鈾核之自裂*

盧鶴紱　曹萱齡

(浙江大學物理學系)

一、引言

1940 年, Flerov 及 Petrzhak[註 1] 利用塗有鈾元素之多片電離室發現鈾核能自行分裂, 並估得其"平均鈾元素"自裂之半壽期爲在 1×10^{16} 年至 1×10^{17} 年之間. 其後即尚未見有直接確量此常數工作之發表.

1942 年, Scharff-Goldhaber 及 Klaiber[註 2] 利用充氫之電離室測量鈾核自發之中子數. 由假定每次鈾核自裂平均放出一個中子, 算得鈾核自裂之半壽期爲 3×10^{15} 年.

Maurer 及 Pose[註 3] 在海平面上, 其後 Pose[註 4] 在深礦中, 亦皆曾察見此來自鈾核之天然中子發射. 彼等用塗硼之正比計數器測得鈾核天然放射中子之半壽期分別爲 2.5×10^{15} 年及 3.1×10^{15} 年.

上述各實驗結果勢難全部同時接受, 否則鈾核自裂時每次即須放出多如 4 個至 40 個中子.

理論上, 自裂與 α 放射同屬量子力學中之隧道效應. U^{235} 及 U^{238} 均得期其自裂. 惟因 U^{235} 之自裂勢壘較低[註 5] 吾人可假定絕大部分之自裂係由於 U^{235}.

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* 1951 年 12 月 26 日收到.

[註 1] G. N. Flerov 及 K. A. Petrzhak, *Phys. Rev.* **58** (1940), 89.

[註 2] G. Schaff-Goldaber 及 S. L. Klaiber, *Phys. Rev.* **70** (1946), 229.

[註 3] W. Maurer 及 H. Pose, *Zeits. f. Phys.* **121** (1943), 285.

[註 4] H. Pose, *Zeits. f. Phys.* **121** (1943), 293.

[註 5] 據 Frankel 及 Metropolis 之計算, U^{235} 及 U^{238} 之分裂閾能分別約爲 6 及 7 Mev (見參考 6 中的第三圖).

本此假設則 U^{235} 自裂之半壽期應爲 10^{14} 年. Frankel 及 Metropolis[註 6] 計算之結果得 U^{238} 自裂之平均壽期爲 10^{26} 年. 關於 U^{235} 自裂之平均壽期亦有類似但較粗率的估計, 其結果爲 10^{15} 年.[註 7] 誠如 Turner 等[註 7 及 8] 所指出, 在此種計算中, Gamow 透過幾率之指數其積分之極限難於確定, 且對稱分裂之假設與實情不全符合能影響分裂所需之閾能及 Gamow 因子中有效質量之數值. 此均使計算之結果頗堪懷疑. 事實上, Frankel 及 Metropolis[註 6] 已發見若核之庫侖能與表面張力能之比略有差異, 即足使此項計算之結果有顯著之改變. 故欲對此類理論之計算加以評議尚需較確的實驗結果.

在此試驗中我們企圖由觀察鈾核自裂生成物所給的電離脈搏直接測定鈾核自裂之半壽期. 因爲我們尚未能企圖將鈾之同位素分開, 只得測量平均鈾之半壽期.

二、關於儀器

我們用一平行板電離室測量裂成物所產生之電離. 室中充有一大氣壓之氫, 故得利用電子之迅速收集. 室之構造如第一圖所示. 由 B 電池所供給之 270 伏特電壓加於兩電極間. 高電壓極載一直徑爲 6.2 厘米之圓形鋁片, 片上澱貼有氧化鈾層. 欲使氧化鈾很均勻的黏貼片上而不脫落是相當困難的. 我們經數次試驗得一比較能均勻澱貼的製法如下：將稱好分量之硝酸鈾結晶溶於少量酒精中, 再與少許無色瀉克之稀溶液相混. 將此混合後之溶液滴少量於鋁片上, 用電爐加熱至約 400°C, 經五分鐘, 取下使冷却. 然後再滴, 再加熱. 如此做下去, 直至所需之分量已全部澱積在片上爲止. 電離室兩電極間之距離約爲 1.2 厘米, 此項深度已足使最大動能之裂成物在未達收集電極之前即被有效停止於氫中.

如 1 圖中所示, 以一 6AK5 小眞空管製成之一單級前置放大器係裝於一附

[註 6] S. Frankel 及 N. Metropolis, *Phys. Rev.* **72** (1947), 914.

[註 7] W. E. Stephens 等, "Nuclear Fission and Atomic Energy", The Science Press (1948), 頁 109—110.

[註 8] L. A. Turner, *Rev. Mod. Phys.* **17** (1945), 292.

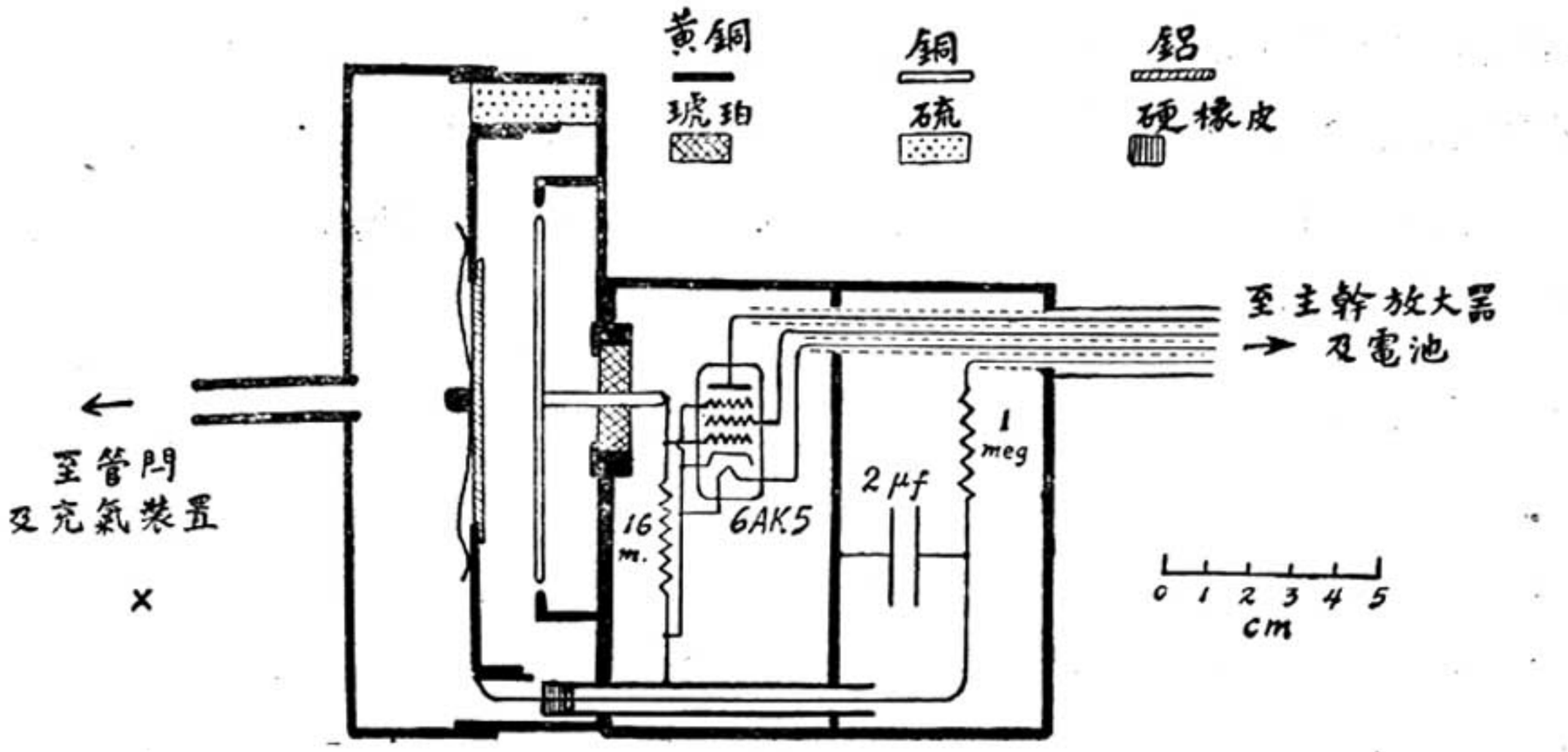

圖 1.

於電離室後壁之黃銅匣中. 主幹放大器之式樣則殊平常,[註 9] 共有四級, 以 6SJ7 眞空管裝成, 每隔一級聯有負反饋. 此放大器系之通頻帶爲由 10^{-4} 秒至 10^{-9} 秒, 係由前置及主幹兩放大器間之配合常數所決定.

電離脈搏經放大後通入一 Cenco 電子示波器, 得觀察之. 由示波器上考之, 知此項脈搏之躍昇時間約爲數微秒, 其分辨時間約爲 20 微秒. 在此實驗中已不需將此時間再縮短, 故未剪截, 得保存示波器上慢掃時脈搏柱之亮度, 以利觀察.

以與 Kanner 及 Barschall[註 10] 所用者相彷彿之人工脈搏發生器校準此放大器系, 知其直線性可準至 230 伏特.

三、程序及方法

在此試驗中共用四個同大小的澱鈾鋁片, 其所含鈾之分量分別爲 1.8, 3.4, 6.7, 及 14 毫克 U/厘米^2. 除含鈾最少之一片外, 其餘三片都已觀察[註 11] 其天然分裂, 每天連續觀察 9 小時, 每片觀察 10 天或 10 天以上. 在每片觀察自裂開始前與結束後, 將一個 3 毫居里之 Ra — Be 中子源[註 12] 放在電離室前(圖 1 中有 × 之處).

[註 9] 係由一原爲游子收集而設計之放大器, 經提高其通頻帶之高頻限, 以適於電子收集, 改換而成.

[註 10] M. H. Kanner 及 H. H. Barschall, *Phys. Rev.* **57** (1940), 373.

[註 11] 大部分時間有兩人同時觀察, 其中至少有一位是本文之作者.

[註 12] 爲王淦昌先生於十數年前所製.

電離室及中子源之週圍即築以一定幾何形式之石臘塊堆. 然後觀察由中子所致之感生分裂所給之電離脈搏, 每次連續觀察數小時. 含鈾最少之一片, 未便觀察其自裂, 只觀察其中子感生分裂. 將所得各片感生分裂之發生頻率外推至鈾層厚度爲零時之值, 即算得每克鈾元素每小時所給感生分裂之次數. 再將三片觀察所給感生零分裂與自裂兩發生頻率之比值作一加權平均比值. 由此二數相比即得每克鈾元素每小時自裂之次數.

示波器上之背景爲很多 α 質粒所致大小不一之脈搏, 分裂所給之脈搏突出其間. 爲避免與 α 粒脈搏相混, 特選定一適於記錄之最低脈高, 其高度約爲 α 脈搏中之最高者之二倍, 超過此高度之脈搏皆被記錄 由觀察所得最高 α 粒脈搏發生之頻率及脈搏之分辨時間算知兩個最高 α 粒脈搏開始相連趨向疊合之機會爲每隔 60 小時不足一次.

將實驗觀得之結果外推至鈾層爲零時之方法如下：

令 t = 鋁片上鈾層之厚度,

R_u = 裂成物在氧化鈾內之平均射程,

R_a = 裂成物在氫內之平均射程,

s_a = 恰足被記錄時之氫內射程 (此氫內所行之最短路程係由選定的最低記錄脈高所決定),

$s_u = R_u(1 - s_a/R_a)$ = 在氫內路程爲 s_a 時在鈾層內所穿過的路程.

在我們的試驗中, $s_a \approx R_a/5$. 因鈾層的面積很大, 邊際效應可以不計. 設有一鈾核在離鈾層表面下 d 處分裂. 則當 $t \leqq s_u$ 時, 此分裂不能被觀察的機會爲 d/s_u, 故整個鈾層內所生之分裂不能被觀察的機會應爲 $f = \frac{1}{2} t/s_u$; 當 $t \geqq s_u$ 時, 在 $d > s_u$ 處之分裂皆不能被觀察, 故其總共不能被觀察的機會應爲 $f = 1 - \frac{1}{2} s_u/t$.

令 F 爲能被觀察之機會, 即 $F = 1 - f$, 則

若

$$t \leqq s_u, \qquad F = 1 - \frac{t}{2 s_u}; \tag{1}$$

若　　$t \geqq s_u$,　　$F = \frac{s_u}{2t}$.　　(2)

四、數據及結果

含鈾 3.4 毫克/厘米^{2}一片，觀察了 10 天，總計 90 小時，共記得 30 次自裂；含鈾 6.7 毫克/厘米^{2}一片，觀察了 10 天，總計 88 小時，共記得 37 次自裂；含鈾 14 毫克/厘米^{2}一片，觀察了 14 天，總計 126 小時，共記得 56 次自裂．每次實驗中，由 α 粒所生之最大脈搏高度之不變，知放大器的情況均稱穩定．在實驗過程中隨時特別當心防止由震動其他意外事故而使放大器發生高如自裂所給者之脈搏，以免影響正確的記錄．

最後用一不澱鈾之空鋁片作試驗．兩次連續觀察各 10 小時，不曾發見有高如自裂所生之脈搏．另一次之 10 小時觀察中，曾發生過一次短期騷擾，其間有兩個脈搏高達分裂所致者，但其形狀則顯然不同，原因不明．從這三次試驗結果，我們就可假定分裂數據中之背景記數爲零．

觀察感生分裂所得數據如下：

鈾層（毫克 U/厘米^2）：	1.8	3.4	6.7	14
每小時感裂次數：	17.8 ± 1.1	26.8 ± 1.5	35.3 ± 1.8	37.8 ± 1.9
感裂與自裂頻率之比：	—	81	84	85

將這些數據化爲每克鈾每小時分裂次數，然後根據 (1) (2) 二式作一曲線使與之最能相合，遂得如圖 2 所示者．由之得感生分裂之頻率爲每克鈾每小時有 400 次，並得 $s_u = 6.0$ 毫克 U/厘米^2，即裂成物在氧化鈾層內之平均射程爲

$$R_u \approx \frac{5}{4} s_u = 7.5 \text{ 毫克 U/厘米}^2.$$

感裂與自裂次數之比之加權平均數爲 83 ± 4．由之得自裂之頻率爲每克鈾每小時有 4.8 ± 0.7 次，即平均鈾自裂之半壽期爲

$$(4.2 \pm 0.6) \times 10^{16} \text{ 年}.$$

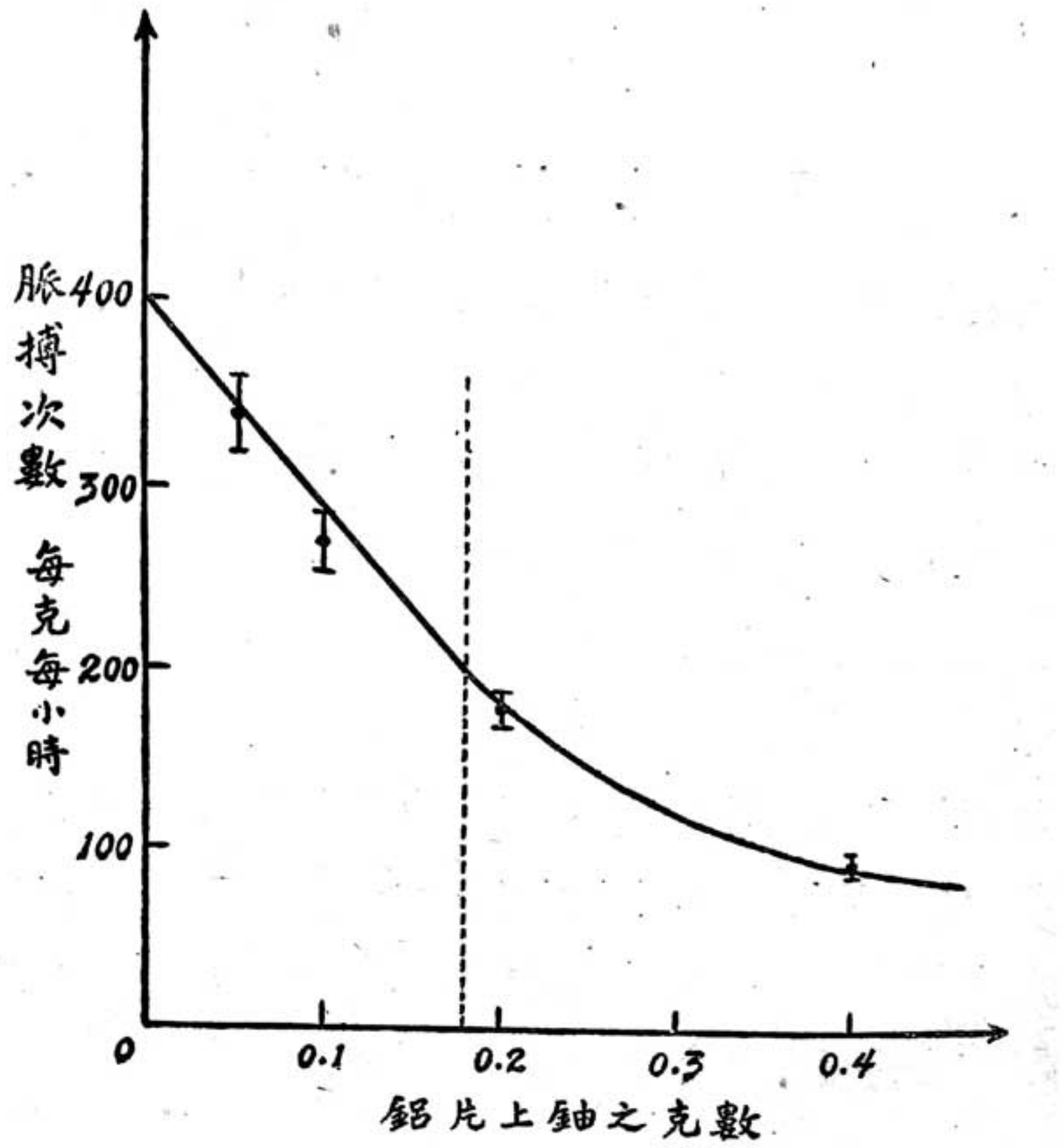
脈搏次數 每克每小時
400
300
200
100
0
0.1
0.2
0.3
0.4
鋁片上鈾之克數

圖 2.

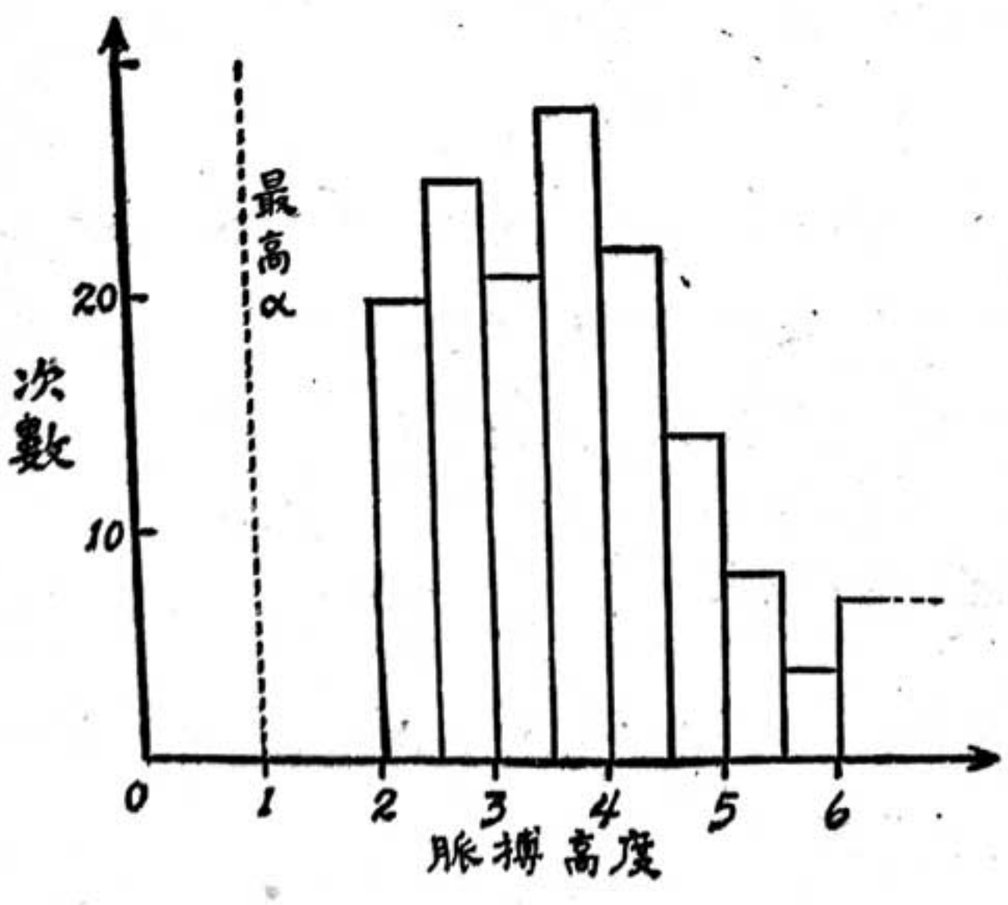
次數
最高α
20
10
0
1
2
3
4
5
6
脈搏高度

圖 4.

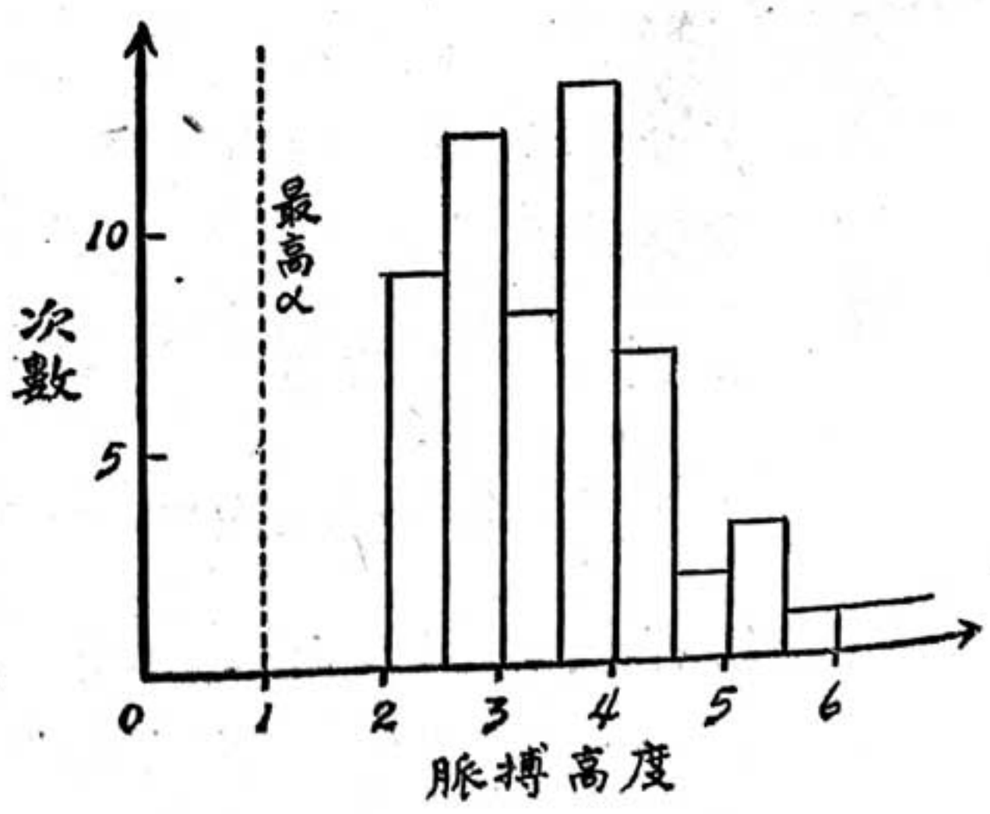
次數
最高α
10
5
0
1
2
3
4
5
6
脈搏高度

圖 4.

吾人若認此自裂絕大部分係由於 U^{235}，則 U^{235} 自裂之半壽期約爲 3×10^{14} 年.

圖 3 與圖 4 分別表出從含鈾最多之一片所觀得感裂及自裂脈搏高度之分佈情形. 由於兩者之分佈情形相似及脈搏發生之無規律，我們更相信確是已見到鈾核的天然分裂.

五、討　論

實驗的結果出乎我們的意料，竟落在 Flerov 與 Petrzhak 所得的數值範圍內. 若接受此項結果，則與 Scharff-Goldhaber, Klaiber, 及 Maurer, Pose 等之實驗結果比較，知每一個鈾核自裂時平均放出多至十個中子，否則就是他們實驗中所測得的中子中有大多數是由於其他來源，譬如可能存在於鈾樣品中或儀器中有如 (α,n), $(n,2n)$, (γ,n) 等核變[註 13] 均能因鈾之存在而產生中子. 此二結論，孰是孰非，尚不能斷定. 然則直接測定與間接測定尚未能符合，仍是一頗值研究的問題.

Segre 及 Wiegand[註 14] 曾由計算得裂成物在 U_3O_8 中之最大射程爲 10 毫克/厘米^2，亦即 9.2 毫克 U/厘米^2. 根據此值，由他們論文中第四圖的吸收曲線的開頭直線部分外推，遂估得裂成物在 U_3O_8 中的有效射程爲 7.1 毫克 U/厘米^2，其平均射程當比此值稍大 適與我們所得的 7.5 毫克 U/厘米^2 堪稱符合.

在我們的計算中只計及統計誤差. 其他被忽略而可能發生誤差，其來源有：(1) 鈾層在鋁片上的澱積難於確實均勻；(2) 製鈾層時因曾用濕克，可能有多少不一的碳元素存留在各鈾層中；(3) 最低記錄脈高之選擇頗嫌隨意；(4) 可能有亂眞計數之存在. 這些誤源所致誤差的大小無法估計.

欲求更精確的測定，首先必須減少統計誤差. 若用金屬鈾作極面，較快的放大器[註 15]，或更加用抵消式電離室[註 16]，則可用很大的鈾面，以圖增加分裂的次數. 再用脈搏延長器及自動脈高筆記器[註 17]，則可使觀察時間增長. 但我們又想到另

[註 13] 因有限次數之鏈裂而多出之中子不必考慮，因爲在他們的試驗中所用的普通鈾的數量不過數千克，而高速中子致鈾分裂的平均路程長約兩米.

[註 14] E. Segre 及 C. Wiegand, *Phys. Rev.* **70** (1946), 810.

[註 15] W. C. Elmore 及 M. Sands, "Electronics, Experimental Technique", (1949).

[註 16] H. W. Koch, J. McElhinney, 及 E. L. Gasteiger, *Phys. Rev.* **77** (1950), 330.

[註 17] J. T. Dewan 及 K. W. Allen, *Rev. Sci. Instr.* **21** (1950), 823; 或參考 15.

一較易又可能較確的辦法，即將鈾化合物溶於能發熒光的溶液中，以閃爍計數器量度自裂的次數，我們現在正在試行這一方法.

最後我們要向幫助我們觀得一部分記錄的賴祖武、平家慶、汪家詠、桑巧娟四位同志致謝.

SPONTANEOUS FISSION OF URANIUM*

By Lu Ho-fu (Hoff Lu) and Tsao Hsuan-ling

Physics Department, Chekiang University

ABSTRACT

An attempt has been made to determine the half-life for spontaneous fission for the "average uranium atom" with an argon-filled parallel-plate ionization chamber, employing the method of electron collection. The high voltage electrode contained an aluminum disk of 6.2 cm diameter on which was deposited a layer of uranium oxide prepared in the following manner. A known amount of uranium nitrate crystals was dissolved in alcohol and mixed with properly thinned colorless lacquer. The procedure then consisted in applying a small amount of the solution on the disk, baking it on an electric furnace to about 400°C for five minutes, and repeating the process again and again until the desired amount of uranium had been deposited. The ionization pulses were observed on a Cenco cathode ray oscilloscope to which the output of the amplifier was connected.

Uranium deposits of 1.8, 3.4, 6.7, and 14 mg U/cm^2 on identical disks were prepared. The spontaneous fission pulses from each of the last three thicker deposits were observed consecutively for ten days or more, the period of observation being nine continuous hours each day. Before and after this observation with each deposit, a three millicurie Ra-Be neutron source was placed in front of the ionization chamber at the position marked in Fig. 1 with a cross, the chamber system and the neutron

* Received December 26, 1951

source being then surrounded by paraffin blocks of reproducible geometry, and the pulses due to fissions thus induced were observed for several hours. With the thinnest deposit only neutron induced fissions were observed. The induced fission results were extrapolated to zero thickness to give the actual rate of induced fissions per gram of uranium. A weighted average of the three observed values for the ratio of the rate of neutron induced to that of spontaneous fissions were used to calculate the actual rate of spontaneous fissions per gram of uranium.

Since the fission pulses occurred in a background of numerous α-particle pulses of varying sizes, only pulses of heights greater than twice those of the largest α-pulses were noted and their heights recorded. As calculated from the observed frequency of occurrence of the largest α-pulses and the resolving time of the pulses, the chance that two largest α-pulses would start to pile up was not more than one in 60 hours.

The method of extrapolating the results to zero thickness was as follows: Let t be the thickness of deposit, R_u the mean range of fission fragments in uranium oxide, R_a that in argon, s_a the minimum detected path-length in argon corresponding to a chosen bias of pulse heights, and $s_u = R_u(1 - s_a/R_a)$ the remaining length of this path that was in the deposit. In our case $s_a \approx R_a/5$. Then, the fraction of the number of fissions occurring at depth d that was not observed was d/s_u if $t \leqq s_u$. Hence, for $t \leqq s_u$, the total fraction of fissions missed was $f = \frac{1}{2} t/s_u$. In $t \geqq s_u$, fissions occurring at depths $d \geqq s_u$ were entirely missed, so that the total fraction of fissions missed was $f = 1 - \frac{1}{2} s_u/t$. Or, the fraction of the number of fissions that was observed was $F = 1 - f$:

$$F = 1 - t/2s_u \quad \text{for} \quad t \leqq s_u; \tag{1}$$

$$F = s_u/2t \quad \text{for} \quad t \geqq s_u. \tag{2}$$

In ten separate runs, totalling 90 hours, 30 spontaneous fission pulses were observed with the 3.4 mg/cm² deposit. In ten separate runs, totalling 88 hours, 37 spontaneous fission pulses were observed with the 6.7 mg/cm² deposit. In 14 separate runs, totalling 126 hours, 56 spontaneous fission pulses were observed with the 14 mg/cm² deposit. Throughout this observation, the gain of the amplifier remained practically constant as indicated by the constancy of the heights of the largest α pulses-

In two separate runs, each of 10 hour duration, in which a blank electrode replaced the uranium-carrying one, not a single pulse of fission size was observed. In another similar 10 hour run, it so happened that a disturbance of unknown origin and short duration occurred among which two pulses of fission sizes but of appearance different from fission pulsis were observed. It was, therefore, safe to assume zero background for the fission counts.

The rates at whieh induced fission pulses were observed were 17.8, 26.8, 35.3, and 37.8 per hour. These values couverted into number of pulses per hour per gram of uranium and were plotted in Fig. 2. The best fit with equations (1) and (2) was obtained with $s_u = 6.0$ mg U/cm², corresponding to a mean range of fission fragments in the deposit material of approximately

$$R_u \approx \frac{5}{4} s_u = 7.5 \text{ mg U/cm}^2,$$

and an actual induced fission rate of 400 per hour per gram of uranium.

The ratios of the observed rate of induced fission to that of spontaneous fission were 81, 84, and 85. Using a weighted average of 83 ± 4 for this ratio, the actual rate of spontaneous fission of uranium was found to be 4.8 ± 0.7 per gram per hour. This corresponds to a half-life for the average uranium atom of

$$(4.2 \pm 0.6) \times 10^{16} \text{ years.}$$

If we attribute spontaneous fission principally to U^{235}, the half-life for U^{235} would be 3×10^{14} years.

In Fig. 3 and Fig. 4 are depicted the distribution of heights of neutron induced and spontaneous fission pulses observed with the thickest deposit. Their apparent similarity together with the randomness of occurrence of the pulses reassured us that we were observing natural fission pulses.

To our surprise rather than expectation, our result falls in the range of values given by Flerov and Petrzhak. This would indicate either that upon each spontaneous fission as many as about ten neutrons were set free or that most of the neutrons observed by Scharff-Goldhaber and Klaiber, and by Maurer and Pose were of secondary origin, being formed in some processes due to the presence of the uranium sample, e.g., (α, n), $(n, 2n)$, (γ, n) reactions or the like.

Only statistical errors were considered in our calculations. Other sources of errors wers: (1) difficulty in obtaining completely uniform layers of uranium deposits, (2) possibility of different carbon contents in the deposits, resulting from the use of lacquer adhesive, (3) the rather arbitrary choice of pulse height bias, and (4) possible presence of unrecognized spurious pulses.