approximately 0.5 MeV. Thorium gave a picture similar to that of uranium, but the number of neutrons was smaller by approximately a factor of four than in the case of uranium.

Mouzon, Park and Richards[59,60] have analyzed with the aid of a Wilson chamber in a magnetic field the γ radiation accompanying the fission of uranium. The expansion of the chamber occurred alternately: during irradiation and 1/2 s after the irradiation was stopped. The energy distribution of the γ quanta in both cases was approximately the same, but the number of quanta during irradiation was considerably greater. Quanta with energies up to 9–10 MeV were observed.

5. YIELDS AND ENERGY DISTRIBUTION OF NEUTRONS FROM URANIUM UNDER THE ACTION OF SLOW NEUTRONS

As the most direct method and one guaranteeing the greatest reliability for determining the number of neutrons emitted in fission one should regard the method of Zinn and Szilard.[61] A valuable feature of this method is the fact that for the determination of the number of neutrons emitted in the course of fission there is no need to know any kind of effective cross sections and also the fact that the conditions of conversion of fast neutrons to slow ones are not significant. Because of this, naturally, the relative accuracy of the result is probably greater than in the following experiments of Halban, Joliot, and Kowarski,[62] of Anderson, Fermi, and Hanstein[63] or of Anderson, Fermi, and Szilard.[64]

The method of Zinn and Szilard essentially consists of determining with the aid of a spherical ionization chamber the number of fast neutrons arising in a cell filled with uranium under the action of radium-beryillium photoneutrons slowed down by paraffin and the number of fission events occurring in the layer of uranium (containing a definite amount of uranium) deposited on the internal walls of a flat ionization chamber replacing the cell containing uranium. The ratio of these two quantities gives us, taking into account the difference in the amount of uranium and certain other corrections, the number of neutrons emitted in an act of fission.

Zinn and Szilard obtained 2.3 neutrons per fission. Taking into account the fact that the fission cross section amounts to[24] $2 \cdot 10^{-24}$ cm^2 and the cross section for radiative capture[65] is $1.2 \cdot 10^{-24}$ cm^2 we obtain the value of 1.4 neutrons per neutron captured by uranium which is in good agreement with the value of 1.5 obtained by Ander-

son, Fermi, and Szilard,[64] but somewhat lower than the value obtained by Halban, Joliot, and Kowarski[62] (3.5 neutrons per fission or 2.2 per capture).

According to measurements made by Zinn and Szilard[61] the energies of neutrons lie within the limits from 1 to 3 MeV, which corresponds to the energy of the neutrons of approximately 2 MeV with respect to the fragment from which they emerge. The maximum value of the neutron energy is obtained when the neutron emerges in the direction of motion of the fragment; the minimum one when the neutron emerges in the direction opposite to the motion of the fragment.

The emission of fast neutrons in the course of fission of uranium was first demonstrated by Dode, Halban, Joliot, and Kowarski[66] with the aid of a very elegant experiment. A source of photoneutrons having an energy not greater than 0.1 MeV was surrounded by a layer of uranyl nitrate. This system was placed in a vessel with 8 l of carbon bisulphide in which 200 mg of phosphorus were dissolved. After six days of irradiation the phosphorus was distilled off from the carbon bisulphide; it showed considerable radioactivity which apparently is a consequence of the endothermic reaction

$$S_{16}^{32}+n=P_{15}^{*32}+p,$$

which proceeds under the action of neutrons having an energy of not less than 0.9 MeV. In veiw of the low energy of the primary neutrons we have to conclude that fast neutrons that bring about the transformation of sulphur into radioactive phosphorus appear on absorption of the slowed down primary neutrons in uranium i.e., in the course of uranium fission. Halban, Joliot, and Kowarski[67] with the aid of an ionization chamber discovered the appearance in fission events of a certain relatively small number of very fast—up to 11 MeV-neutrons.

A number of French and American publications[62–64] was devoted to the determination of the number of neutrons emitted in fission by measuring the total number of neutrons present (in a quasi-steady state) in a large vessel with a solution of a uranium compound in water (or a mixture of a uranium compound and water) containing at its center a neutron source.

With the aid of a dysprosium detector or some other similar method the density ρ of slow neutrons was determined at different distances a from the neutron source and the integral $\int \rho a^2 da$ was determined in the presence and in the absence of uranium or upon the replacement of the uranium salt by another salt. At first Halban, Joliot, and Kowarski[62] found some increase in the integral when the 1.6-molar solution of ammonium nitrate was replaced by a solution of uranyl nitrate of the same concentration. Starting with a distance of 13 cm from the source the neutron density increases in the case of the solution of uranyl nitrate; for $a=25$ cm ρ increases by approximately a factor of five. Since the radon-beryllium photoneutrons used in this case cannot initiate a reaction of the n, 2n type, i.e., ejection of neutrons, the authors assume that the additional number of neutrons is associated with the emission of more than one neutron as a result of a fission event, brought about by the capture of a neutron.

In the next paper[62] already a definite value for the number of neutrons ν_f ejected as a result of a fission event was obtained. The aforementioned integral, as one can easily see, must be proportional to the quantity $Q\tau$ where Q is the number of neutrons produced per second (from the principal source and from uranium fission) and τ is the average lifetime of a neutron. Denoting the value of the integral by S and noting that $\tau \sim 1/\Sigma c_i\sigma_i$[1)] where c_i and σ_i are the concentrations and the neutrons capture cross sections, for atoms present in the system under consideration, one can write

$$S \sim Q\tau \sim Q\frac{1}{\Sigma_i c_i \sigma_i},$$

$$S' \sim Q'\tau' \sim Q'\frac{1}{\Sigma_i' c_i \sigma_i},$$

where the unprimed and primed letters refer to the absence and presence of uranium respectively or

$$\frac{S'}{S}=\frac{Q}{Q}\frac{\Sigma_i c_i \sigma_i}{\Sigma_i' c_i \sigma_i}.$$

Further the number of additionally generated neutrons can in the first approximation be expressed in the following manner:

$$Q'-Q=\Delta Q=Q\frac{c_U\sigma_f}{\Sigma_i c_i \sigma_i}\nu_f,$$

where σ_f is the cross section for capture with fission. From these equations we obtain in the first approximation

$$\nu_f=\frac{S'-S}{S}\frac{\Sigma_i c_i \sigma_i}{c_U\sigma_f}+\frac{\Sigma_i' c_i \sigma_i-\Sigma_i c_i \sigma_i}{c_U\sigma_f}.$$

Halban, Joliot, and Kowarski[62] obtained for $(S'-S)/S$ the value of 0.05 ± 0.01. Assuming the capture cross sections for thermal neutrons, expressed in units of 10^{-24} cm^2, to be equal to: for hydrogen 0.27 ± 0.03, for uranium 1.3 ± 0.45, and for uranium fission 2, the value of 3.5 ± 0.7 was obtained for ν_f. Taking resonance absorption of neutrons in the process of slowing down into account is accomplished by introducing the "cross section for resonance absorption" which was determined by special experiments of Halban, Kowarski, and Savitch[26] and was taken to be equal to 6.4 ± 1.1 (in these experiments measurements were made of the change in the activity of a gold detector in placing between it and the source of partially slowed down neutrons of two-centimeter layers of a solution of ammonium nitrate and uranyl nitrate of the same concentrations as in the experiments on determining the neutron density).

The determination of the distribution of the neutron density and the total number of neutrons in the system of a uranium compound and water were also carried out by Anderson, Fermi, and Hanstein[63] and by Anderson, Fermi, and Szilard.[64] In the last more exact investigation into 540 l of a 10 percent solution of $MnSO_4$ 52 cylindrical jars were

placed which were either filled with uranium oxide U_3O_8 of a total amount of 200 kg, or left empty. At the center a source of photoneutrons was placed. The activity of the $MnSO_4$ solution which is proportional to the total number of thermal neutrons in the volume turned out in the presence of uranium to be greater by 10% than without uranium. Without going into the details of additional experiments and recalculation of the experimental data we quote the results. The authors without indicating a value of the probable error assume that in place of one thermal neutron absorbed in uranium there arise on the average 1.5 neutrons.

In order to recalculate the yield of neutrons per fission event (and not per absorption of a neutron by uranium) one can use the data of Anderson and Fermi[65] according to which for a total capture cross section of uranium for thermal neutrons equal to $3.2\cdot 10^{-24}$ cm^2 the capture with fission accounts for $2\cdot 10^{-24}$ cm^2 and simple capture (with γ emission) accounts for $1.2\cdot 10^{-24}$ cm^2. From this we obtain

$$\nu_f=1.5\,\frac{2+1.2}{2}=2.4.$$

This lies between the values obtained by Zinn and Szilard (2) and by Halban, Joliot, and Kowarski (3.5 ± 0.7).

Hagiwara[68] also obtained analogous results and he, just as the French authors of Ref. 67, notes the presence of neutrons with energies up to 10 MeV.

6. CHAIN DECAY OF URANIUM

Chain decay based on fast neutrons. The emergence of more than one neutron upon absorption by uranium of one neutron in principle makes possible the realization of a nuclear chain reaction with branching chains. Halban, Joliot, and Kowarski[62] first noted that the discovery of the big yield of neutrons in fission is a step towards realization of an exothermic nuclear chain reaction. The quantitative treatment of the problem of the possibility of macroscopic decomposition of uranium was for the first time proposed by Perrin.[69] In this paper Perrin considered the action of only fast neutrons. Therefore the effective cross sections were assumed to be constant. It is assumed that uranium or some compound of it are placed in the shape of a sphere of radius R at the center of which there is a neutron source yielding Q_0 neutrons per second. Denoting by $F(r,t)$ the number of neutrons per cubic centimeter as a function of the time t and of the distance r of a volume element from the center Perrin introduces the equation

$$\frac{\partial F}{\partial t}=D\Delta F(r,t)+\left[c_U(\nu_f-1)\sigma_f-\sum_i c_i\sigma_{ci}\right]\bar{v}F(r,t) \quad (1)$$

for the variation of the neutron concentration with time. In this equation the first term on the right hand side represents the variation of the concentration due to the diffusion of neutrons, and the second one the change in the concentration due to the appearance of new neutrons and due to the absorption of neutrons. The notation used here is as follows: D is the diffusion coefficient for neutrons, c_U is the concentration of uranium atoms, c_i is the concentration of atoms of other substances, ν_f as before is the number of neutrons formed in the course of fission, σ_f is the cross section for capture involving fission, σ_{ci} is the cross section for the capture of an atom of the ith substance, $\bar{v}$ is the average velocity of the neutrons.

If the mean free path of neutrons is considerably smaller than the radius of the sphere then one can write $D=\bar{v}\lambda/3$; here

$$\lambda=\frac{1}{\Sigma c_i\sigma_{si}},$$

where σ_{si} is the cross section for the scattering of atoms of the ith substance. Taking this into account Perrin rewrote equation (1) in the form

$$\frac{\partial F}{\partial t}=\frac{\lambda}{3}\Delta(\bar{v}F)+\left[c_U(\nu_f-1)\sigma_f-\sum c_i\sigma_{ci}\right]\bar{v}F. \quad (2)$$

In the limiting steady-state regime we have $\partial F/\partial t=0$ (when $\partial F/\partial t>0$ the rate of reaction grows with time); then in place of (2) we obtain

$$\Delta(\bar{v}F)+a^2(\bar{v}F)=0, \quad (3)$$

where

$$a^2=\frac{3}{\lambda}\left[c_U(\nu_f-1)\sigma_f-\sum_i c_i\sigma_{ci}\right].$$

Taking into account the boundary conditions (flux near the center) and spherical symmetry Perrin obtains the solution of (3) in the following form:

$$\bar{v}F=\frac{3Q_0}{4\pi\lambda}\,\frac{1}{\sin aR}\,\frac{\sin a(R-r)}{r}.$$

The neutron density becomes infinite for $R_{cr}=\pi/a$. For $R>R_{cr}$ according to Perrin an explosive reaction of uranium fission will occur.

For the case of powder-like U_3O_8 Perrin, assuming its density to be equal to 4.2 g/cm^3 and taking $\sigma_{sU}=6\cdot 10^{-24}$ cm^2, $\sigma_{sO}=2\cdot 10^{-24}$ cm^2, $\sigma_f=10^{-25}$ cm^2, $\nu_f=3$ and neglecting absorption in oxygen, obtained $R_{cr}=130$ cm and for the crtical mass of U_3O_8 he obtained the value of 42 t.

Perrin assumed that the critical mass can be reduced if one surrounds the uranium-containing mass by some "neutron isolation," for example, by lead or iron. With a layer of iron of 35 cm the critical mass of uranium oxide according to Perrin's calculations reduces to 12 t.

Peierls[70] generalized Perrin's calculations and showed that the critical conditions found by Perrin do not depend on the position of the neutron source. Along with this Peierls also examined a system in which the probability of branching is very great so that the critical dimensions of the system are less than the mean free path of a neutron and the use of the differential equation of diffusion is inadmissable. This last case does not have any relation to uranium disintegration.

The essential defect of Perrin's work is that it does not take into account slowing down of neutrons occurring both in elastic as well as in inelastic collisions of neutrons with

TABLE I.

E_0 \ ν_f	1,5	2	3
3	0,63	0,84	1,26
2	0,3	0,4	0,6

nuclei of uranium and other elements (oxygen). Zel'dovich and Khariton[71] showed that even in the case of an infinitely extended mass of U_3O_8 an explosion (an infinitely branched chain) is impossible due to the large value of γ—the probability of the fact that a neutron formed with energy E_0 is slowed down without having had time to produce a new fission event prior to reaching the energy E_{cr} which is equal approximately to 1.5 MeV below which it no longer can produce fission.

For the quantity γ Zel'dovich and Khariton using the equations that determine the variation in time of the average energy E of the neutrons and the number of neutrons in scattering

$$\frac{dE}{dt}=-E\bar{v}\sum_i \sigma_{si}c_i\lambda_i,$$

and

$$\frac{dN}{dt}=-N\bar{v}\sum_i \sigma_{ci}c_i,$$

where

$$\lambda_i=\frac{2m_i}{(m_i+1)^2}$$

(m_i is the mass of the ith nucleus expressed in terms of the neutron mass), obtained the expression

$$\gamma=\exp\int_{E_0}^{E_{cr}}\psi(E)d\ln E,$$

where $\psi=\Sigma c_i\sigma_{ci}/\Sigma c_i\sigma_{si}\lambda_i$ for ψ not dependent on the neutron energy $\gamma=(E_{cr}/E_0)^{\psi}$. The possibility of an explosion is determined by the inequality

$$\nu_f(1-\gamma)>1, \tag{4}$$

which expresses the necessity of appearance of more than one neutron to replace the one absorbed or slowed down below E_{cr}. On carrying out calculations of the quantity $\nu_f(1-\gamma)$ for different possible values of ν_f and E_0, Zel'dovich and Khariton obtained for U_3O_8 in an infinite mass the values of $\nu_f(1-\gamma)$ given in Table I.

In these calculations the value of 1.5 MeV (Ref. 13) was taken for E_{cr}, $\sigma_{sO}=2\cdot10^{-24}$ cm^2, $\sigma_{sU}=6\cdot10^{-24}$ cm^2 [2)], $\sigma_{cO}=0$, $\sigma_{cU}=\sigma_f=0.5\cdot10^{-24}$ (Ref. 25).

As can be seen from Table I oxygen greatly impedes the realization of a chain disintegration of uranium. The criterion $\nu_f(1-\gamma)$ becomes greater than unity only for the most favorable combination of ν_f and E_0.

One might think that the use of pure uranium would guarantee the realization of a chain explosion based on fast neutrons, but the presence of inelastic scattering according to the calculations of Zel'dovich and Khariton also in this case (very favorable from the point of view of elastic collisions) leads to a powerful breaking off of the chains and, apparently, to the impossibility of an explosion.

It should also be noted that the presence of energy losses by the neutrons in elastic collisions strongly limits the possibility of utilizing the "neutron insulation" proposed by Perrin since the neutrons returning from the comparitively distant layers of the "insulator" will have energies lower than E_{cr}.

Haenny and Rosenberg[73] tried to solve experimentally the problem of the possibility of realizing a chain reaction in uranium. They placed a source of neutrons inside a 14-kg block of uranium oxide U_3O_8 and measured the increase due to uranium of the number of fast neutrons emitted by the Rn–Be source surrounded by 4 cm of paraffin. In order to record only the fast neutrons a hexane chamber[74] was used as a detector. The authors on surrounding the source by uranium obtained a 20% increase in the number of fast neutrons instead of a 5% decrease which according to their opinion should be associated with the absorption in uranium and assumed that this increase signifys a possibility upon a further increase in the layer of uranium surrounding the source of an infinite "amplification" of the neutron source up to catastrophic values. However these conclusions are not sufficiently well founded. In view of the indefiniteness of the spectrum of the neutrons from the source being used (Rn–Be+4 cm of paraffin) and it not being clear whether the hexane chamber records specifically those neutrons which are capable of inducing further fission, in interpreting the observed 20% amplification one should be particularly careful otherwise one can easily arrive at erroneous conclusions.

Perrin[75] also examined the behavior of a system using uranium (uranium oxide) with the addition of a certain amount of water and cadmium. The presence of hydrogen atoms produces relatively rapid conversion of a portion of fast neutrons into slow and thermal ones. If we now have a system with a mass exceeding the critical mass based on the simultaneous action of fast and thermal neutrons and the entire system begins to heat up due to liberation of heat as a result of fission, then an increase in temperature will lead to a decrease in the capture cross section of uranium for thermal neutrons (the cross section is inversely proportional to the velocity of the neutrons). And capture of thermal neutrons which occurs primarily as a result of the admixture of cadmium remains practically unchanged. As a result the increase in temperature will have as a consequence the transition of the system from an overcritical state into a subcritical one since the critical dimension must grow with an increase in temperature. Consequently a self-regulating "explosion-safe" system is obtained. Perrin[75] presents the following example for U_3O_8 containing 3% of water and 0.01% of cadmium, $R_{cr}=65$ cm at room temperature and 80 cm at 900°.

The calculations of Perrin[75] contain in addition to the lack of taking into account elastic and inelastic slowing down of neutrons yet another significant omission. Perrin

assumes that the conversion of fast neutrons into slow ones under the conditions examined by him takes place with the same yield (approximately 85%) that was obtained by Halban, Kowarski, and Savitch[26] for a 1.6-molar solution of uranyl nitrate in water. In actual fact at low (3%) by weight concentrations of water in uranium the resonance capture of neutrons by uranium will, as is shown below, be very great and will lead to obtaining thermal neutrons from fast ones with a yield of approximately 15%. At the same time such a quantity of water practically liquidates, due to the loss of energy in elastic collisions with protons, fission by fast neutrons. We therefore arrive at the conclusion that the numbers quoted by Perrin are quite unreal since also in pure uranium the occurance of an explosion is not very probable, and the addition of a small amount of hydrogen only worsens the situation.

Adler and Halban[76] at the same time as Perrin arrived at practically the same conclusions as did Perrin.

Chain reaction based on slow neutrons. The second variant of realizing a chain explosion of uranium—an explosive disintegration based on slow neutrons—can be conceived in the following manner. Assume a system consisting of uranium and a dilutant that has the purpose of rapidly slowing down the neutrons from the initial energies down to thermal ones for which the fission cross section again becomes sufficiently great. Rapid slowing down is necessary in order that the neutrons would "jump over the resonance level of uranium of mass 238" with minimum losses. In order to achieve effective slowing down it is necessary to take larger quantities of dilutant (for example water). However, with the large amount of dilutant one will encounter the fact of capture of neutrons by nuclei of the dilutant and this will lead, just as resonance capture, to breaking the chains. Anderson, Fermi, and Szilard[64] pointed out this double role of the dilutant; they note the impossibility at the present time to answer the question whether it is possible in general to realize a chain explosion in a mixture of uranium with water.

A detailed quantitative analysis of chain fission of uranium based on slow neutrons was made by Zel'dovich and Khariton.[77] They formulate the condition for initiation of an explosion, i.e., the condition for coming into existence of infinitely branching chains[78] in the following manner. The discussion, just as in Ref. 71, is given for the case of an infinitely large system. Assume that per unit time in the system there arise N fast neutrons (as a result of fission events brought about by neutrons and also from external neutron sources). Of them φN will "survive" in the course of slowing down through the region of resonance capture and will become slow, thermal neutrons (φ is the probability for a fast neutron to slow down without undergoing resonance capture by a nucleus of uranium of mass 238). Of the φN "surviving" neutrons θ will be captured by uranium nuclei (the remaining $(1-\theta)\varphi N$ neutrons will be captured by nuclei of the moderating admixture, for example hydrogen). If for each slow neutron captured by uranium there are formed ν new fast neutrons then there will be formed an additional $\nu\theta\varphi N$ neutrons. Let N_0 be the number of neutrons arising in the system from external sources. Then by definition

$$N=N_0+\nu\theta\varphi N, \tag{5}$$

or

$$N=\frac{N_0}{1-\nu\theta\varphi}. \tag{6}$$

From this we obtain the critical condition for initiation of an explosion

$$\nu\theta\varphi>1. \tag{7}$$

We note that the number $\theta\varphi N$ contains also slow neutrons being captured by uranium of mass 238 with the formation of uranium of mass 239 and subsequent β-emission (this type of capture according to the Breit–Wigner formula is practically absent[3)] in the range of energies from somewhat lower than the resonance energy to energies close to thermal when the effective cross section for capture begins to grow in proportion to $E^{-1/2}$) and neutrons captured by uranium of mass 235 then undergo fission. Since ν_f introduced by us earlier refers to the event of fission then

$$\nu=\nu_f\frac{\sigma_f}{\sigma_f+\sigma'_{cU}}=\nu_f\frac{\sigma_f}{\sigma_{cU}}, \tag{8}$$

where σ_f is the cross section for the capture of a neutron by uranium with fission, σ'_{cU} is the cross section for the simple (radiative) capture of a neutron by uranium and σ_{cU} is the total cross section for capture.

The quantity θ is expressed in the following way:

$$\theta=\frac{c_U\sigma_{cU}}{c_U\sigma_{cU}+c_H\sigma_{cH}}. \tag{9}$$

The principal problem in the calculation is the determination of the quantity φ depending on the composition of the system. Using for the case of dilution by hydrogen the fact that in each collision with a nucleus of hydrogen the neutron is scattered in energy with uniform density over the entire interval from zero to the energy prior to a collision, and by introducing w the probability of being captured in such a collision Zel'dovich and Khariton obtained the integral equation

$$\varphi(E)=[1-w(E)]\frac{1}{E}\int_0^E\varphi(\varepsilon)\mathrm{d}\varepsilon,$$

the integration of which taking into account the fact that for E which to any significant extent exceeds E_r, φ approaches a certain asymptotic value and leads to the expression

$$\varphi=\exp\left(-\int_0^\infty w\mathrm{d}\ln E\right). \tag{10}$$

The probability of being captured in the first collision when the neutron energy is equal to E is evidently expressed in the following way:

$$w=w(E)=\frac{c_U\sigma_{cU}(E)}{c_U\sigma_{cU}(E)+c_H\sigma_{sH}(E)}. \tag{11}$$

TABLE II.

η*)	1	2	4	8	17	62
θ	0,422	0,855	0,748	0,547	0,410	0,160
φ	0,251	0,377	0,501	0,613	0,716	0,840
$\theta\varphi$	0,231	0,331	0,374	0,366	0,284	0,134

*)The values of η equal to 17 and 62 correspond to the experiments of Anderson, Fermi, and Szilard,[64] and of Halban, Joliot, and Kowarski.[62]

If we had exact data on the energy dependence of σ_{cU} then the calculation of φ could be made by numerical integration of (10). Calculating φ with the aid of the Breit–Wigner formula with the values $\sigma_r=3000\cdot 10^{-24}$ and $\Gamma=0.2$ (these values more or less correspond to the available data on the absorption of slow neutrons in uranium) and assuming $\sigma_{sH}=20\cdot 10^{-24}$ cm^2 Zel'dovich and Khariton obtained for the case of equal atomic concentrations of hydrogen and uranium ($\eta=1$) the value $\varphi=0.84$. However such a calculation is not very reliable in view of the fact that the formula corresponding to a single resonance level apparently is inapplicable to the case under consideration.[65] The direct experiment of Halban, Kowarski, and Savitch[26] yields $\varphi=0.86$ for $\eta=62=c_H/c_U$ and this corresponds to a considerably larger capture under equal conditions.

In connection with the unreliability of using the Breit–Wigner formula Zel'dovich and Khariton used the fact that $\int w d\ln E$ is inversely proportional to the square root of the ratio c_H/c_U earlier denoted by η, i.e.

$$\varphi=\exp\left(-\alpha\sqrt{\frac{c_U}{c_H}}\right)=\exp\left(-\frac{\alpha}{\sqrt{\eta}}\right).$$

The constant quantity α can be determined directly from the experiments of Halban, Kowarski, and Savitch[26] in which c_H and c_U are known and the value of φ is determined. Having determined α one can then calculate the value of φ for any arbitrary composition, with the result not depending on the number and distribution of resonance levels in the uranium nucleus. In this way one obtains $\alpha=1.36$ and then with the aid of elementary calculations one obtains the values of θ and $\theta\varphi$ for different ratios of concentrations of hydrogen and uranium.

These values are given in Table II.

The maximum value of $\theta\varphi$ corresponds to $\eta=c_H/c_U\approx 4$ and is equal to ~ 0.375. Consequently the critical conditions for the initiation of an explosion (7) will be satisfied if ν will be greater than 1/0.375, i.e., greater than 2.75. Analyzing with the aid of the method discussed above the results of Joliot *et al.*,[62] Zel'dovich and Khariton[77] obtain for ν the value of 1.95; consequently the maximum possible value of the criterion $\nu\theta\varphi$ is $0.375\cdot 1.95=0.73$. This means that for no composition of the mixture of uranium with water are infinite branching of the chains and an explosion of uranium possible. The greatest observable effect can be an increase in the intensity of a neutron source due to neutrons obtained from fission.

This increase according to (6) will be

$$\frac{N}{N_0}=\frac{1}{1-\nu\theta\varphi}\approx 4.$$

An effect of just this order of magnitude was observed by Halban, Joliot, Kowarski, and Perrin[79] in investigating the distribution of neutrons in a large mass (300 kg) of uranium mixed with water in different ratios.

It is of interest to note that the analysis of the data of Joliot *et al.*[62] carried out "theoretically," i.e., on the basis of the Breit–Wigner formula with one level and correspondingly with the value $\varphi=0.84$ for $\eta=1$, which gives $\alpha=0.168$ (instead of $\alpha=1.36$ in the case of an "empirical" determination using the data of the paper by Halban, Kowarski, and Savitch[26]) gives for $\nu\theta\varphi$ the value of 0.68, i.e., very close to the already quoted value of 0.73. It is easy to understand the reason for such an insensitivity of the value of the criterion $\nu\theta\varphi$ to the method of calculation, i.e., to the value of φ used for the calculation. The smaller is the value of φ that we use for the calculation, the greater will be the value of ν that we shall obtain (since otherwise the experimentally observed distribution of the neutron density would not be guaranteed). But the product of the two quantities $\nu\varphi$ will change very little. This circumstance to a large extent contributes to the reliability of the obtained value of $\nu\theta\varphi$.

7. PATHS TOWARDS THE REALIZATION OF A CHAIN EXPLOSION OF URANIUM BASED ON THERMAL NEUTRONS

We have arrived at the conclusion that the thermal explosion of a mixture of uranium with water is not possible for any ratio of the amounts of uranium and of water. To achieve an explosion, i.e., to increase the value of $\nu\theta\varphi$ to a value exceeding unity one could attempt to utilize two routes: 1) replacing hydrogen by another dilutant, 2) enrichment of uranium by the light isotope with atomic weight 235. Let us examine each of these routes.

Let us assume that we replace hydrogen which serves for slowing down the neutrons by some other element with atomic weight m, capture cross section σ_c and scattering cross section σ_s. In each collision of a neutron with a nucleus of the "moderator" the energy of the neutron will be changed on the average by $2m/(m+1)^2$ of the value of the energy assuming that the collisions occur as in the case of elastic spheres). In the case of hydrogen this relative change of energy is equal to 0.5, in the case of deuterium it is equal to 0.45, in the case of helium to 0.32, in the case of carbon to 0.142, in the case of nitrogen to 0.125 etc. If in addition we take into account that for hydrogen in the energy interval of interest to us the cross section for scattering is approximately equal to $20\cdot 10^{-24}$ cm^2 and for elements from deuterium to nitrogen it is approximately equal to $2\cdot 10^{-24}$ cm^2, then in the first approximation one will be able to say that deuterium slows down by a factor of 10 "worse" than hydrogen, helium by a factor of 17, carbon by a factor of 35, nitrogen by a factor of 40 etc.

We now note that hydrogen under otherwise equal conditions could provide the condition for an explosion if its capture cross section for thermal neutrons were not $0.27\cdot 10^{-24}$ cm^2, but by a factor of ~ 5 lower (this result is

TABLE III.

Element	D	He	C	N	O
Maximum explosive capture cross section	0,0054	0,0032	0,0015	0,0013	0,0012
Capture cross section	< 0,03	—	< 0,01	1,3	<0,01

obtained if to Table II one adds a number of lines with the values of θ calculated on the assumption of different gradually decreasing values of σ_{cH} and examines for what value of σ_{cH} the condition $\nu\theta\varphi=1$ will first be satisfied. Further, we can assert that in order to guarantee the possibility of an explosion of a mixture of uranium with some kind of moderator the capture cross sections of the latter should be lower than the "explosion cross section" of hydrogen which is equal to $\sim 0.054\cdot 10^{-24}$ cm^2, by approximately the same factor that the given moderator slows down neutrons "worse" than hydrogen. As a result we obtain the following "maximum capture cross sections leading to an explosion," expressed in units of 10^{-24} cm^2 for different elements which could be used as moderators (Table III).

For comparison we have given in the third line of Table III the data of Frisch, Halban, and Koch[80] concerning the capture cross sections of several light elements. It is evident that the data on the capture cross sections need to be made more precise in the future.

We now examine what degree of enrichment n of the uranium isotope with atomic weight 235 is needed in order that the condition $\nu\theta\varphi=1$ will be fulfilled on dilution with hydrogen.

In contrast to the cases that have just been examined where the calculation had to be carried out very approximately for different contents of uranium-235 the calculation can be carried out strictly within the framework of the method presented above. It is only necessary in all the formulas which contain the quantity σ_f to replace it by the quantity $n\sigma_f$.

The calculation carried out in this manner showed that the quantity $\nu\theta\varphi$ attains the value 1 for $n\approx 1.85$. We remind the reader that this number refers to an unlimited volume. In the case of a finite volume a somewhat greater enrichment is necessary, the greater the smaller is the amount of the mixture.

It should be noted that approximately a twofold enrichment of those fairly large amounts of uranium which are necesary for realizing a chain reaction (as will be shown in the next section) represents a very massive problem close to impracticability.

8. THE CRITICAL MASS

In a finite mass of uranium with a moderator it is necessary, as has been pointed out by Perrin[69], to take into account simultaneously the change of neutron density in time associated both with diffusion and also with different nuclear processes (fission and capture). We construct the corresponding equation for the balance of the density of thermal neutrons ρ in a system not provided with an external source of neutrons:

$$\frac{\partial\rho}{\partial t}=D\Delta\rho+f\rho.$$

Here D is the diffusion coefficient for thermal neutrons, while the quantity f can be expressed in the following manner:

$$f=u\left(c_U\sigma_{cU}\nu\varphi-c_U\sigma_{cU}-\sum_i c_i\sigma_{ci}\right),$$

where the first term corresponds to the number of slow neutrons appearing per unit volume as a result of fission events taking into account the circumstance that only the portion φ is slowed down without being captured in the resonance level of uranium-238. The last term corresponds to the capture of neutrons by nuclei of the atoms of the moderator.

The solution of this equation for a spherical mass can be written in the form

$$\rho=\sum_k \psi_k(r)\exp\left[t\left(f-x_k\frac{D}{r^2}\right)\right].$$

It can be easily seen that one can restrict oneself to consideration of the first eigenfunction and the first eigenvalue since the subsequent terms of the sum will have ever decreasing significance as the time t increases. Therefore one can take the solution to be of the form

$$\rho=c_0\psi_0e^{pt},\quad \text{where } p=f-\chi_0\frac{D}{r^2},$$

from which we find the critical dimension of the sphere

$$r_{cr}=\sqrt{\frac{\chi_0 D}{f}}=\pi\sqrt{\frac{D}{f}}=\frac{\pi}{\sqrt{3}}\sqrt{\frac{\lambda_s\lambda_c}{\nu\theta\varphi-1}},$$

where λ_s and λ_c are the mean free paths of neutrons for scattering and for capture respectively.

We consider a concrete example. Assume that the system consists of a mixture of U_3O_8 with heavy water of 99.5% quality with a quantity of U_3O_8 of 17.5 weight % which corresponds to $c_U:c_D=1:100$. The capture cross sections of deuterium and oxygen will be taken equal to zero. We shall assume that the uranium oxide is uniformly distributed in the water. Then taking

$$\sigma_{sU}=6\cdot 10^{-24}\ \text{cm}^2,\quad \sigma_{sD}=2.5\cdot 10^{-24}\ \text{cm}^2,$$

$$\sigma_{sH}=35\cdot 10^{-24}\ \text{cm}^2\ \text{(for slow neutrons)},$$

$$\sigma_{sO}=2.5\cdot 10^{-24}\ \text{cm}^2,\quad \sigma_{cU}=3.2\cdot 10^{-24}\ \text{cm}^2,$$

$$\sigma_{cH}=0.27\cdot 10^{-24}\ \text{cm}^2,\quad \nu=1.95,$$

finding for φ from Table II with the aid of considerations stated on a previous page[4)] the value of 0.7 and calculating using equation (9) $\theta=0.959$, we obtain $\nu\theta\varphi\approx 1.3$ and further

$$R_{cr}\approx\frac{\pi}{\sqrt{3}}\sqrt{\frac{4.6\cdot 450}{0.3}}\approx 150\ \text{cm},$$

or for the critical mass of uranium oxide and water

$$M_{cr}^{U_3O_8} \approx 2.5 \text{ t}, \quad M_{cr}^{D_2O} \approx 15 \text{ t}.$$

Finally, we consider briefly the question of the nature of the course of the process in time in a system in which critical conditions may be attained for the branching of the chains (4) and (7) limiting ourselves to the assumptions and conclusions of the theory. As we have seen (cf., above) under the given conditions above the limit the intensity of the reaction and the neutron density increase exponentially. However, if the probability of the branching of the chain is at all significant the time of increase of the rate of reaction by a factor of e turns out to be of the order of 10^{-7} s (for fast neutrons).[5] During a time of 10^{-4}–10^{-5} s there will be enough energy liberated to evaporate the entire mixture and we have assumed that the reaction was initiated by cosmic neutrons. With such a rapidly developing reaction we can no longer leave aside the discussion of the process itself of creating supercritical conditions, as has been done, for example, by Flügge.[82] It turns out that it is necessary to conduct a detailed analysis of the transition over the limit, i.e., of the course of the reaction near $p=0$, where the behavior of the system depends sensitively on a number of factors which can be neglected far from the limit. Thus, the expenditure of uranium and the appearance of new nuclei capable of capturing neutrons impede the branching of the chains. Calculations show that even stronger (by a factor of $\sim 10^8$) is the effect of thermal expansion brought about by the liberation of energy of the nuclear reaction. The significance of thermal expansion is related to the fact that for a mixture of constant composition it follows from Perrin's formulas that

$$(R\delta)_{cr} = \text{const}$$

(δ is the density of uranium or of uranium-containing substance), or

$$(R^3\delta^3)_{cr} \sim (M\delta^2)_{cr} = \text{const}$$

(M is the mass) so that the critical mass increases with a decrease of the density and in a system of a given mass the decrease of density leads to a deterioration of the condition for branching of the chains.

The very short relaxation time for the development of neutron chains leads to the situation that for any method of surpassing the critical conditions (addition of uranium, bringing together two parts of the system) the nuclear reaction proceeds with such intensity that as a result of the consumption of uranium and the thermal expansion on the average at all times the condition $p=0$ is maintained and the reaction stops practically simultaneously with the cessation of providing more uranium or of bringing together parts of the system.[83] The chain reaction that has begun then stops without affecting the critical mass of uranium. In this respect the system essentially differs from explosives and due to its self-regulation would be on the contrary quite convenient for power production applications, while special devices (for example the addition of cadmium; cf., Refs. 71, 72) are useless.

It is interesting to note that the existence of even a small amount (1%) of delayed neutrons emitted with a half-period of ~ 10 s after fission, significantly delays the development of the reaction near the critical conditions, however the relaxation time even in this case remains sufficiently small in order that the conclusions drawn above would remain valid.

9. PROSPECTS FOR POWER PRODUCTION

The material presented in the preceding sections indicates that at the present moment it is still not possible to make final conclusions concerning the possibility or impossibility of realizing in uranium a nuclear reaction of fission with infinitely branching chains. If such a reaction can be realized then, as has been indicated in the preceding section, regulation of the rate of reaction is automatically realized, guaranteeing its smooth development in spite of the tremendous amount of energy available to the experimenter. This circumstance is exceptionally favorable for the use of the reaction for energy production. We shall therefore provide—even though this does amount to dividing the skin of an as yet unkilled bear—some numbers which characterize the possibility of the use of uranium for power production.

If the process of fission utilizes fast neutrons, and consequently the reaction involves the principal uranium isotope (U^{238}), then the heat content of uranium amounts to $Q_U = 2 \cdot 10^8$ eV$=4.6 \cdot 10^9$ kg cal mole$^{-1} = 1.8 \cdot 10^{13}$ kg cal/t; the corresponding number for coal is $Q_C = 8 \cdot 10^6$ kg $\cdot$ cal/t. With the ratio $Q_U/Q_C \approx 2 \cdot 10^6$ and the ratio[6] of the prices of uranium and coal on world markets of ~ 500 the cost of a calorie from the principal uranium isotope turns out to be approximately by a factor of 4000 cheaper than from coal (if, of course, the processes of "burning" and heat extraction will not turn out to be in the case of uranium considerably more expensive than in the case of coal). In the case of slow neutrons the cost of a "uranium" calorie (if one uses the above numbers) will be, taking into account the fact that the occurence of the U^{235} isotope is equal to 0.007, only by a factor of 30 cheaper than a "coal" calorie with all other conditions being equal.

The world production of uranium ores and compounds amounted in 1934 to approximately[84] 700 t (not including USA). In converting this to uranium this gives ~ 300 t, which is equivalent to $6 \cdot 10^8$ t of coal, i.e., more than half of the world annual production of coal if the fast neutron process can be used, and only $5 \cdot 10^6$ t if slow neutrons are used. A relatively widespread occurence[85] of uranium (some authors consider that the occurence of uranium is the same as the occurence of copper), probably will give the possibility in case of necessity to increase the production considerably.

We should expect that the question of the possibility or impossibility of realizing the chain "burning" of uranium using slow, or fast neutrons will be decided in the near future. Until a positive solution of this problem is available a more detailed analysis of the prospects of energy production associated with uranium is hardly expedient.

Supplement. At the May session of the Academy of Sciences of the USSR K. A. Petrzhak and G. N. Flerov reported that they have observed the appearance of fragments of uranium in the absence of a neutron source. The number of fission events observed by them was very small. Petrzhak and Flerov point out that if the fragments are associated with spontaneous decay of uranium then the corresponding half life is equal to 10^{16} years, if it is U^{238} that is decaying, or 10^{14} and 10^{12} years in the case of U^{235} and U^{234} respectively.

No analogous effect has been observed for thorium.

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1)It is assumed that all the capture cross sections vary in inverse proportion to the neutron velocity.

2)According to the data of the most recent investigations[27,72] one should adopt a somewhat larger value for σ_{sU}, but this will not significantly affect the result of the calculation.

3)The Breit–Wigner formula for the cross section for capture by a nucleus of a neutron having an energy E in case of a single resonance level has the following form:

$$\sigma_E=\sigma_r\sqrt{\frac{E_r}{E}}\frac{(\Gamma/2)^2}{(E-E_r)^2+(\Gamma/2)^2},$$

here σ_r as can be easily seen is the value of σ_E for $E=E_r$, i.e., at resonance, Γ is the width of the resonance level. The formula gives a maximum at $E=E_r$ and then as the neutron energy decreases the value of σ_E falls off rapidly (the more rapidly the smaller is Γ), remaining quite small until E decreases to such a value that $(E_r/E)^{1/2}$ becomes sufficiently large.

4)Oxygen and deuterium were assumed to be equivalent to an additional amount of hydrogen and an effective value of η was found. Then from Table II a value of φ corresponding to the obtained value of η was found by interpolation.

5)Adler[81] discusses in detail the kinetics of the process within the indicated time interval in the special case of a certain number of neutrons concentrated at the initial moment in the center of the sphere. These calculations relating to the process occurring during a time shorter than the diffusion time have no relation to the occurence of the real process discussed by us below.

6)The ratio of prices is taken for uranium oxide. Metallic uranium at present is considerably more expensive, and it would be just what is needed for using the principal isotope.

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